

Modern advances in mechanics of materials with hydrogen

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Abstract

Accumulation of hydrogen inside metals leads to hydrogen embrittlement. Examination of the hydrogen influence on the mechanical properties of materials, as well as explanation of the mechanisms of its transport, especially diffusion, is one of the most actual problems in mechanics of materials. Within the framework of this paper we review existing models of hydrogen diffusion and describe the phenomenon of localization of plastic deformations. According to the retrospective of the models development, the most well-known and universally recognized mathematical description of hydrogen transport is based on the elementary diffusion equation and a number of equations for the chemical reactions of hydrogen trap filling. Moreover, the main experimental method for studying the hydrogen motion in a solid generally does not take into account diffusion. Another important fact is that most of the existing results have been obtained by studying specimens charged with hydrogen. According to our analysis this approach can not be used for investigation of the hydrogen influence on the mechanical properties of materials. In this regard we developed the model of multichannel diffusion of hydrogen. On the basis of this model, we also developed an experimental-calculation method for measuring the binding energies of hydrogen. In this study we also observe modern approaches for modeling of localization of plastic deformation, considering it as a process of loss of stability of motion. We underline that all problems of localization of plastic deformation intersect with the problems of formation of local areas of hydrogen embrittlement. The dynamic instability caused by hydrogen is associated with its diffusion. One can conclude that it is necessary to consider dynamic unstable processes when solving problems of mechanics of materials. Finally, it is necessary to take into account the spatial inhomogeneity of both plastic deformations and the distribution of hydrogen when calculating the strength.

1 Introduction

Modern problems in mechanics are closely related to the special properties of materials due to technology development. Exploitation of most of the metal structural

components is realized with minimal stock factor and under stress levels equal or even exceeding the yield stress. The mass fraction of special high-strength steels in the modern car body is 70 %. Highly alloyed high-strength steels are the only ones that are used in construction of pipelines. Forged titanium is widely used in aviation. Details of modern building constructions are connected by high-strength hardware instead of rivets. Nowadays superhigh-strength steels and nanostructured materials with tensile strength three to four times higher than that of common ones are developed and used.

Elastic modules are fundamental values underlines the binding energy in a metal matrix. Metals with extreme properties are generally obtained by changing the internal structure. It means that the yield line of the original matrix is used to increase strength. In this case, the maximum plastic deformations substantially decrease down to zero in some nanocrystalline metals.

Under these conditions, the influence of relatively small imperfections and local defects becomes determinative. One of these factors is dissolved hydrogen. Unlike other components of alloys, it has a large diffuse mobility and can enter the metal from the external medium, both directly in the gaseous state, and due to various corrosive processes. Therefore, one can say that in nature there are no metals completely hydrogen free.

2 Mechanics of materials containing hydrogen

The significant effect of dissolved hydrogen on the metal properties was first discovered by M.Fremy in 1861 in his investigation of the effect of gases on the steels properties. He studied the meteorite iron specimens and found that steel can be obtained from these specimens only if hydrogen is removed. Since then, any new technology for the metals production and production of many other materials is faced with the problem of the destructive effect of hydrogen at an increasingly low level of its concentrations in the solid material.

At the beginning of the 20th century, one had to contend with the Krupповaya disease, or in other words, discontinuities of rolled metal due to the mass production of rolled steel. The relative mass concentrations of hydrogen that cause these deviations are of the order of $4 \cdot 10^{-6}$ in steels. Later, the problem of the brittleness of aluminum alloys was appeared. The corresponding level of hydrogen concentrations was of the order of $4 \cdot 10^{-7}$. "Hydrogen problems" appeared in the production of titanium, zirconium, heat-resistant nickel alloys.

There is a vast amount of reference devoted to the examination and investigation of "hydrogen problems". The number of scientific publications related to the investigation of hydrogen influence on the materials structure and properties represents between 20 and 30 thousand over the last 150 years.

Nowadays elimination of the negative effect of hydrogen on strength, plasticity, cold-brittle strength, fatigue strength, impact strength and other physical and mechanical properties of materials can be considered as one of the main research direction for mechanics.

The accumulation of hydrogen inside metals always leads to hydrogen embrittlement. Hydrogen transport occurs mainly due to its diffusion in a solid. Therefore, both

explanation of the hydrogen influence on the mechanical properties of materials, and explanation of the mechanisms of its transport are important in terms of mechanics. Initially, the process of hydrogen transport in a solid was considered as a diffusion process described by the Fick equation with a diffusion coefficient depending on temperature according to the Arrhenius law. The parameter of this law is the diffusion activation energy. Moreover, in 1930s Gorsky established that the deformation of the material matrix changes its concentration gradient and thus can lead to diffusion induced by mechanical stresses.

Experimental studies have shown that the application of the Fick equation for the data approximation leads to a huge scatter in the values of the diffusion coefficients and the activation energy for the same materials. Danken and Smith [7] explained this variation by the fact that there is a threshold value of the concentration depending on the method and temperature regime of the sample treatment when hydrogen saturation of metals. Therefore, they introduced the concept of bound hydrogen and distributed in the material hydrogen traps in the explanation of hydrogen transport in a solid. Traps were understood as boundaries of multicrystalline grains, foreign inclusions, internal defects (dislocations, microcrack vacancies, etc.) [22, 36, 37]. As the “traps theory” for description of the hydrogen transport developed, more and more complex mathematical models were used, from McNabb and Foster [28] to the Oriani model [30]. At the same time, the equation of the hydrogen diffusion did not change, only additional equations for filling and emptying the distributed hydrogen traps were introduced. In case of presence of a large number of parameters, such as concentration, diffusion activation energy, the capacity of traps, different activation energies for the collection and desorption of hydrogen from traps, they can be selected to approximate almost any experimental result. Generalization of all models and a comparison of the experimental results with the results of mathematical modeling are carried out in [16, 19]. In [19], in addition to the values $\ln K_B / K$ of activation energies and diffusion constants, different activation energies of hydrogen capture and release from traps were introduced.

The Oriani model is supported by an experimental technique for measuring the binding energies of hydrogen, which is called the “thermal desorption spectra (TDS) method”. The justification of this method was given 60 years ago by Kissinger [20]. According to this paper, the energy state changing and the hydrogen diffusion process in a solid are described as first-order chemical reactions. Therefore, diffusion is considered as a fast process in relation to the process of hydrogen releasing from the traps. In this regard, the experimental procedure for interpreting the obtained experimental data neglects the diffusion equations in the transport model. Hydrogen diffusion was taken into account in one paper [32], but in the framework of the Oriani model [30].

Thus, the most well-known and universally recognized mathematical description of hydrogen transport is based on the most elementary diffusion equation and the set of equations of chemical reactions of trap filling. The main experimental method for examination the hydrogen movement in a solid generally does not take into account diffusion. It leads to the fact that in the fundamental physical handbook [14] it is written that the values of the hydrogen diffusion coefficient and its activation energy, which are given in the tables of the handbook, are the result of averaging of

numerous experimental data and are fair “at best, only for magnitude order”.

This situation can be explained by the great importance of the hydrogen problem. According to the long history of the technology development, all scientific research in this area has been repeatedly checked up experimentally. Therefore, despite the great importance of specific data, significant technological failures has been avoided due to large-scale tests.

Several basic approaches for the modeling of the influence of hydrogen on the strength of materials can be identified, such as taking into account the influence of hydrogen on the nucleation and motion of dislocations, taking into account the influence of hydrogen on the development of cracks, taking into account the internal pressure of hydrogen in the metal and, finally, “physical approaches”, based on taking into account the potential energy of hydrogen-material matrix interaction.

The motion and formation of dislocations and their effect on local plasticity near the peaks of cracks lead to local plasticity because of the very high concentration of dislocations. The mechanism of hydrogenenhanced local plasticity (HELP) was first described by Birnbaum H.K., Sofronis P. [6]. The constitutive equations for the material taking into account local changes in material properties at the mouth of the microcrack were proposed later in [40] and [9, 10]. These equations were obtained on the basis of physical considerations on the interaction potentials between hydrogen and dislocations.

At the same time, according to calculations performed by the authors of the model presented in [40], significant changes in mechanical properties in HELP occur at local relative mass concentrations of hydrogen of the order of 10^{-2} . For most metals it is an unattainable high concentration. Steels, as well as more aluminum alloys, crack themselves up to complete destruction without any external load even at much lower concentrations.

Calculation of local plasticity in the framework of the theoretical examination of a crack with a spherical tip shows that the local hydrogen concentrations at the tip of the crack are only 100 times higher than the average ones [42]. Given that the average values are usually about 10^{-6} , the local concentrations do not exceed 10^{-4} . Thus, the verification calculation does not confirm that local accumulation of hydrogen necessary for triggering physical mechanisms of local plasticity is possible under the influence of external mechanical loads.

There are still many uncertainties surrounding the model noticed by the authors. In particular, there is a nonlinear dependence of the internal potential on the stresses magnitude and hydrogen concentration. Since huge local concentrations many times greater than observed in practice are considered, all nonlinearities are of significant importance.

It was noted in [17] that the HELP model requires enormous computational resources in solving any applied problem, therefore, the only way out is to use the continuum model of dislocation development, however, this replacement is often inadequate and the authors propose to use the growth criterion for submicrocrack, ie, reduce all hydrogen problems to Modeling the development of a crack with a manually adjustable reduction in fracture toughness.

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The hydrogenenhanced decohesion model (HEDE) is similar to HELP [43]. The difference is that HEDE takes into account the decrease in the formation energies of free fracture surfaces with increasing local hydrogen concentration.

It should be noted that the HELP and HEDE models have become generally recognized in state-of-art science. Explicit discrepancies, including mentioned above discrepancies with experimental data, are ignored. The latest scientific discussions are reduced to using these models simultaneously to describe the same material [11, 12]. This is very difficult to realize because of the computational complexity. Therefore, only quasistatic problems considering uniaxial stretching of cylindrical samples are solved.

Standard modeling of the hydrogen-induced cracks development, taking into account the reduction in crack resistance, is also a common approach. At the same time, the model does not relate to real physical mechanisms of hydrogen influence. In addition, it turns out that the consideration of the same model within the framework of problems with different scales yields significantly different results [1, 2].

The stress tensor changing based on the internal pressure created by hydrogen penetrating into the crystal structure of metal also does not allow to examine the effect of low hydrogen concentrations. Within the framework of this approach, the visible effect of hydrogen is detected only at average concentrations above 10^{-5} [15], which are about ten times greater than the real threshold for steels.

Molecular dynamics is also used to model hydrogen embrittlement [41, 44]. Nevertheless, it allows to describe only micromechanisms at the apex of a microcrack or dislocation because of the smallness of the modeled ensembles. The same disadvantage is possessed by the quantum mechanical approach [8, 39]. It can be used only to describe the behavior of cracks in ideal crystals or to model the behavior of individual microcracks and dislocations because of the large heterogeneity of real metals.

3 New approaches to description of hydrogen destruction

It is necessary to underline one more discrepancy of modern approaches to hydrogen research. Almost all results and models were obtained by investigation of specimens charged with hydrogen. Initially, the saturation was carried out in gaseous hydrogen under pressure. Nowadays, 99% of the results are obtained by hydrogen saturation in salts solutions or in an electrolyte under the action of an electrical potential.

We detected experimentally the skin effect when saturating the metal samples with hydrogen in a solution of salts according to the NACE Standard TM0284-2003. After saturation of the samples, hydrogen is concentrated in a thin surface layer less than $50 \mu\text{m}$ thick. Our results in combination with the latest data from other studies devoted to the uneven distribution of hydrogen concentrations during cathodic

hydrogenation [26, 45] actually put an end to the technology of charging samples with hydrogen to study the effect of hydrogen on the properties of metals.

Developed by us model of multichannel diffusion of hydrogen has been verified by using various materials. It is also confirmed by the experiments of other scientists. We suppose this model to be promising for describing the transport of small concentrations of hydrogen in a solid. On the basis of this model, we constructed an experimental-calculation method for measuring the binding energies of hydrogen.

We have established that the limitations of the quasistatic approaches, as well as the methods of separation of motions, do not allow us to obtain certain predictive results in the case of complex systems involving several contacting details, which is important for practical applications [5]. Also, it is not possible to describe the process of loss of stability of deformation, which is characteristic for hydrogen embrittlement of steels, when zones of local hydrogen saturation and associated zones of local plastic deformations are formed.

This problem is especially important in terms of the fact that zones of plastic deformations localization are sources for the microcracks initiation, their formation triggers the destruction process.

4 Localization of plastic deformations

The inhomogeneity of the plastic deformation of metals and the "stress dropping" accompanying its formation in case of uniform deformation are the main manifestations of the Portevin-Le Chatelier effect [35]. Later detailed studies of plastic deformation fields made it possible to establish that during plastic deformation, plastic deformation bands are formed. The characteristic distance between these bands depends, in particular, on the grain size [46].

A lot of references are devoted to the explanation and modeling of the Portevin-Le Chatelier effect, see the review [38]. The main mechanism is the motion or even "jumping" of dislocations inside the metal during plastic deformation and their interaction with vacancies, foreign inclusions and other structural imperfections that contribute to the unstable motion of dislocations and the formation of plastic deformation bands. Moreover, the effect itself is often used for testing various models of displacement of dislocations and their interaction with the metal structure [46].

This dislocation approach has a significant drawback. The mechanical characteristics calculated on the base of this approach highly depend on the dislocation density on the grain surface (for example, see [31]). This value can not be directly measured. Simulation of the behavior of a large number of dislocations requires the specification of the generalized energy characteristics of this ensemble, which requires the building of additional physical microparameters. The presence of a large number of parameters in the material characteristics makes it possible to approximate any experimental dependences and ensure fully correspondence between results of the dislocation modeling and the experimental data [46]. It is difficult and sometimes impossible to measure these parameters even in case of complete destruction of the test specimen (for example, in case of beam irradiation in an electron microscope). In other words, the dislocation model does not allow us to obtain a quantitative prediction without preliminary adjusting of the parameters based on the the results

of testing the material with the same initial dislocation density.

These problems led to the emergence of generalized models of the localization of plastic deformation, considering it as a process of loss of stability of motion. In this regard, deformation with a small constant velocity is considered. Elements cause instability of solutions are introduced into the equations of a continuum.

There are three main approaches:

1. Determination of the dependence of the speed of change of stresses on the speed of change of deformations, which has a site with a negative slope, see for example [21, 34].
2. Introduction of non-linear dependencies based on simulation of the diffusion of vacancies and associated dislocations and non-linear constitutive equations of the material in the equations of continuum, for example [18, 27].
3. Description of the dislocations formation as a random process developing in time, transition to stochastic dynamic equations of continuum[23]. In this case, the nonuniformity of the deformation process is described as stochastic instability.

Thus, the loss of stability of a uniform uniaxial deformation of material is modeled either by direct violation of the energy conservation law (by specifying an unstable part of its characteristic or by instability in a stochastic system describing the behavior of dislocations) or by complicating the continuum model by introducing moment stresses, grains rotations and plastic shears, or by means of a bifurcation point arising from the introduction of additional nonlinear relations in the constitutive equations [24]. These nonlinear relationships are often obtained from consideration of the processes of nucleation and migration of dislocations. The process of localization of plastic deformations is considered as dynamic and wave process.

These assumptions contradict some experimental data and the initial statement of the problem, as underlined by the authors of the models described above. For example, in [3], the interaction of dislocations with structural inhomogeneities is considered as the main physical mechanism for localization of plastic deformation. In [46] an experimental dependence of the geometric characteristics of local bands on grain sizes was obtained.

One way or another, the authors of all the above-mentioned articles agree that the original natural heterogeneity of the material can play an essential role in the occurrence of localization of plastic deformation.

At the same time, except for [4], in all the papers known to us such an inhomogeneity is taken into account binary. For example, in [13] it is given as a soft metal model containing more rigid grains of another metal. In [25], the inhomogeneity is given as a separate deformation model for grain boundaries.

An important manifestation of the effect of localization of plastic deformation is the formation of a system of intersecting bands on the surface of a material such as a “chessboard”. For the first time this term was proposed by V.E. Panin in [33]. According to his research, the effect of the “chess” distribution of normal and tangential stresses on interfaces was experimentally discovered and theoretically

confirmed. Also conclusion about the important role of the observed effect in various phenomena of nature was made.

An alternative explanation for the formation of the “chess” surface character is given in Morozov [29]. The authors considered the compression of different elastic bodies. Within the framework of a geometrically linear formulation, both isotropic elastic bodies in a two-dimensional formulation (plates, plates on an elastic base) and transversely isotropic elastic half-spaces and layers were investigated. It was shown that exceeding of the critical value of the compressive load lead to yield of material from the loading plane. In this case, wave formation occurs in the material, which leads to the appearance of a regular structure on the surface, which also resembles the “chessboard”.

5 Localization of plastic deformations and hydrogen embrittlement as a single problem of modern mechanics of solids

All problems of localization of plastic deformation intersect with the problems of formation of local zones of hydrogen embrittlement. They are a manifestation of the dynamic instability of the material. The only difference is that the dynamic instability caused by hydrogen is associated with its diffusion, whereas in the case of plastic flow it is associated with the migration of dislocations. That is, additional degrees of freedom, which can give energy for the development of instability, appear. It is difficult to model an unstable process related to localization, since all approximate approaches have an approximation error that increases in time and can not be estimated from above.

Many continuum material makes it possible to describe the emerging instability as a structural transition. Nevertheless, recent data indicate failure of a homogeneous material model, since localization of plastic deformations leads to volumetric heterogeneity of mechanical properties. The surface effect, both in redistribution of hydrogen concentrations and in plasticity, leads to talk about new approaches to modeling the behavior of materials outside the elastic region.

These new approaches shall take into account the redistribution of diffusely mobile components and the special role of the solid surface during plastic deformation and material destruction.

6 Conclusion

Accounting of dynamic unstable processes in the framework of solution of mechanics problems is necessary due to the logic of research development and the problems of state-of-art technology. The main problem is that the accuracy of any approximations falls with time, since the error is also an unstable process.

When calculating the strength, it is necessary to take into account the spatial inhomogeneity of both plastic deformations and the distribution of hydrogen, since they directly affect the mechanical properties of materials.

Plastic waves are accompanied by the transfer of diffusely mobile hydrogen. The presence of a boundary surface layer makes the influence of the boundary conditions on these processes not essential.

Under these circumstances, it is natural to use the wave approximation when studying the processes of redistribution of natural small hydrogen concentrations in structural materials under the influence of external loads, taking into account the mutual influence of hydrogen diffusion, hydrogen accumulation in critical areas, heterogeneity of plastic deformations and changes in the mechanical properties of the material associated with the accumulation of hydrogen.

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