DISLOCATION MODEL OF HYDROGEN-ENHANCED LOCALIZING OF PLASTICITY IN METALS WITH BCC LATTICE*

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A mechanism of the influence of hydrogen-enhanced localizing of plasticity on the stage of initiation of a submicrodefect and growth of a macrocrack in metal with bcc lattice is proposed. A mathematical model of hydrogen embrittlement in metals with bcc lattice was constructed, which allows for the effect of hydrogen-enhanced localizing of plasticity and hydrogen influence on surface energy of a submicrocrack. It is established that metal susceptibility to hydrogen embrittlement is increased with grain size refinement.

Keywords: *arc welding, welded joints, high-strength low-alloyed steels, hydrogen brittleness model, metals with bcc lattice, grain size, hydrogen-enhanced localizing of plasticity, brittle fracture*

In welding of HSLA steels there exists the probability of initiation of hydrogen-induced cold cracks in the welded joint, which are a manifestation of hydrogen brittleness (HB) of metal under specific conditions of thermodeformational cycle of welding [1]. This phenomenon is based on the mechanism of hydrogen interaction with dislocations [2–5], leading to a change of properties of dislocation clusters [6, 7].

Properties of both individual dislocations and dislocation clusters largely determine the mechanical properties of metals. The difference between theoretical yield point and the really observed value, metal capability for plastic deformation, strengthening, temperature dependence of yield point and many other properties are attributed to presence of dislocations in the real metals [8–12]. Main regularities of HB are also well described in terms of dislocation theory that is confirmed by experiments correlating plastic deformation rate in the metal and its HB sensitivity [1, 2, 13], as well as electron microscopy studies of hydrogen influence on behaviour of dislocation clusters and microcrack growth [14]. Under the impact of hydrogen, metal mechanical characteristics can deteriorate several times, and it is important to take it into account in prediction of the reliability and fatigue life of structures. This work describes HB mechanism allowing for the effect of hydrogen-enhanced localizing of plastictiy (HLP) on micro- and macrolevels and physicomathematical model of dislocation interaction in hydrogen-containing metal with bcc lattice.

Physical model of HB. Dislocations are capable of interacting with each other owing to presence of stress fields around them. Two edge dislocations of one sign located in one slip plane are repulsed with force F_D inversely proportional to distance r between them [15]:

$$F_D \sim 1/r. \quad (1)$$

In hydrogen-containing metal the nature of interaction of edge dislocations changes somewhat. In addition to the force of interaction with the stress field of adjacent dislocation F_D, an additional force is applied to the edge dislocation that is caused by interaction of this dislocation stress field with hydrogen atoms concentrated around the adjacent dislocation [6, 7]. Thus, an additional force of attraction F_H arises between two edge dislocations of one sign located in one slip plane, which is caused by presence of hydrogen atoms around the dislocations. The magnitude of this force depends on metal temperature and hydrogen concentration. The force of interaction of hydrogen atom with edge dislocation rises in inverse proportion to the square of distance between them [16]. Therefore, allowing for certain approximations, dependence of force F_H on distance between the dislocations has the following form:

$$F_H = \frac{\alpha}{r^2}, \quad (2)$$

where α is the coefficient of proportionality.

Therefore, in hydrogen-containing metal the resultant force of interaction is smaller than that in metal without hydrogen. Effect of reduction of the resultant force of interaction between edge dislocations $F_{RES} = F_D - F_H$ in the metal, containing hydrogen, was experimentally confirmed in [14], and was called hydrogen-enhanced localized plasticity.

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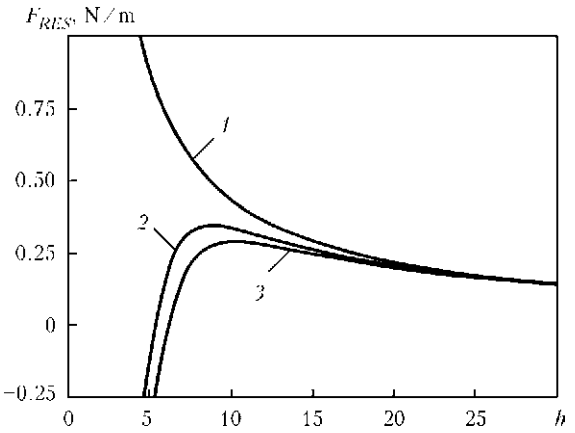

Figure 1. Dependence of resulting force of interaction of two edge dislocations F_{RES} on distance between them h (in modules of Burgers vector) at different concentration of diffusible hydrogen (metal temperature T = 300 K) [17]: *1* — 0; *2* — 5; *3* — 10 cm^3/100 g

In work [17] it is shown that a low hydrogen concentration characteristic for metal of welded joints from HSLA steels [18] is sufficient for appearance of the effect of HLP in iron. The limiting stage of initiation of a submicrodefect is the coalescence of two first dislocations in the cluster tip, as a much smaller force is required for dislocation joining the already formed submicrodefect [19]. Therefore, reduction of F_{RES} magnitude essentially facilitates coalescence of edge dislocations into microdefect nucleus. In hydrogen-containing iron the value of force, required for coalescence of the first two edge dislocations into submicrodefect nucleus, can drop 2 times and more (Figure 1). Therefore, under certain conditions presence of Cottrell atmospheres essentially changes the properties of dislocation clusters, leading to a change of metal mechanical properties.

As is seen from Figure 1, in hydrogen-containing metal there is a certain threshold distance between the dislocations, after achievement of which hydrogen influence on the force of interaction of dislocations rises noticeably. Therefore, hydrogen presence starts affecting the macroscopic strength of the metal after the dislocation density has reached a certain threshold value.

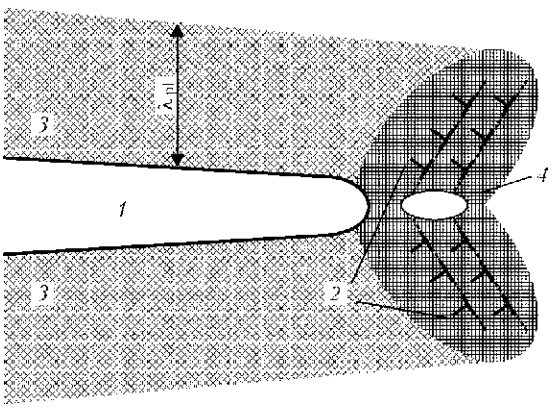

Figure 2. Schematic of macrocrack growth in the metal: *1* — macrocrack; *2* — field of plastic deformations ahead of its tip; *3* — layer of plastically deformed metal; *4* — microdefect

Initiation and propagation of a submicrocrack in the metal grain allowing for the effect of HLP be divided into three stages:

- hydrogen atoms concentrate around edge dislocations present in the cluster, thus reducing the force of repulsion between the dislocations and faciliting their coalescence;
- under the impact of external stresses the dislocations in the cluster tip coalesce and form a submicrocrack nucleus. Hydrogen present on these dislocations is partially entrapped by submicrocrack stress field, and partially penetrates into its volume;
- submicrocrack growth leads to fracture of metal grain and formation of a stable microdefect, which is capable of growing up to macrodimensions under the impact of external stresses and hydrogen.

One of the most probable mechanisms of macrocrack development in the metal is formation of a microdefect ahead of its tip and its subsequent coalescence with the crack (Figure 2) [10]. Under the impact of stresses, a plastic strain region forms in the metal ahead of the growing crack tip. During crack growth it leads to formation under its surface of a layer of plastically deformed metal, λ_{pl}, the thickness of which depends on applied load: the higher the load, which should be applied for formation of a microdefect ahead of the crack tip, the greater the λ_{pl}. Energy required for macrocrack growth consists of two parts — energy of free surface formation, and energy of subsurface layer of plastically deformed metal. In the metal not containing hydrogen, specific energy required to form such a layer, is by several orders of magnitude higher than the specific energy of crack-free surfaces [20]. Owing to the effect of HLP, the stress required to form a microdefect ahead of macrocrack tip, decreases considerably. Therefore, in a sample containing hydrogen value of λ_{pl} and percent of plastically deformed metal are much lower than in metal without hydrogen. Effect of HLP reduces the most energy-consuming component of crack growth, namely, formation of a subsurface layer λ_{pl}. Therefore, in hydrogen-containing metal with bcc lattice, macrocrack propagation should proceed in a more brittle mode, and should require much less energy, which is exactly what was observed in the experiments [1, 2].

Based on the proposed HB mechanism, an assumption can be made about the difference in fracture processes on macrolevel in samples of one and the same metal with and without hydrogen. While at the initial stage of plastic deformation dislocation density is low, change of dislocation structure in the samples proceeds in a similar mode, irrespective of hydrogen content. It means that at mechanical testing on macrolevel metal properties practically coincide. With increase of metal plastic deformation dislocation density rises. When as a result of increase of dislocation density the average distance between them reaches a certain

threshold value, the effect of HLP will be manifested in hydrogen-containing metal. As a result, more intensive formation of submicrodefects starts in the metal that will lead to an irreversible change of the mechanical properties of such a metal on macrolevel.

Thus, there should be a threshold plastic deformation, the value of which depends on metal sensitivity to HB, testing temperature and hydrogen concentration. If metal containing hydrogen is deformed to a smaller degree of plastic deformation, then after hydrogen degassing its mechanical properties will recover. After achievement of threshold plastic deformation, however, owing to presence of a large number of microdefects in the sample, metal properties after degassing will not recover, and it will still fail in a more brittle manner than the metal that did not contain any hydrogen initially.

HB mathematical model. Dislocation rearrangements leading to initiation and propagation of a submicrocrack in hydrogen-containing metal can be described with application of either force [3, 19] or energy criterion [21, 22]. Application of force criterion of submicrocrack initiation, when behaviour of each individual dislocation is calculated proceeding from forces acting on it, gives a more detailed picture and is easily realized, when studying the behaviour of a small number of dislocations, as it was shown in the case of two dislocations interaction. Nonetheless, an energy criterion of submicrocrack initiation and propagation is used in this work for calculations. This is related to the fact that if a cluster has five and more dislocations, then in case of application of force approach for calculation the derived system of equations has no analytical solution, even when there are no Cottrell atmospheres around the dislocation. Now, allowing for hydrogen influence makes such calculations more complicated by an order of magnitude. For metal containing no hydrogen the problem of dislocation behaviour in a large cluster can be solved, if continuum model of the cluster is considered instead of discrete dislocations [11, 15, 19]. With such a problem definition the cluster is characterized not by the position of each individual dislocation, but by dislocation density $\rho(x)$ in a point. However, as shown by calculations, continuum model gives an inadequate assessment of the stressed state inside the cluster proper [11]. Therefore, when allowing for the influence of the effect of HLP, when it is the question of interaction inside the dislocation cluster proper, this continuum model cannot be applied. Energy criterion of submicrocrack formation and growth allows circumventing this problem.

At simulation of metal HB the classical schematic of submicrocrack formation from a plane cluster of edge dislocations was taken as a basis. In the case of metal containing no hydrogen, the total energy of plane cluster of edge dislocations–submicrocrack system is equal to [10, 21–24]

$$W = \frac{nb^2G}{4\pi(1-\nu)} \ln \frac{4d}{L} + \frac{(N-n)^2b^2G}{4\pi(1-\nu)} \times \\ \times \ln \frac{4\pi\sqrt{e}(1-\nu)d}{(N-n)Gb} \tau_{eff} - \frac{\pi(1-\nu)L^2}{8G} \sigma^2 + 2\gamma L, \quad (3)$$

where b is the Burgers vector; G is the shear modulus; ν is the Poisson's ratio; L is the submicrocrack length; n is the number of dislocations which formed the submicrocrack; N is the maximum number of dislocations in the cluster; σ is the principal normal stress; γ is the specific surface energy of the metal which changes as a result of hydrogen impact; d is the size of metal grain; e is the Napierian base; τ_{eff} is the effective tangential stress in the slip plane.

Penetrating into the submicrocrack volume, hydrogen decreases its specific surface energy. During submicrocrack growth its volume increases, and dislocations joining it contribute new portions of hydrogen. This leads to a constant change of hydrogen pressure inside the submcirocrack and to a change of metal specific surface energy. Therefore, in case of hydrogen-containing metal, the last addendum in equation (3) should be replaced by integral

$$2 \int_0^{L_f} \gamma(L, N, n, T) dL, \quad (4)$$

where L_f is the final length of the submicrocrack.

In hydrogen-containing metal the work which should be done to bring together dislocations removed from each other to distance r, is found from the following equation:

$$\int_{+\infty}^{r} F_{RES} dr = \int_{+\infty}^{r} F_X dr - \int_{+\infty}^{r} F_H dr, \quad (5)$$

where $\int_{+\infty}^{r} F_X dr$ is the work which should be done in the metal containing no hydrogen, and, therefore, it is already allowed for in equation (3).

With shortening of the distance between the dislocations, their stress fields are superposed. As a result, the energy of the bond of hydrogen with dislocations becomes higher. Let us assume that at the beginning, when dislocations are located at a considerable distance from each other, the total energy of their bond with hydrogen is equal to W_d, and when they are at distance r – W_d^r from each other, then according to the law of energy conservation

$$\int_{+\infty}^{r} F_H dr = W_d^r - W_d. \quad (6)$$

Therefore, hydrogen, located on dislocations during shortening of interdislocation spacing, gives part of its potential energy to the submicrocrack

nucleus, thus reducing the work required for dislocation coalescence:

$$\int_{+\infty}^{0} F_H dr = W_L^0 - W_d, \qquad (7)$$

where W_L^0 is the energy of hydrogen bond with submicrocrack nucleus.

Integrating over the entire region taken up by Cottrell atmosphere, we will find the energy of the bond of hydrogen with edge dislocation per a unit of length:

$$W_d = \int\int_{\Omega_d} u_d(x, y) C_d(x, y) dx dy \qquad (8)$$

where Ω_d is the area taken up by Cottrell atmosphere around the dislocation; $C_d(x, y)$ is the hydrogen concentration calculated on the basis of Fermi–Dirac statistics.

Energy of interaction of a hydrogen atom, located in point (x, y) relative to dislocation nucleus, is calculated from equation [16]

$$u_d(x, y) = -\frac{Ay}{x^2 + y^2}, \qquad (9)$$

where A is the material constant determined experimentally.

In the first approximation at evaluation of the energy of hydrogen bond with submicrocrack W_L the latter can be regarded as a superdislocation. Therefore, W_L value is found in a similar way:

$$W_L = \int\int_{\Omega_L} U_L(x, y) C_L(x, y) dx dy \qquad (10)$$

where Ω_L is the region taken up by Cottrell atmosphere around the submicrocrack; $U_L(x, y)$ is the energy of the bond between hydrogen atom and submicrocrack.

Allowing for equations (7), (8) and (10), total change of submicrocrack energy caused by hydrogen and $(N - n)$-dislocations that did not join the defect can be written as

$$W_H = -(W_L + (N - n)W_d) = -\int\int_{\Omega_L} U_L C_L dx dy - (N - n) \int\int_{\Omega_d} u_d C_d dx dy \qquad (11)$$

Allowing for equations (3), (4) and (11), energy of plane cluster of edge dislocations–submicrocrack system for hydrogen-containing metal is equal to

$$W = \frac{(nb)^2 G}{4\pi(1 - \nu)} \ln \frac{4d}{L_f} + \frac{(N - n)^2 b^2 G}{4\pi(1 - \nu)} \ln \frac{4\pi\sqrt{e}(1 - \nu)d}{(N - n)Gb} \tau_{eff} - \frac{\pi(1 - \nu)L_f^2}{8G} \sigma^2 + 2\int_0^{L_f} \gamma dL - \int\int_{\Omega_L} U_L C_L dx dy - (N - n) \int\int_{\Omega_d} u_d C_d dx dy \qquad (12)$$

To calculate the behaviour of dislocations and submicrocrack, it is necessary to differentiate equation (12) by L_f and n. Derived equations are used to determine the conditions, at which the submicrocrack becomes unstable, i.e. breaks up.

At calculation it was assumed that the maximum slip length of edge dislocation is equal to grain size, while hydrogen concentration in the metal is equal to 10 $cm^3/100$ g.

Quantity of hydrogen, which is transferred by edge dislocation to the site of submicrocrack initiation, was determined on the basis of relationships proposed in [25]. Number of dislocations in the cluster N and τ_{eff} was evaluated on the basis of Hall–Petch relationship [11].

Derived relationships were the basis to perform computer simulation of fracture of iron grain with bcc lattice by dislocation mechanism (Figure 3). In calculation it was assumed that in the temperature range T = 350–250 K shear modulus and Poisson's ratio remain constant. Therefore, breaking stress of a grain of metal not containing hydrogen, will also remain constant in this range. If HLP is taken into account in the mathematical model, the stress, which is to be applied to break the grain, decreases, and microdefect length and number of dislocations joining it increase. In some cases, lowering of the strength of hydrogen-

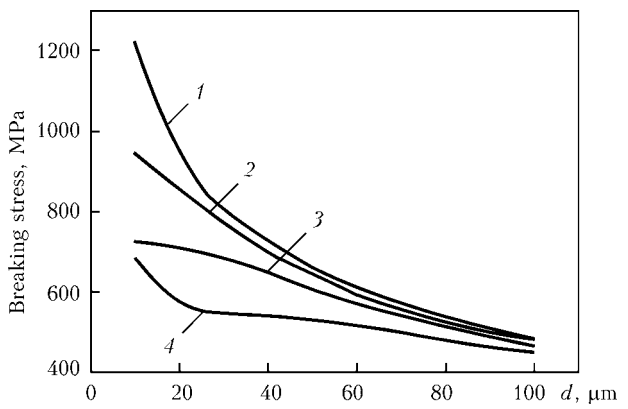

Figure 3. Dependence of breaking stress of grain of iron with bcc lattice on its size d at different temperatures: *1* — grain of metal without hydrogen at T = 350–250; *2* — 350; *3* — 300; *4* — 250 K

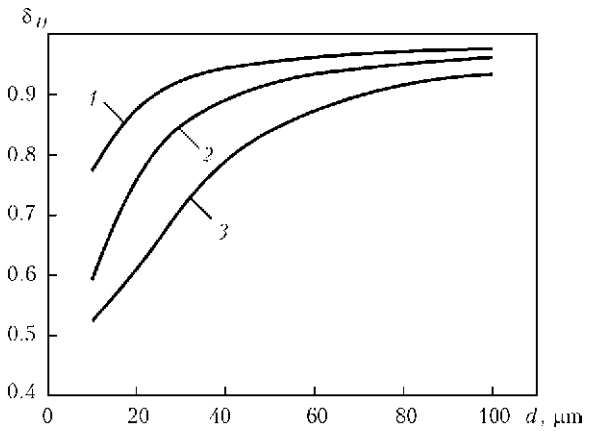

Figure 4. Dependence of relationship $\delta_H = S_C^H / S_C^0$ on size of grain d at different temperatures: *1* — T = 350; *2* — 300; *3* — 250 K

containing iron is equal to 40–50 % of the nominal value. A certain increase of breaking stress on curve *4* in Figure 3 at d = 25–10 μm is attributable to the nature of HB temperature-rate dependence [1, 2]. It is seen that in the metal containing hydrogen the effect of saturation can be manifested, when grain refinement practically does not increase its strength (see Figure 3). Therefore, increase of steel strength due to grain refinement is rational only up to a certain limit value of d. Note that d value depends on metal sensitivity to HB, hydrogen concentration and operating temperature range.

Metal susceptibility to HB can be expressed through decrease of breaking stress [1]:

$$\delta_H = S_C^H / S_C^0,$$

where S_C^H, S_C^0 is the true breaking stress of metal grain with and without hydrogen, respectively. According to performed calculations at comparatively large values of grain size d (in the range from 100 up to 50 μm), dependence of the degree of lowering of metal brittle strength δ_H can be approximated by a straight line (Figure 4). This is in agreement with experimental data given in [2, 26], which were obtained for armco-iron and low-carbon steel. At further grain refinement, however, metal sensitivity to BH rises abruptly. At other conditions being equal, grain refinement raises the degree of lowering of metal brittle strength. It should be noted that the absolute value of breaking stress S_C^H of hydrogen-containing metal still grows somewhat with grain refinement (see Figure 3).

As was noted above, in calculation it was assumed that the length of dislocation slipping is equal to grain size d. Therefore, according to the results given in Figure 4, HB degree rises with shortening of free sliding length of dislocations. Thus, derived conclusion on higher HB susceptibility of metal with finer grain can be interpreted as a theoretical explanation for the experimental fact that metal HB susceptibility rises with increase of its strength.

CONCLUSIONS

1. A mechanism of HB of metals with bcc lattice was proposed, which allows for the effect of hydrogen-enhanced localizing of ductility on the micro- and macrolevel.

2. A model of fracture of metal grain by the dislocation mechanism for hydrogen-containing metal was improved.

3. It is shown that at all other conditions being equal metal grain refinement will lead to an increase of HB degree, although the absolute value of breaking stress of hydrogen-containing metal increases with grain refinement.

4. It is shown that for hydrogen-containing metal increase of its strength due to grain refinement it rational only up to a certain limit, which depends on hydrogen concentration, operating temperature and HB sensitivity.

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