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МЕТАЛЛИЧЕСКИЕ ПОВЕРХНОСТИ И ПЛЁНКИ

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Analysis of Metal Corrosion under Conditions of Mechanical Impacts and Aggressive Environments

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Based on standpoints of the surface physics, fracture mechanics and electrochemistry, a mathematical model of the physical and chemical processes near the crack tip of a metal under mechanical loads in aqueous electrolyte solutions is developed. Calculations of the energy and electrochemical characteristics are performed for the steel 20 in the 3% solution of sodium chloride. Parameters of the Tafel-type relationship between the anode current and the difference of electrode potentials are analysed. Well-known Kaeshe expression for the current density on the juvenile surface of a crack bottom is generalized both by linear approximation of the dependence of corrosion current density on surface energy of plastic deformation of the metal and with accounting for increase of mechanical tensile stress up to yield limit.

Key words: metal, crack, juvenile surface, mechanical tension, electrochemical overpotential, surface energy, corrosive current.

З позицій фізики поверхні, механіки руйнування та електрохемії сформульовано математичний модель фізико-хемічних процесів у вершині тріщини металу при його механічному навантаженні у водному розчині електроліту. Проведено розрахунки енергетичних та електрохемічних характеристик для криці 20 у 3% розчині хлориду натрію. Проаналізова-

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но параметри рівняння типу Тафелевого між анодним струмом і ріжницею електродних потенціялів. У лінійному наближенні зв'язку густини корозійного струму з поверхневою енергією пластичного деформування металу та з урахуванням зростання механічних розтягувальних напружень до границі плинности узагальнено відоме співвідношення (Kaeshe) для густини струму на ювенільній поверхні дна тріщини.

Ключові слова: метал, тріщина, ювенільна поверхня, механічне напруження, електрохемічне перенапруження, поверхнева енергія, корозійний струм.

Исходя из положений физики поверхности, механики разрушения и электрохимии сформулирована математическая модель физико-химических процессов в вершине трещины металла при его механическом нагружении в водном растворе электролита. Проведены расчёты энергетических и электрохимических характеристик для стали 20 в 3% растворе хлорида натрия. Проанализированы параметры уравнения типа Тафеля между анодным током и разницей электродных потенциалов. В линейном приближении связи плотности коррозионного тока с поверхностной энергией пластического деформирования металла и с учётом увеличения механических растягивающих напряжений до предела текучести обобщено известное соотношение (Kaeshe) для плотности тока на ювенильной поверхности дна трещины.

Ключевые слова: металл, трещина, ювенильная поверхность, механическое напряжение, электрохимическое перенапряжение, поверхностная энергия, коррозионный ток.

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1. INTRODUCTION

It has been established that physical and chemical description of corrosion process on surfaces of metal constructions as a stationary process of oxidation with formation of passivating films on the metal surface does not reflect adequately the whole complexity of degradation processes in metals [1–3]. In particular, real technical system is normally operated under conditions of aggressive environments and mechanical fields (stress corrosion [4, 5]) followed by formation of cavities, corrosion pits and cracks, and their growth. Thus, corrosion processes in such objects should be treated comprehensively with taking into account the mechanical factor and probable electrochemical processes. In another words, the corrosion as by its essence is an electrochemical process with formation of new surfaces and developed cracking.

The paper is concerned with an impact of external corrosive environment and mechanical loading on variations in mechanical and electrochemical parameters characterizing corrosion current in the crack tip under mechanical loading within zero to yield limit σ_T of a metal.

This study aims to analyse macroscopic relationships of surface phys-

ics, destruction mechanics, and electrochemistry to establish relationships between energy variations and plastic deformation of the surface, overpotential of anodic reaction for a δ -wide juvenile surface (JS) (of the width δ), and electric current in the crack tip at the boundary of elastically deformed metal and ambient corrosive environment.

2. OBJECT OF STUDY AND MATHEMATICAL MODEL

The object of study is a loaded metal with a crack on its surface in aqueous electrolyte solution.

Passive films presented on the surface are destroyed at the crack tip under loading, and the δ wide JS and plastic deformation zone appear [3]. The said juvenile surface is the just formed surface of a metal, free of oxides and other contaminations [3, 4]. In the first approximation, we treat the geometric parameter δ at the crack tip as its opening displacement δ_{1C} . The crack tip and the JS in particular extend into the body volume. Near the crack tip, the cathode and anode reactions take place. Corrosive dissolution corresponds to the metal anode reaction. We address the crack tip as an anode (A), beyond it on the side surfaces there is the cathode region (K) [4]. The 'A–K' system forms an electrochemical couple.

To obtain quantitative assessment of impact of aggressive environment on energy characteristics of surface layers, we use analytical expressions, which connect cracking parameters and intensity of electrochemical reactions in the cracks vicinity.

The stress intensity coefficient (SIC) $K_{\rm 1SCC}$ [Pa·m^{1/2}] relates to the crack tip opening displacement $\delta_{\rm 1C}$ and overpotential ζ of the metal dissolution reaction by relationships [3, 6, 7] (here, the overpotential means deviation of the electrode potential from its equilibrium thermodynamic value during live electrode polarization [8, 9]):

$$K_{1SCC} = \sqrt{(W_{pL} - z_{si}F\rho\delta\zeta/M)E/(1 - v^2)}, \quad K_{1SCC} = \sqrt{E\sigma_T\delta_{1C}},$$
 (1)

where $z_{\rm si}$ —formal charge of solvated ions, F—the Faraday constant, δ —width of approaching micro crack front [m], M—molecular mass of a metal [g/mole], $K_{\rm 1SCC}$ —threshold value of the SIC (minimal value corresponding to onset of the corrosive crack propagation), $W_{\rm pL}$ —specific energy spent for plastic deformation of the subsurface level when new (juvenile) surface is formed in it, E and V—elastic modulus and Poisson's coefficient, respectively, σ_T —yield limit of metal.

Parameter $W_{\rm pL}$ can be found in well-known Griffith-Irwin-Orowan relationship (the strength criterion) [10]:

$$\sigma_* = \sqrt{4EW_{\rm pL}/[\pi L_T(1-v^2)]}, \quad \sigma_* = \sqrt{4EW_{\rm pL}/(\pi L_T)},$$
 (2)

where L_T —length of the crack.

Here in formulae (2), first formula is written for a plane deformation, second one—for a plane stressed state, σ_* —critical stress, $W_{\rm pL} = J/2$, J—the Rice's integral having energetic meaning [11].

In paper [9], empirical relationship linking the SIC to the $W_{\rm pL}$ has been established based upon the study of contact deformation of different steel brands:

$$K_{\rm 1SCC} = a_1 \sqrt{W_{\rm pL}} - a_2$$
, $a_1 = 2.26 \cdot 10^8 \, rac{{
m N}^{1/2}}{{
m m}}$, $a_2 = 6.98 \, {
m MPa \cdot m}^{1/2}$. (3)

The Kaeshe relationship for the current density $I_{\rm as}$ at the crack tip complements the model [4]:

$$I_{\rm as} = \frac{\alpha \chi \Delta \psi_{\rm ak}}{\delta \ln([h+c+r]/\delta)},$$
 (4)

where α —angle at the crack tip, χ —electric conductivity of electrolyte, $\Delta \psi_{ak}$ —ohmic variation of potential between anode and cathode sections (anode—a tip, cathode—edges of a crack), c—crack depth, h+c+r—total depth of defect (cavity and crack), h—depth of cavity, r—curvature radius at the crack tip.

Expression (4) was written for a crack in unloaded metal. However, to operate elements of real constructions like pipelines, one needs to take into account conditions of corrosion under loading (stress corrosion) [12]. Therefore, it is necessary to modify expression (4) by adding mechanical parameters.

For this purpose, let us consider a model of cylindrical pipe of radius R and wall thickness d, having aforementioned defects and being in conditions of aggressive environment (Fig. 1).

Let us assume that the metal is in an electrolyte solution and has a surface defect in form of the cavity with a crack at its tip. Figure 2

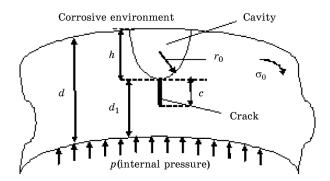


Fig. 1. Element of pipe with cavity (h) and crack (c) under impact of internal pressure p in corrosive environment.

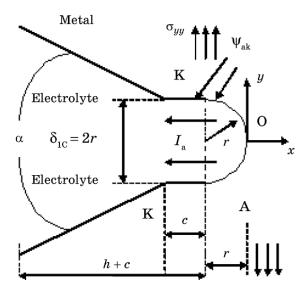


Fig. 2. A cavity (h) with a crack (c) in a pipe with markings of cathode (K) and anode (A) sections. Total depth of the defect h+c+r; α , δ_{1C} —angle and the crack tip opening displacement, $\Delta \psi_{ak}$ —difference of potentials between the anode and cathode sections, I_a —density of anode corrosive current.

shows projection of it on the xOy plane. External corrosive environment (in both the cavern and the crack) is an aqueous electrolyte solution. Under impact of mechanical load (uniaxial tension in the direction of the Oy axis with corresponding stresses σ_y), destruction of passive films at the crack tip occurs and the JS of width δ and length L is formed as well as the region of plastic deformations when $\sigma_{yy} = \sigma_T$ (σ_T —material yield limit). We model projection of the JS by a semicircle πr (Fig. 2).

The crack tip and the JS extend into the body volume in the direction of the x-axis toward the pipe centre almost perpendicularly to the boundary. Near the crack tip, the cathode and anode electrochemical reactions take place characterizing corrosion process of the metal dissolution. We consider the crack tip as an anode (A), and beyond its limits on the side surfaces, there is the cathode region (K). The 'A-K' system forms an electrochemical couple (Fig. 2).

In the first approximation, we assume that the empirical relationship between the crack opening displacement δ_{1C} and geometric parameter δ characterizing the JS width holds true

$$\delta_{1C} = \xi_{\delta} \delta$$
,

and corresponding value of the empirical constant $\xi_{\delta} = 1$.

In paper [9], relationship (4) is generalized by accounting for the $W_{\rm pL}$ and internal pressure $p_{\rm cr}$ acting upon the cylindrical pipe:

$$I_{\rm a} = I_{\rm as}(1 + \beta_W W_{\rm pL}) = \frac{\alpha \chi \Delta \psi_{\rm ak}}{\delta \ln(c/\delta)} (1 + \beta_W W_{\rm pL}), \tag{5}$$

$$p_{\rm cr} = \frac{2\sqrt{2}d\sigma_T}{3K_{\rm t}D} \frac{(1.5 + K_z)(r_0 + c)^4}{(r_0 + c)^4 + 0.5r_0^2(r_0 + c)^2 + r_0^4},$$
 (6)

where

$$K_{z} = \left(\frac{d_{1} - c}{2} \frac{2(d_{1} - c) + 3r_{0}}{d_{1} - c + r_{0}}\right) \left(\frac{d}{K_{t}} + \frac{r_{0}}{3} \left\{\frac{r_{0}^{3}}{(d_{1} - c + r_{0})^{3} - 1}\right\}\right)^{-1}, \beta_{W} = \frac{r_{0}}{d_{1} - c},$$

 $K_{\rm t}$ =(2.021-1.301 β -0.727 β ²-0.147 β ³) $d(d_{\rm 1}-c)$ —the notch sensitivity index, whose procedure of calculation is presented in [13], β_W —empirical coefficient of proportionality, r—radius of curvature at the crack tip, r_0 —critical value of r when plastic deformations occur at the crack tip.

The critical pressure (6) corresponds to a condition of reaching limiting, plastic, state according to the Huber-Mises-Hencky yield criterion [12]:

$$\sigma_{y}^{2} + \sigma_{z}^{2} + \sigma_{y}\sigma_{z} = \sigma_{T}^{2} / 3, \ \sigma_{y} = pD / (2d), \ \sigma_{z} = pvD / (2d),$$
 (7)

where σ_T —yield limit of the pipe metal, x, y, z—rectangular Cartesian coordinates, p—internal pressure inside the pipe.

In the relationships (7), we took into account for the mechanical stresses tensor $\sigma_y \equiv \sigma_{yy}$ and for σ_z that the pipe thickness d is considerably less than diameter D.

Relationships (1)–(7) compose a mathematical model for assessment of changes in the effective surface energy during plastic deformation, electrochemical overpotential and in the current density of the metal dissolution reaction at the crack tip on the metal surface during its loading in aqueous electrolyte solution (*i.e.* under conditions of stress corrosion).

3. BEHAVIOUR OF ENERGY AND ELECTROCHEMICAL PARAMETERS

3.1. Accounting for a Resistive Layer

The specific of cracking in electrolyte solution is emergence of a 'resistive layer' (RL) of the $H_{\rm L}$ thick and with practically dry its surface [14]. It is called a layer of complete hydration (solvation). Resistance of the RL is larger than the resistance of the diluted solution by 2–3 orders of magnitude [14]. Over time, when crack resides in the electrolyte, its thickness increases.

Let $\mathbf{r}(t)$, $\xi(t)$ be radius vectors of internal and external surfaces of the RL, correspondingly; $\Sigma(t) = 2\pi \xi^2(t)$, $\sigma(t) = 2\pi r^2(t)$ —areas of internal and external surfaces of the RL, which we assume to be concentric hemispheres.

In vector form, with accounting for spherical symmetry, relationships for the $H_{\scriptscriptstyle L}$ will be

$$\mathbf{H}_{\mathrm{L}} = \mathbf{r}(t) - \xi(t). \tag{8}$$

Motion of surfaces $\sigma(t)$, $\Sigma(t)$ with radii r(t), $\xi(t)$ is defined by the relationship [14]:

$$\frac{\partial \mathbf{r}}{\partial t} = \mathbf{I}_{a}(\sigma) \frac{M}{\rho z_{si} F}, \frac{\partial \xi}{\partial t} = \mathbf{I}_{\xi}(\Sigma) \frac{M}{\rho z_{si} F}.$$
 (9)

Here, $I_a(\sigma)$, $I_{\xi}(\Sigma)$ are densities of currents outflowing from these surfaces and depending on coordinates of the points, z_{si} —valence, ρ —density of material, M—molar mass.

The dissolution current of the crack bottom $i_a(\sigma) = i_a(\eta, c_{as}, c_{bs})$ depends on threshold concentrations of activated ions c_{as} , aqueous solvent c_{bs} and polarization η of the metal.

Polarization $\eta = \eta(\sigma)$ of the metal is connected with anode potential by relationship [14]:

$$\eta(\sigma, \mathbf{r}) = \varphi_a - \Delta \varphi_{rs}(\sigma, \mathbf{r}) - \Delta \varphi_0(\mathbf{r}), \tag{10}$$

where $\Delta\phi_{rs}(\sigma, \mathbf{r})$, $\Delta\phi_0(\mathbf{r})$ —ohmic potential drops in the volume of the RL. Losses in the electrolyte are relatively small.

In the first approximation, for the radius change, it is possible to use formula following from considerations of spherical symmetry and accounting for the Faraday law [14]:

$$r(t) = \left(\frac{3}{2\pi} \frac{IMt}{zF\rho}\right)^{1/3}.$$
 (11)

3.2. Calculation of the Change in Effective Energy and Overpotential at the Crack Tip

For the steel 20 ($T = 20^{\circ}$ C and p = 100 kPa), physical and mechanical characteristics are as follow [15, 16]:

$$z_{si} = 2$$
, $E = 213$ GPa, $\sigma_T = 245$ MPa, $v = 0.3$, $\rho = 7860$ kg/m³. (12)

With using (1)–(3), (12) and numerical data of paper [3], we obtain

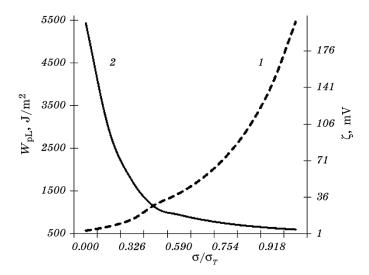


Fig. 3. Dependences of surface energy of plastic deformation $W_{\rm pL}(\sigma/\sigma_T)$ [J/m²] (1) and overpotential $\zeta(\sigma/\sigma_T)$ [mV] (2) within the range of mechanical tensile stresses $\sigma/\sigma_T = 0-1$ for the steel 20 in the 3% NaCl solution.

changes in effective energy during plastic deformation of the metal subsurface layer $W_{\rm pL} = f_1(\sigma/\sigma_T)$ and overpotential of anodic dissolution reaction $\zeta = f_2(\sigma/\sigma_T)$. Results corresponding to calculations of these dependences are presented in Fig. 3.

One can see from the dependences of Fig. 3 that the effective surface energy $W_{\rm pL}$ during plastic deformation grows by 9.5 times, and the overpotential ζ decreases by 40 times. These changes can be assessed quantitatively by the difference of maximal and minimal values attributed to their mean value:

$$w_1 = 2 \frac{W_{
m pL~max} - W_{
m pL~min}}{W_{
m pL~max} + W_{
m pL~min}} = 1.62, \ w_2 = 2 \frac{\zeta_{
m max} - \zeta_{
m min}}{\zeta_{
m max} + \zeta_{
m min}} = 1.90.$$
 (13)

3.3. Calculation of Electrochemical Parameters

To assess impact of tensile stresses on intensity of corrosion processes in the steel 20 residing in the 3% NaCl solution, in particular, at the crack tip at the moment of passive films' destruction (when anode current i_a considerably grows), we will use experimental data [3] approximated by the Tafel-type expression [3]:

$$I_a = I_0 \exp(DE/a), DE = E_0 - E_a,$$
 (14)

where a—the Tafel parameter for anode process.

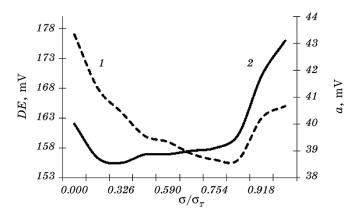


Fig. 4. Dependences of the electrode potential difference $DE(\sigma/\sigma_T)$ [mV] (1) and the Tafel coefficient $a(\sigma/\sigma_T)$ [mV] (2) of the relative tensile stress within the range of $\sigma/\sigma_{0.2} = 0-1$ for the steel 20 in the 3% NaCl solution.

By these data, we calculate values of the DE and a, presented in Fig. 4. Minimal values of the $DE(\sigma/\sigma_T)$ and $f_4(\sigma/\sigma_T)$ determined by the data of Fig. 4 are $DE_{\min} = 156 \,\mathrm{mV}$ when $\sigma/\sigma_T \approx 0.8$ and $a = 38.6 \,\mathrm{mV}$ when $\sigma/\sigma_T = 0.39$, correspondingly.

Because $DE_{\text{max}} = 177 \text{ mV}$ when $\sigma/\sigma_T = 0$ and $a_{\text{max}} = 43.1 \text{ mV}$ when $\sigma/\sigma_T = 1.0$, then we obtain the relative variations of the parameters:

$$w_3 = 2 \frac{DE_{\text{max}} - DE_{\text{min}}}{DE_{\text{max}} + DE_{\text{min}}} = 0.126, \ w_4 = 2 \frac{a_{\text{max}} - a_{\text{min}}}{a_{\text{max}} + a_{\text{min}}} = 0.110.$$
 (15)

By comparing expressions (13) and (15), it is evident that the relative variations of the effective energy and overpotential during increase of mechanical stress are considerable larger than the relative variations in the Tafel equation.

Comparison of data of Figs. 3 and 4 allows to state that there is no correlation between the $W_{\rm pL}$, ζ on the one hand and the DE, $a(\sigma/\sigma_T)$ on the other.

At the same time, in Fig. 3, we can allocate three ranges of the σ/σ_T variation. In the first range ($\sigma/\sigma_T \approx 0$ -0.16), DE and a are decreased. In the second range ($\sigma/\sigma_T \approx 0.16$ -0.84), parameters DE and a are changed insignificantly. In the third one ($\sigma/\sigma_T \approx 0.84$ -1), they grow considerably. Assessments of the said parameters can be indicative of changes of the electrode reaction type on the JS.

3.4. Generalization of Relationship between Electric Voltage and Current at the Crack Tip with Accounting for Mechanical Stress

Let us use experimental dependences [3] of current density i_a for the steel 20 in corrosive environment with different values of crack open-

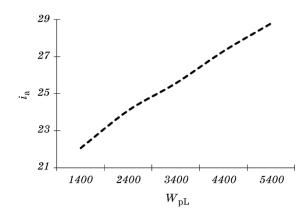


Fig. 5. Dependence of anodic dissolution current i_a [A/m²] on the plastic deformation energy $W_{\rm pL}$ [J/m²] within the range of $\sigma/\sigma_T = 0.59-1$ for the steel 20 in corrosive environment.

ing displacement $\delta_{\rm 1C}$ for stresses $\sigma/\sigma_T=0-1$. From dependences presented, $i_{\rm a}=f({\rm lg}\delta_{\rm 1C})$ and $i_{\rm a}=f(\sigma/\sigma_T)$, one can see that until $\sigma/\sigma_T=0.59$ ($\delta_{\rm 1C}=1.9~\mu{\rm m}$), density of anode current practically does not change, but after $\sigma/\sigma_T=0.59$, the $i_{\rm a}$ grows nonlinearly. Results of calculations by the LS-method has shown that dependence of the plastic deformation energy $W_{\rm pL}$ on the $i_{\rm a}$ ($i_{\rm a}=f_5(W_{\rm pL})$) after $\sigma/\sigma_T=0.59$ is practically linear (Fig. 5).

With considering the data of [3], for expression (5), we have:

$$I_{as} = 19.86 \text{ A/m}^2, \ \beta_W = 0.0000843 \text{ m}^2/\text{J} \ (W_{DL} = 1400 - 5456 \text{ J/m}^2).$$
 (16)

With accounting for the approximation (6) with respect to E_0 , E_a [3], which contains the surface layer energetic characteristic, $W_{\rm pL}$, we generalize well-known relationship (Kaeshe) (4) for the current density at the crack tip:

$$I_{\rm a} = I_{\rm as} e^{DE/a} (1 + \beta_W W_{\rm pL}) = \frac{\alpha \chi \Delta \psi_{\rm ak}}{\delta \ln(c/\delta)} e^{DE/a} (1 + \beta_W W_{\rm pL}). \tag{17}$$

Expression (17) describes dependence of the anodic dissolution current on electrochemical characteristics of the crack tip χ , ψ_{ak} , DE, a, the geometric α , δ , c, and the effective energy of plastic deformation W_{nL} .

Let us consider an example of a pipe made of the pipe steel X-70. According to [3], the limiting value of the crack critical depth $c_{\rm crg}=3$ mm is the criterial one.

Let us set following parameters:

$$P = 5 \text{ MPa} = 50 \text{ atm}, h = 4 \text{ mm}, d = 10 \text{ mm}, d_1 = 6 \text{ mm},$$

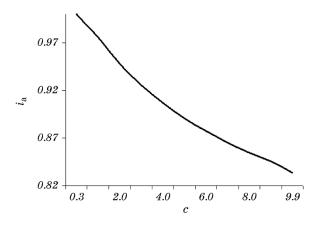


Fig. 6. Dependence of the corrosion current $i_{\rm a}$ on the crack length c.

TABLE 1. A pipeline resource.

Option	Corrosive rate, mm/year	The time of achieving a crack of critical depth of $0.7d$ over the years (17), years
1	0.3	8.9
2	1.0	2.6
3	1.5	1.8

$$D = 2R = 0.76 \text{ m}, \sigma_{\theta} = 372 \text{ MPa}, p_{cr} = 9.3 \text{ MPa}.$$
 (18)

With accounting for (12), (19) by means of (6), we obtain:

$$c_{\rm cr0} = 1.52 \text{ mm}.$$
 (19)

Here, $c_{\rm cr0}$ —initial value of the crack critical depth when condition of plasticity holds true at the crack tip. Then, the crack expands from $c_{\rm cr0}$ to $c_{\rm crg}$ due to corrosion (anodic dissolution).

Let initial condition for anodic current is:

$$I_{\rm a} = 1 \frac{{
m mm}}{{
m year}} \ {
m for} \ c_0 = 0.3 \ {
m mm}.$$
 (20)

With taking into account data [3], information in Figs. 1–3, and relationships (8)–(11), (17)–(20), we obtain the dependence $I_a = I_a(c)$ (Fig. 6) and assessment for a pipeline resource (Table 1) for three options of the corrosion initial speed.

4. CONCLUSIONS

A mathematical model based on standpoints of the surface physics and electrochemistry was expanded for assessment of the surface energy of plastic deformation, overpotential and current density for the reaction of metal dissolution at the tip of crack for loaded metal in aqueous electrolyte solution. Dissolution of metal on the juvenile surface has been treated with accounting for the coefficient of stresses intensity. The Kaeshe relationship for current density at the crack tip for loaded metal under uniaxial tension has been generalized with accounting for the $W_{\rm pL}$.

Based on analysis of dependences among the surface energy of plastic deformation, $W_{\rm pL}$, the anode energy overpotential, ζ , and the load (with stress σ/σ_T), it is found that, within the range of variation of tensile stresses from zero to the yield limit for the steel 20 in the 3% NaCl solution, the $W_{\rm pL}$ grows by 9.5 times, and the ζ decreases by 40 times.

Dependences of the electrode potential difference DE and coefficient a in the Tafel-type equation on the relative tensile stress $\sigma/\sigma_T = 0-1$ for the steel 20 in 3% NaCl solution were calculated. Based upon these dependences, we obtained information on changes in the character of electrode reactions on the juvenile surface.

The proposed technic of study of the energetic and electrochemical parameters of the stress-deformed state of metal pipe structures can be used for assessment of a number of physical and mechanical parameters of the corroding technical system (a pipeline), necessary to increase its service life.

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