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IMPROVING THE CORROSION RESISTANCE OF HOT-ROLLED STEEL PIPES

ПІДВИЩЕННЯ КОРОЗІЙНОЇ СТІЙКОСТІ СТАЛЬНИХ ГАРЯЧЕКАТАНИХ ТРУБ

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Abstract. ***Purpose.*** *Substantiation of the use of surface plastic and dynamic surface deformation (SPD, DSD) combined with anticorrosion inhibitory treatment of general-purpose carbon steel pipes to increase wear resistance of pipelines.*

Methodology. *Samples of hot-formed steel taken from test pipe fragments were subjected to various modes of surface plastic and dynamic surface deformation using corrosion inhibitors. Plastic deformation was carried out with a brush tool with different values of brush pressing against the pipe and processing time. Studies of the protective ability of inhibitors were carried out by an accelerated method with periodic moisture condensation according to DSTU ISO 6270-2:2015. A comparative metallographic study of the metal structure of pipes subjected to PPD and DPD with inhibitors was carried out.*

Results. *The results of experimental studies of the surface corrosion resistance and metallographic structure of 139x5 mm pipes made of steel grade 20 using three types of inhibitors and various modes of dynamic plastic deformation are presented.*

Scientific novelty. *The dynamic surface deformation of metal, which refines its grain structure, creates conditions for formation of "penetration channels" for the inhibitor into the metal structure. The use of DPD makes it possible to reduce the grain size in the surface layer, which suggests possibility of controlling the depth of the inhibitor penetration and, as a result, the corrosion resistance of pipes, depending on the operational requirements.*

Practical value. *It has been established that the use of dynamic surface deformation in combination with a certain inhibitor makes it possible to increase the corrosion resistance by 11–19 times.*

Key words: *hot-rolled pipe, general corrosion, inhibitor, plastic deformation, penetration channel, microstructure.*

Introduction.

Existing technologies for the production of hot-rolled pipes result in the formation of scale and coarse-grained decarburized layers on their surfaces. This contributes to the corrosion process of the pipe metal and leads to a decrease in its load-bearing strength, thereby increasing the likelihood of accidents during pipeline operation.

Metal failure due to corrosion is a significant problem in most industrial sectors, and its control and prevention must strike a balance between effectiveness and cost-effectiveness. Growing concern about environmental damage has greatly influenced this field, as corrosion protection must comply with waste management regulations in different regions [1]. The problem is exacerbated by the placement of pipeline routes in soil. On one hand, this hinders the monitoring of the pipeline's condition, while on the other hand, it causes environmental damage [1, 2].

Formulation of the problem.

Corrosion processes occur both on the surfaces and inside the walls of the pipe. Surface corrosion often occurs locally, but its progression can lead to premature failure of the entire structure. Internal corrosion poses an equally significant danger. Spreading across the pipe's cross-section, it can cause failure even though the pipe's surface remains visually undamaged [3].

The chemical composition of the steel significantly influences the corrosion rate. This influence is noticeable even in the case of closely related carbon steels [6].

Samples of hot-rolled pipes and Grade 20 steel were selected as the research materials. Although Grade 20 steel is widely used in pipeline construction, it does not possess high corrosion resistance. For this reason, galvanic coatings such as chrome, zinc, etc., are typically applied to it for operation in aggressive environments.

Depending on the chemical composition and production processes, a different structural state and its determining parameters are formed in steel products. The type of structure, its morphology (for example, the texture of hot-rolled and cold-rolled products), defects, grain size, dislocation density, which affect the limits of application of the selected material [5, 7].

The authors [5] conducted a detailed review of the influence of various structural states on corrosion resistance. However, there is no clear consensus on this matter. Nevertheless, it has been demonstrated that a coarse-grained structure contributes to increased corrosion resistance. Formation of a fine-grained structure is a very ambiguous factor affecting the degree of corrosion resistance. It has been shown in [8] that the small size of crystallites and the high density of dislocations accelerate dissolution of Fe in CO_2 medium.

It has been shown [5] that the size and spatial distribution of the cementite phase are an important aspect of corrosion, since the more noble cementite acts as cathodic centers, thereby affecting the corrosion resistance of the material.

One aspect of corrosion research is the study of texture in relation to the corrosion resistance of materials. It has been established that the activation energy of the dissolution of a close-packed surface is higher than that of a loose-packed one. When evaluating the texture and its role in corrosion, dislocation effects must be considered. Dislocations are understood as one-dimensional defects in materials and are closely related to development of the texture, since the high density of dislocations caused by tensile deformation causes weakening of the crystallographic orientation. This phenomenon is closely related to corrosion, as high corrosion rates have been obtained at locations where dislocations cross the surface.

The lower corrosion resistance of cold-rolled steel compared to that of hot-rolled steel is due to the high surface energy in the direction {100} since the plane with low surface energy provides a slow dissolution rate of atoms due to its closed atomic packing and results in high corrosion resistance.

As soon as the use of inhibitors to prevent corrosion of carbon steels is often the most economical option, it is of considerable interest to the industry to determine the limits of application of film-forming corrosion inhibitors. Commercial formulations of inhibitors almost never use a single molecule due to observed synergistic effects that increase their effectiveness.

It follows from the previous discussion that both external corrosion and internal corrosion are hazardous to pipeline systems. While in relation to external corrosion certain measures have been developed to combat it, this cannot be said in relation to corrosion occurring inside the pipe wall. The task is to increase the corrosion resistance of pipes.

One of the directions of the search is to find ways of anticorrosion inhibitory treatment of structural crystalline surfaces along the thickness of the pipe wall, which can be achieved by introducing the inhibitor to the crystalline structures.

A prerequisite for the possibility of the inhibitor introduction into crystalline structures is the very fact of the occurrence of corrosion, developing according to the type of electrochemical corrosion, that is, with participation of an electrolyte – an aqueous medium. Electrochemical corrosion refers to the type of heterogeneous corrosion, with participation of two phases – liquid (water) and solid; the surfaces of the elements of crystalline structures inside the bulk of the material act as a solid phase. Only one molecular layer of moisture can act as an aqueous medium [9]. Therefore, there are mechanisms for ingress of moisture into the monolith of the material, and, so, inhibitors can also enter it in a similar way to provide anti-corrosion inhibitory treatment.

Main part.

From practice, examples of penetration and movement of atoms and molecules inside monolithic bulks of materials are known. The phenomenon of segregation is also known, which consists in "instantaneous" movement of atoms of some elements in the bulk of metal to the surfaces [10]. In the practice of a number of processes, cutting fluids (coolants) are successfully used in the areas with extreme conditions. So, for example, during cutting, the pressure in the cutting center develops up to 50 MPa, and the temperature can locally rise to several tens of thousands of degrees. The positive effect is associated with the ability of the active principle of the coolant –

polarized dipole molecules of surface-active substances (surfactants) to penetrate into foci with extreme conditions. To ensure penetration into a zone with extreme conditions, "penetration channels" and driving forces are needed to ensure movement of surfactant molecules. Creation of "penetration channels" is provided by surface plastic deformation, its implementation options – roller, roller deformation with adjustable slip, dynamic deformation with brushes. In addition, the penetrating ability is affected by surface microroughness, its mechanical and chemical properties [11]. The driving forces are electric forces, the presence of electric dipoles in surfactant molecules, the presence of electric charges on the walls of microcracks. In [10], the fact of detecting charges on the walls of microcracks inside the material mass is noted. It can be assumed that this is due to formation of double electrical layers, which is typical for breaking any contacts. Some identified features of the use of coolant – the mechanisms of movement along the "penetration channels" can be taken as a basis for developing the task.

Previous studies conducted by the authors on the surface treatment process using a hydro-abrasive jet under pressure with the introduction of an inhibitor into the hydro-abrasive suspension have allowed the establishment that the anti-corrosion effect was absent when the surface was solely treated with the inhibitor outside of the hydro-abrasive treatment. This led to the conclusion that the anti-corrosion effect is associated with the surface deformation-induced strengthening effect of the hydro-abrasive jet on the surface. Simultaneously, the appearance of intense luminescence during the treatment was noted, which was attributed to the occurrence of electrostatic discharges from double electric layers upon contact breaking between abrasive particles and the material [12].The use of high-frequency deformation treatment and its effect on the corrosion properties of materials are most often considered [13] in terms of the impact of vibration on the weld pool, which contributes to development of a fine-grained structure, suppression of the formation of fine dispersed particles, especially at the grain boundaries of the final microstructure. Improvement of the corrosion characteristics of welded joints is probably associated with suppression of the formation of intermetallic compounds, due to mechanical vibration during solidification of the weld [14].

The relationship between vibration of the solid metal material is shown in [15]. The positive effect of the use of vibration during hardening of steel 35CrMoV on its properties, including corrosion resistance, is shown.

It has been suggested that the increased anti-corrosion effect is associated with the external deformation effect of the surface layer, which leads to refinement of the grain structure.

It is advisable to use inhibitors having a relatively simple chemical formula and minimal molecular sizes. For example, the inhibitor NaNO2 has a molecular weight of 68 g/mol, which is comparable to the molecular weight of water of 18 g/mol. Comparable dimensions create the same conditions for penetrating power. At the same time, it has been found that the same corrosion inhibitor can exhibit a different protective effect for different steel grades of pipes, which may be due to high sensitivity of the reagent to the nature of metal [16].

Identification of "penetration channels" can be carried out on the basis of modern ideas about the structures of nanomaterials and nanotechnologies. It has been established that during crystallization there is a mismatch in orientation of the crystallographic planes of neighboring crystal lattices, which reaches 5° in conventional materials [17]. In nanomaterials, the mismatch can reach several tens of degrees [9]. Significant disorientation also takes place during grain grinding by traditional deformation methods (PPD, DPD, etc.).

If there are mismatches in the placement of the crystallographic planes of the neighboring crystals being formed, appearance of free volumes at the junctions of the surfaces of neighboring crystalline formations is inevitable. This causes appearance of some residual porosity, free volumes, and in conventional materials with a small degree of mismatch up to 5° it significantly increases for nanostructures due to significant increase in mismatch up to several tens of degrees In nanomaterials, the amount of free volume significantly increases due to the increase in lattice mismatch by several tens of degrees. Under these conditions, there is no dense contact between the surfaces of contacting crystal elements, and the formation of free volumes is inevitable. As a result, grain boundary pores are located along the grain boundaries (GBs) and their clusters, and the majority of excess (free) volume is concentrated there [18]. In coarse-grained polycrystals, the free volume manifests as point defects, dislocation nuclei, and with grain size reduction, additional free volume in the form of imperfect GBs is added to them [12].

They are almost always group [18], which also predisposes possibility of movement of inhibitors in ionic form in the bulk of the material.

But, in addition to the free volumes in the structures, the near-border area directly adjacent to the GBs, which have a special structure and properties, deserves special attention. Grain boundaries "are the sources of dislocations and sinks for them". It is also noted that nanomaterials are distinguished by exceptionally high diffusion mobility of atoms, which is 5-6 orders of magnitude higher than that of conventional materials. In [18], the phenomenon of segregation is described, which consists in "instantaneous" movement of atoms of some elements in the bulk of metal to the surfaces. The "instantaneity" of movements should be linked to the extremely high rapidity of microlevel processes, which is 10^{-6}–10^{-11} s [17], and to the presence of "free volumes" in the structures that came into view only in connection with development of nanomaterials.

The noted features are key in the issue related to deformational refinement of grains and the behavior of inhibitor ions – the possibility of their penetration into the bulk of material. It can be seen from the foregoing that during plastic deformation of the surface layer with crushing of grains, conditions are created for the formation of "penetration channels" in the material from the complex of free volumes present in the bulk in combination with free volumes of movable deformable layers. From the practice of surface hardening treatment by deformation, examples are known when the deformation effect extends to the depth of up to 35–40 mm [19]. This corresponds to the wall thicknesses of hot-rolled pipes, therefore, the existing experience can be used for anti-corrosion treatment of pipes in the bulk of material throughout the entire thickness of the walls. Equipment for similar processing of external and internal surfaces of pipes has been developed. As a result, the pipe wall can be processed from two sides.

Considering the high brevity of micro-level processes (10^{-6}–10^{-11} s), the time of crushing-grinding of grains, corresponding to the direct crushing effect, is incommensurably large (it can be taken, in the first approximation, within 0.01–1 s). This exceeds the duration of microlevel processes by several orders of magnitude and means that during the time of deformation impact on the material, a large number of acts can occur for penetration of inhibitor ions into the bulk of material, their transportation through the "penetration channels", and formation of protective passivating films on the elements of the crystal structure. In addition to transportation of inhibitor ions along the "penetration channels", the previously noted diffusion conduction along grain boundaries can be carried out, where, due to misorientation of crystal lattices, interlayers of 1–5 nm with the high content of dislocations are formed along the grain boundaries (see above). These interlayers, in conditions of mobility and vibrational phenomena of a wide range of vibrations, can also contribute to solving the problem. The noted ultra-high diffusion permeability of nanomaterials should be associated with grain refinement, which can be compared with the common practice of hardening treatment with grain crushing [20].

Experimental studies of corrosion resistance of pipes.

The purpose of the experiment was to establish, on the basis of a previously conducted theoretical analysis, effectiveness of the use of inhibitors in combination with dynamic surface deformation by brush treatment to increase the corrosion resistance of pipes, as well as structural changes in the thickness of the pipe wall.

The specific feature of the work of brush tools is the high-speed impact of an elementary tool – a wire, followed by a deformation-cutting effect and the same high-speed separation from the surface to be treated. It should be noted that the type of action of the wire has common features with the type of action during hydroabrasive processing, where intense electrical phenomena are observed.

Hot-rolled pipes 139x5 mm in size made of steel grade 20 were subjected to dynamic brushing. Three types of inhibitors and their modifications were studied. Dynamic surface plastic deformation was carried out at the rate of movement of the brushes relative to the pipe surface of 9 m/s.

The speed mode and processing time were chosen in accordance with the results of studies [10, 21]. The brush feed in relation to the pipe surface was regulated in the range from 1.5 mm to 6 mm. The time for processing the pipe section, corresponding to the width of the brush tool, did not change – 15 sec.

Accelerated tests of the protective ability of conservation compositions in combination with surface dynamic deformation were carried out with periodic moisture condensation according to DSTU ISO 6270-2:2015.

The results of tests with periodic moisture condensation for 24 hours are presented in Figures 1–4.

Analysis of the results of the corrosion resistance of pipes presented in the figures allows us to draw the following conclusions:

- the type of inhibitor used (up to 2 times), affects the degree of corrosion damage. It is possible that other types of inhibitors, in contrast to those used, can improve this indicator;

- the degree of corrosion damage (up to 3 times) depends on the intensity of DPD, carried out by changing the brushes feed and has pronounced minimum;
- dilution of the inhibitor leads to decrease in its protective properties by several times. The best result is achieved when using the inhibitor at 100% concentration;
- the depth of surface defects increases the degree of corrosion damage. The initial surface preparation during descaling and the optimal value of DPD should be carried out almost simultaneously;
- the result shown in Fig. 4 indicates a certain optimum degree of brush feed, i.e. pressed against the surface of the pipe. Increase in feed leads to increase in the depth of surface defects and, as a result, deterioration in the corrosion properties of the surface;
- the presented dispersion field of the average size of ferrite grains on the surface indicates the absence of relationship with the degree of corrosion. At the same time, the trend line suggests possible decrease in corrosion damage with decrease in grain size.

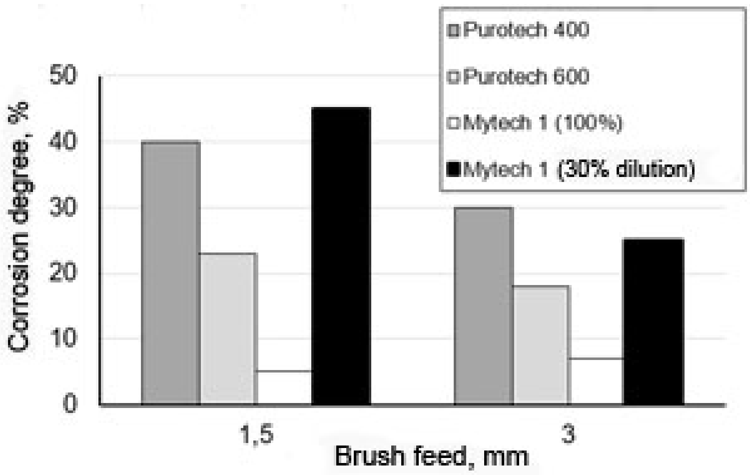


Figure 1 - Effect of the inhibitor type and brush feed on corrosion degree

Author's development

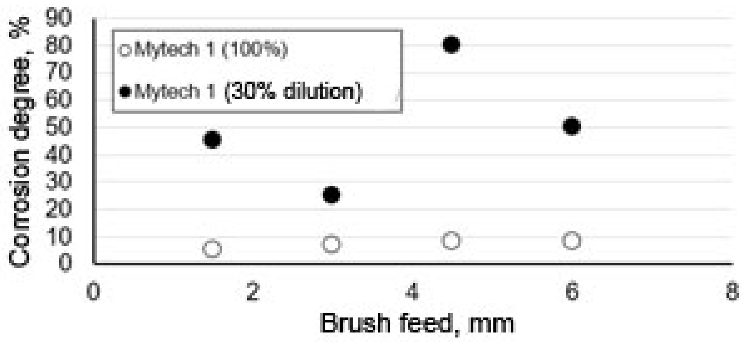


Figure 2 - Dependence of the degree of damage to corrosion on brushes feed and the degree of inhibitor dilution

Author's development

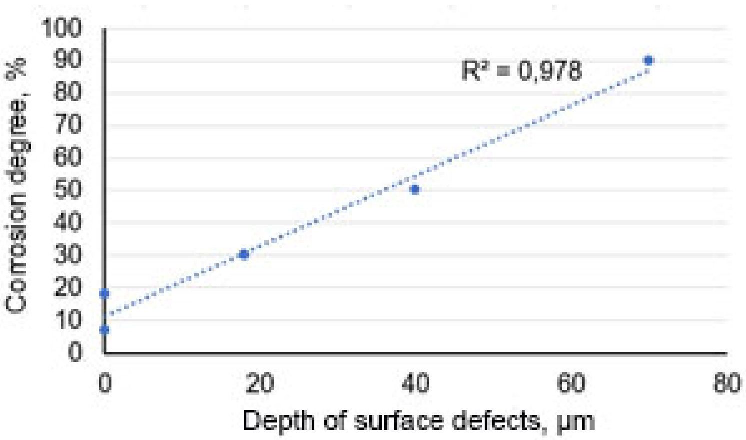


Figure 3 - Dependence of the degree of corrosion damage on the depth of surface defects

Author's development

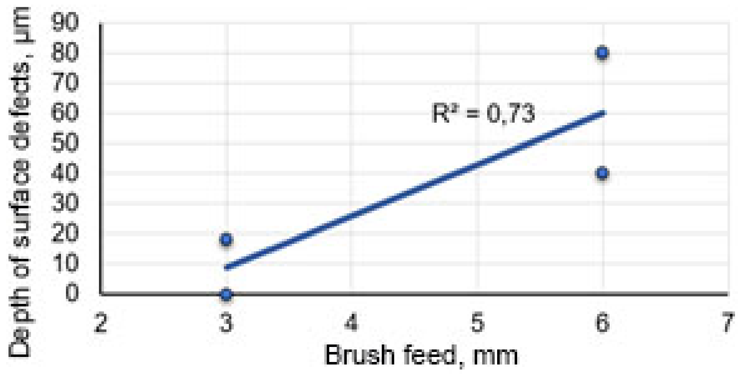


Figure 4 - Dependence of the depth of defects on the feed of brushes and processing time (15 s action)

Author's development

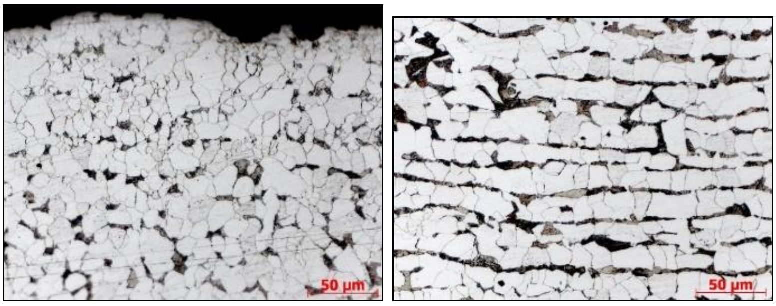
Study of the structure of pipes subjected to surface plastic deformation with inhibitors. Based on the results of corrosion tests, 6 samples of hot-rolled pipes were selected according to the results for each type of inhibitor.

The structure of all presented samples is identical and is characterized by an elongated ferrite-pearlite structure. In this case, the ferrite grains are equiaxed, and the pearlite grains are elongated (banding score on the inside of pipes is 4, according to GOST 5640). Herewith, decrease in banding of the structure is observed with approaching the outer surface of the pipe. Closer to the outer surface, the degree of pearlite grains deformation decreases, and at ½ of the wall thickness an equiaxed ferrite-pearlite structure is observed.

The outer surface of the pipe is somewhat decarburized and consists of extremely fine grains of ferrite.

It can be assumed that after cooling immediately after hot deformation, the pipes or pipe fragments were subjected to thermal action from the outside, which led to development of primary and collective recrystallization processes.

A typical cross-sectional structure of samples is shown in Figure 5.

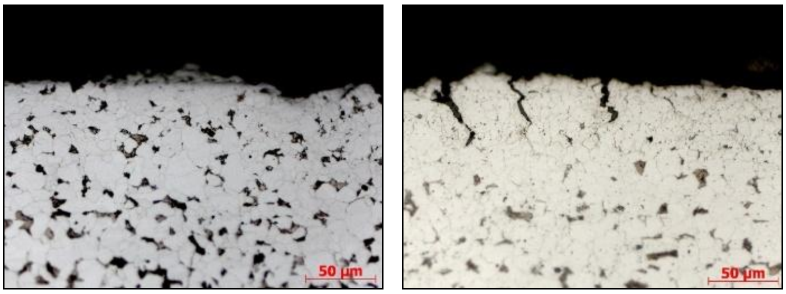


a b

Figure 5 - Structure of hot-rolled pipes: ***a*** *– outer surface;* ***b*** *– cross-section structure close to the inside surface*

Author's development

The depth of defects on the outer surface of the pipe reaches 65-75 µm. The average grain size on the surface (decarburized layer) is 5.4 µm (Figure 6).



a b

Figure 6 - Structure of the outer surface of hot-rolled pipes: ***a*** *– sample 4;* ***b*** *– sample 5*

Author's development

The results of corrosion resistance tests and studies of the metallographic structure of pipe samples are presented in Table 1.

Table 1 - Corrosion resistance and average grain size depending on the type of inhibitor and processing mode

Sample No.	Inhibitor type	Feed, mm/ Processing time, sec	Depth of surface defects, µm	Average grain size, µm	Corrosion damage degree, %
1	Without inhibitor	2/30		5.4	95
2	Purotech 400	3/15	65 ÷75	7.2	30
3	Purotech 600	3/15		7.8	18
4	M1	3/15		4.3	5
5	M1	6/15		3.8	8
6	M1 (30%)	6/15		5.2	50

Author's development

Steel 20 is not a corrosion resistant material, therefore the effect of using a corrosion inhibitor can be seen most clearly.

The use of brush deformation in the specified ranges will obviously not affect the microstructure of the material. This treatment creates residual compressive stresses on the surface that improve the fracture potential. As shown by the results given in the table, the use of brushes contributed to deeper penetration of the inhibitory reagent to the juvenile steel surface. Intensive brushing may have a reverse defect – the corrosion rate, as well as the depth of damage to the material, may increase. Presumably, this is due to the fact that having a sufficiently low strength, steel is deformed, with formation of the smallest surface defects. It is these small areas that turned out to be with a high level of stress and structural defects. As is known, the high density of dislocations and the level of stress contribute to development of oxidation processes in oxidizing and corrosive environments.

However, the use of brush surface treatment in some power and time intervals contributes to increase in corrosion resistance. The effect of brushing will be especially noticeable when processing hot-rolled material. Since hot-rolled steel is always covered with a small layer of scale (the thickness and composition of the scale varies depending on the deformation temperature, the initial material, the rate and degree of deformation, and subsequent processing). Application of an inhibitory layer to areas with scale without the use of additional treatment or the effect increasing the inhibitor penetration into the metal surface, in fact, leaves the so-called "white spots", i.e. such scale microsites, which, when the material enters the corrosive medium, are primarily the channels for penetration of the destructive medium deep into the material to the juvenile surface, which is direct consequence of pitting corrosion appearance.

The use of brush processing of the material does not completely remove the scale microlayer, it is like a kind of "stuffing" of the inhibitor on the surface of the material. It allows to fill all voids in the scale layer with an inhibitor, since scale is a fairly loose product by its nature.

Attention should be paid to the negative result of brushing. Decrease in the positive impact of the effect can also be considered in terms of reducing the surface roughness. Decrease in surface roughness not only improves mechanical properties, such as fatigue characteristics, but also, according to [22], increases corrosion resistance.

And at the same time, later studies [23] showed that the quality of the surface in relation to its corrosion resistance is not stable. Roughness removal does improve corrosion resistance, however, polished specimens tend to show lower resistance results compared to the original condition. Therefore, in the future, it is necessary to conduct additional studies on the dependence of duration, frequency and intensity of brushing, taking into account the nature of the material, mechanical properties, surface quality and other factors, the degree of steel aging.

Based on the study, it can be assumed that the use of brush-packed application of an inhibitor on a clean, cold-rolled metal surface may not have such pronounced results, since there is no scale.

The study showed that there are prospects for further research in this area.

The use of brushing allows the surface treatment to be carried out simultaneously with application of the inhibitor.

Brush processing allows increasing the level of compressive stresses, which, to a certain level, contribute to increase in corrosion resistance. In addition, in the process of vibration processing, particles of Fe_3O_4, Fe_2O_3, FeO, formed on the surface under the influence of high temperatures during production of rolled products, are removed.

1. The low corrosion resistance of transport lines made of hot-rolled pipes is associated with the technological features of their production – the presence of scale on the surfaces and coarse-grained decarburized layers on the surfaces. Improvement in performance can be achieved by careful removal of scale, surface plastic deformation combined with inhibitor treatment of the pipe surface, the penetration depth of the inhibitor can be significantly increased if the preparation reaches a juvenile (scale-free) metal surface.

2. Surface-plastic deformation, which refines the grain structure, creates conditions for formation of "penetration channels" of the inhibitor in solution to the elements of the crystal structure of the material

3. The use of surface dynamic deformation in combination with an inhibitor makes it possible to increase the corrosion resistance of pipes by 11...19 times.

4. The type of inhibitor used significantly affects the corrosion resistance (up to 8 times with these types).

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Abstract. ***Ціль.*** *Обґрунтування застосування поверхневої пластичної та динамічної поверхневої деформацій (ППД, ДПД) суміщених з антикорозійною інгібіторною обробкою сталевих вуглецевих труб загального призначення для підвищення зносостійкості трубних магістралей.*

Методика. *Зразки гаряче деформованої сталі, відібрані від тестових фрагментів труб, були схильні до різних режимів поверхневої пластичної та динамічної поверхневої деформацій з використанням інгібіторів корозії. Пластичну деформацію здійснювали щітковим інструментом з різною величиною притискання щіток до труби та часу обробки. Дослідження захисної здатності інгібіторів проводили прискореним методом під час періодичної конденсації вологи ДСТУ ISO 6270-2:2015. Проведено порівняльне металографічне дослідження структури металу труб, підданих ППД та ДПД з інгібіторами.*

Результати. *Представлені результати експериментальних досліджень поверхневої корозійної стійкості та металографічної структури труб 139х5 мм із сталі марки 20 при використанні трьох типів інгібіторів та різних режимів динамічної пластичної деформації.*

Наукова новизна. *Динамічна поверхнева деформація металу, що подрібнює зеренну структуру, створює умови для утворення "каналів проникнення" інгібітора в структуру металу. Застосування ДПД дозволяє зменшити розмір зерен у поверхневому шарі, що дозволяє припустити можливість управління глибинним проникненням інгібітора і, як наслідок, корозійної стійкості труб залежно від експлуатаційних вимог.*

Практичне значення. *Встановлено, що застосування динамічної поверхневої деформації у комплексі з певним інгібітором дозволяє збільшити корозійну стійкість у 11 – 19 разів.*

Ключові слова: *гарячекатана труба, міжкристалітна корозія, піттингова корозія, інгібітор, пластична деформація, канал проникнення, мікроструктура*

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