

ЕКОЛОГІЯ ТА РЕСУРСОЗБЕРЕЖЕННЯ

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DEPENDENCE OF CORROSION ACTIVITY OF AQUATIC- PETROLEUM MIXTURES ON CHARACTERISTICS OF AQUATIC ENVIRONMENTS

The processes of metal corrosion in water-oil mixtures depending on the level of water mineralization, reaction of the environment, and ratio between the volumes of oil and aqueous solutions at temperatures of 20 °C and 80 °C were studied. It is shown that the corrosiveness of the medium against metals becomes higher with increasing level of water mineralization and acidic content with decreasing pH of the medium and increasing temperature. The corrosion rate of metals is significantly reduced in the presence of oil. Increase in the oil content in water-oil mixtures has little effect on the corrosion rate of metals. Increase in the hardness of water in the presence of acetic acid does not reduce the corrosion rate of metals and, in some cases, there is an increase in the corrosion activity of water with increasing hardness.

Keywords: oil, mineralization, corrosion of metals, corrosion rate, inhibitors.

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Formulation of the problem. Despite significant progress in the development of new materials characterized by high strength, reliability, corrosion resistance, the use of non-alloy metals in the mining industry remains quite common, as well as in the construction of water, gas and oil pipelines. Even more common is the use of steel structures in existing enterprises, mines, wells, local pipelines. Today, in 40–70 % of cases, premature destruction of metal structures due to corrosion. These phenomena are often the cause of man-made accidents and catastrophes that cause significant economic and environmental damage. The problem is especially acute in the oil and gas and petrochemical industries, where corrosion poses a great danger to equipment, tanks, pipelines in the systems of extraction, transportation and refining of oil and gas. Corrosion of metals during oil and gas production in the early stages of their processing is dangerous. Both reservoir and separation reservoir waters and crude oil are corrosive. It is believed that the main reason for the high corrosion activity of water-oil mixtures is the presence of various sulfur compounds in the aqueous phase and a high level of mineralization. But little effect has been studied on the corrosion activity of these mixtures, the reactions of their environment, which are often weakly acidic due to various reasons. However, it is not known whether it is always possible to reduce the harmfulness of corrosion by simple neutralization. Therefore, to solve the problems of protection of metals from corrosion, it is important to determine the influence of various factors on the corrosion activity of water-oil mixtures.

Analysis of previous research. It is known that water-oil mixtures formed in the processes of oil extraction and primary refining are quite complex [1] and are characterized by increased corrosion activity due to the presence of sulfur compounds [2]. These media are corrosive not only in the extraction, but also in the transportation, storage and refining of oil [3, 4]. Corrosion aggressiveness in water-oil environments largely depends on the ratio of oil and water, mineralization of the water fraction [5]. Modern approaches to the use of inhibitors to protect metals from corrosion are based on determining the dependence of corrosion activity of media on their composition [6, 7]. One of the main factors of high corrosion activity of water-oil mixtures is the high content of salts in mine waters formed in oil production processes [8, 9]. During oil transportation, the corrosive activity of the environment is due to the high content of mineralized water (10–30 %) and temperature (~ 70–80 °C) [10, 11]. However, the data in the literature do

not fully analyze the impact of individual factors and their impact on the corrosion activity of the environment as a whole.

The aim of this work was to determine the effect of water mineralization, pH, temperature, ratio of water to oil, aeration conditions on the corrosion activity of water-oil mixtures.

Methods of work. Metal samples from steel St 3, steel St 20, brass L-62, copper M-2 were used in the work. The corrosion rate was measured by massometry [12]. Distilled and tap water were used as the corrosive medium. Characteristics of tap water: hardness 4.7–5.1 mg-eq/dm³, calcium concentration 3.8–4.0 mg-eq/dm³, alkalinity 4.6–5.0 mg-eq/dm³, pH 7.1–7.8, chloride concentration 35–75 mg/dm³, sulfate concentration 23–65 mg/dm³. Sodium chloride and sulfate solutions with a concentration of 1 to 100 g/dm³, aqueous solutions of acetic acid with a concentration of 0.5 to 12.5 mg/dm³, solutions of sodium sulfite and metabisulfite with concentrations of 1 to 10 mg/dm³ were used as model solutions.

During corrosion tests, metal samples were in an aqueous medium in the presence of oil and without its use. The corrosion test period was 96–150 hours at 20 °C and 6 hours at 80 °C. The volume to water ratio ranged from 140 cm³/10cm³ to 180cm³/5cm³. The corrosion rate was calculated by reducing the mass of metal samples according to the formula:

$$\frac{(m_1 - m_2)}{S \cdot \tau}, \text{ g/m}^2 \cdot \text{h} \quad (1)$$

where m_1 – mass of the sample before the test, g;

m_2 – mass of the sample after the test, g;

S – sample area, m²;

τ – test duration, h.

Presenting main material. It is known that the corrosive activity of aqueous solutions of mineral salts against metals increases with increasing concentration [12]. For sodium chloride, the highest corrosion aggressiveness is observed in 3.0 % solution. This is confirmed by the results shown in Figure 1. In this case, solutions of chloride and sodium sulfate were used.

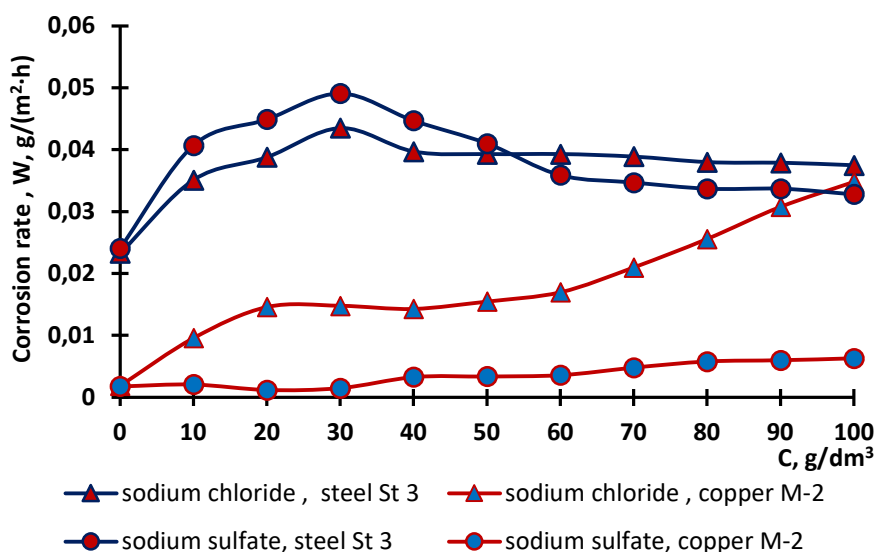


Fig. 1 – Dependence of the corrosion rate of steel ST 3 and copper M-2 on the concentration of chloride and sodium sulfate at a temperature of 20 °C

As can be seen in the figure, the corrosion rate of steel St 3 in the concentration range from 10 to 100 g/dm³ varies little. The same dependence was observed for sodium sulfate solutions. Although at a concentration of 30 g/dm³ the corrosion rate of steel is slightly higher than at other concentrations, both for sodium chloride solution and for sodium

sulfate solution. The decrease in the corrosion rate of metals at salt concentrations higher than 30 g/dm³ is explained by a decrease in oxygen solubility with increasing mineralization of solutions.

Another dependence is observed when using copper samples. As can be seen from Fig. 1, the corrosion rate of copper increases with increasing sodium chloride concentration. If in the case of steel the corrosion rate in desalinated water reached 0.0233 g/(m²·h) and at a NaCl concentration of 100 g/dm³ increased to only 0.0375 g/(m²·h), then in the case of copper M-2 in the same concentration range sodium chloride corrosion rate increased 18 times – from 0.0019g/(m²·h) to 0.0348 g/(m²·h). That is, in a 10 % solution of sodium chloride, the corrosion rate of copper M-2 is the same as that of steel St 3, although in distilled water it was 12 times lower. This effect is due to the fact that with increasing salt concentration increases the electrical conductivity of solutions, which leads to a significant increase in the distances between the cathode and anode zones on the metal surface, which generally prevents the formation of a dense protective oxide film on its surface. This is of great importance for copper because its corrosion resistance is largely due to the strength of the oxide film. It is obvious that the use of copper products and its alloys in mineralized aquatic environments is impractical. In general, given the high corrosion activity of 3 % sodium chloride solution, it was mainly used in subsequent experiments.

When studying the chemical characteristics of formation waters, it was noted that at a high level of their mineralization, waters with a weakly acidic environment were often found (5<pH<6). It is possible that in addition to mineral acids, they may have been present and organic acids. The effect of organic acids on the corrosion activity of distilled, tap water and 3 % sodium chloride solution can be assessed by the results shown in table 1. When increasing the concentration of acetic acid in these media from 0.0 g/dm³ to 5.0 g/dm³ there was a decrease in pH – in distilled water from 6.28 to 3.15, in tap water from 7.0 to 3.09, and in 3 % sodium chloride solution from 6.25 to 2.84. In all cases, there was a significant increase in the corrosion rate of steel St 3. In distilled water at 20 °C, the corrosion rate increased by almost an order of magnitude, and at pH 3.15 it reached 0.2036 g/(m²·h).

Table 1 – The dependence of the corrosion rate of steel St 3 on the concentration of acetic acid and pH of the medium when tested in distilled water (I), tap water (II) and 3 % sodium chloride solution (III)

№	[CH ₃ C(O)OH], g/dm ³	pH			T=20 °C			T=80 °C		
		I	II	III	W, g/(m ² ·h)					
1	0.0	6.28	7.01	6.25	0.0286	0.0459	0.0567	0.8471	1.0237	0.7920
2	0.5	4.46	3.46	3.35	0.0445	0.0772	0.0882	1.6272	1.8912	3.0496
3	2.5	3.70	3.22	2.98	0.1313	0.0992	0.1039	3.1034	2.6994	3.6530
4	5.0	3.15	3.09	2.84	0.2036	0.1568	0.1245	3.7985	3.4860	4.5690

Surprisingly, with increasing concentration of acetic acid, the rate of corrosion in sodium chloride solution increased the least, and at pH 2.84 it reached 0.1245 g/(m²·h), although at pH 6.25 it was twice as high as in distilled water. At a temperature of 80 °C, the corrosion rate increased the most in 3 % sodium chloride solution from 0.7920g/(m²·h) to 4.5690 g/(m²·h), which is more than 4 mm/year. It is clear that at 80 °C in distilled water at pH 6.28 there was an acceleration of the corrosion rate, mainly due to the increase in the rate of the oxidation reaction of iron with oxygen. At pH ~ 6, oxygen is the main depolarizer in the oxidation of iron. At pH ~ 3 significantly increases the rate of oxidation of iron with the release of hydrogen. At a temperature of 80 °C, probably in all environments, the corrosion rate increases due to hydrogen depolarization. Although corrosion processes with oxygen depolarization still play a significant role. After all, the concentration of oxygen in water at 80 °C reaches 5.29 mg/dm³. This is enough for the oxidation of the metal. Especially when you consider that when the temperature rises by 15 °C, the rate of the chemical reaction doubles. In this case, with increasing temperature from 20 °C to 80 °C, the corrosion rate at pH 6–7 increases 29.6 times, in tap water 22.3 times, and in 3 % NaCl solution 14.0 times.

Obviously, the solubility of oxygen is most reduced in the salt solution, so here the increase in corrosion rate is the smallest. In a weakly acidic environment, when hydrogen depolarization makes a significant contribution to the corrosion process, and the pH values in NaCl solution were the lowest, the increase in the corrosion rate was the highest – 36.7 times. For tap water this figure reached 22.3 times, and in distilled water 18.7 times. Obviously, in the latter two cases, the process of oxidation of iron with oxygen plays a significant role in the acidification of water. Therefore, the increase in the rate of the corrosion process is slightly lower than the calculated one – 32 times, when the oxygen concentration decreased from 9.17 mg/dm³ at 20 °C to 5.29 mg/dm³ at 80 °C. It should be noted that in a solution of sodium chloride, where the electrical conductivity is highest, corrosion processes are faster due to the separation of the anode and cathode zones, regardless of the nature of the depolarizer – oxygen or hydrogen.

The effect of oil on the corrosion processes of steel St 3 in the water mixture can be judged by the results shown in tables 2 and 3.

Table 2 – The dependence of the corrosion rate of steel St 3 in 3 % sodium chloride solution on the concentration of acetic acid, pH and the ratio of the volume of aqueous solution (V_w) and oil (V_o) at 20 °C

№	$V_w, \text{ cm}^3$	$V_o, \text{ cm}^3$	$[\text{CH}_3\text{C}(\text{O})\text{OH}], \text{ g/dm}^3$	pH	$W, \text{ g}/(\text{m}^2 \cdot \text{h})$
1	150	0	–	6.25	0.0567
2	140	10	–	6.25	0.0123
3	150	0	0.50	3.33	0.0882
4	140	10	0.50	3.33	0.0206
5	150	0	2.50	2.98	0.1039
6	140	10	2.50	2.98	0.0394
7	150	0	5.00	2.84	0.1245
8	140	10	5.00	2.84	0.0488
9	140	10	6.25	2.82	0.1184
10	140	10	12.50	2.71	0.1246
11	140	10	13.50	2.63	0.1973

Table 3 – The effect of acetic acid concentration, pH, ratio of aqueous solution (V_w) and oil (V_o) on the rate of corrosion of steel St 3 in 3 % NaCl solution at 80 °C

№	$V_w, \text{ cm}^3$	$V_o, \text{ cm}^3$	$[\text{CH}_3\text{C}(\text{O})\text{OH}], \text{ g/dm}^3$	pH	$W, \text{ g}/(\text{m}^2 \cdot \text{h})$
1	150	0	–	6.41	0.7920
2	140	10	–	6.41	0.2497
3	150	0	0.50	3.35	3.0496
4	140	10	0.50	3.35	1.7457
5	150	0	2.50	2.96	3.6530
6	140	10	2.50	2.96	1.7565
7	150	0	5.00	2.81	4.5690
8	140	10	5.00	2.81	2.3707

As can be seen from table 2, when adding to 140 cm³ of 3 % NaCl solution 10 cm³ of oil at pH=6.25, the corrosion rate of steel at a temperature of 20 °C is reduced by 4.6 times. This is due to the deterioration of the diffusion of oxygen into the aqueous medium, as well as the possible formation of a protective film of oil on the metal surface. When lowering the pH to 3.33; 2.98 and 2.84 by increasing the doses of acetic acid to 0.5; 2.5; and 5.0 g/dm³, the corrosion rate of steel in the presence of oil, compared to aqueous solution without oil, decreased by 4.2, respectively; 2.63; 2.55 times. This suggests that as the pH decreased, the oxygen depolarization process played an increasingly smaller role, while the effect of hydrogen depolarization increased.

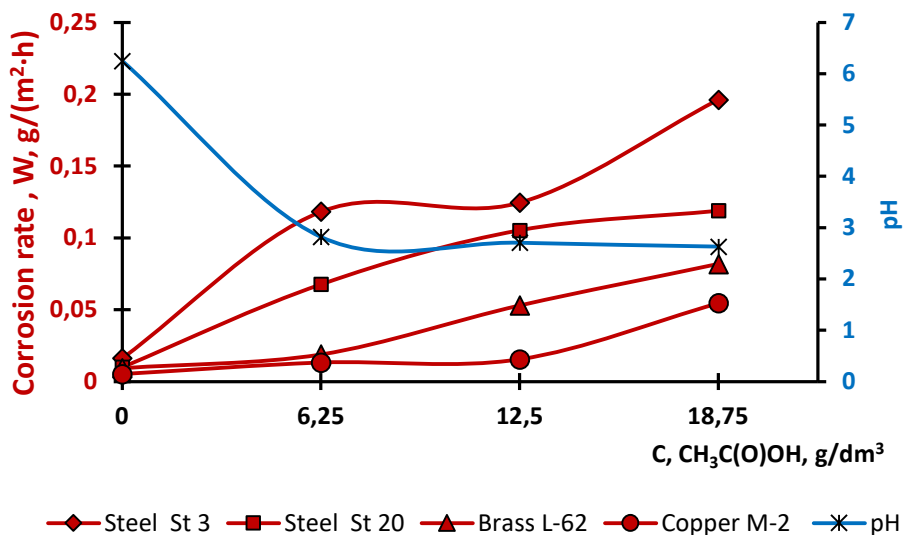
It should be noted that at the concentration of acetic acid over 6 g/dm³ (pH<2.82) the corrosion rate of steel increased significantly and reached 0.1184–0.1973 g/m²·h. This indicates that in this case the corrosion is mainly due to hydrogen depolarization.

The addition of oil to a 3 % NaCl solution significantly affected the corrosion rate of steel St 3 and at a temperature of 80 °C. The concentration of acetic acid and the pH of the medium had a significant effect on the corrosion rate. In a neutral environment, the addition of oil reduces the corrosion rate by 3.2 times, and when the water is acidified with acetic acid and when oil is added, the corrosion rate is reduced by 1.7–2.0 times. This indicates an increase in the effect of hydrogen depolarization on steel corrosion under these conditions. As can be seen from Figure 2, with the acidification of a solution of sodium chloride in the water-oil mixture, the corrosion rate increases not only in steel St 3 but also in other metals. The corrosion rate of brass and copper is also significant. This is due to the high electrical conductivity of solutions and their significant acidity.

The significant effect of hydrogen depolarization on the process of metal corrosion (steel St 3) in the water-oil mixture is evidenced by the results shown in table 4. In this case, sodium metabisulfite was added to 3 % NaCl solution at a concentration of 1–10 g/dm³. The pH of the aqueous medium decreased from 6.25 to 3.84. There was a film of

oil on the surface of the water, and in experiments 2–6 the samples with the water-oil mixture were hermetically sealed. Under these conditions, the diffusion of oxygen into water was impossible. The corrosion rate mainly depended on hydrogen depolarization and increased from 0.0222 g/(m²·h) at pH 6.25 to 0.7039 at pH 3.84. It is interesting to note that the addition of only 5 cm³ of oil to 180 cm³ of aqueous solution provided a significant reduction in the diffusion of oxygen into the water. The corrosion rate in this case reached 0.0272 g/(m²·h) (experiment 1). In the hermetically sealed volume, it was almost the same and reached 0.0222 g/(m²·h).

Fig. 2 – Change in pH of 3% NaCl solution and corrosion rate of steel St 3, steel St 20, brass L-62 and



copper M-2 with increasing concentration of acetic acid at a temperature of 20 °C at a ratio of aqueous solution and oil 140 : 10

Table 4 – The dependence of the corrosion rate of steel St 3 on the concentration of sodium metabisulfite in 3 % NaCl solution and the ratio of the volume of aqueous solution (V_w) and oil (V_o) at a temperature of 20 °C in hermetically sealed beakers

№ п/п	V _w , cm ³	V _o , cm ³	[Na ₂ S ₂ O ₅], g/dm ³	pH	W, g/(m ² ·h)
1	180	5	-	6.250	0.0272*
2	180	5	-	6.250	0.0222
3	180	5	1	4.468	0.0647
4	180	5	3	4.006	0.2523
5	180	5	5	3.853	0.3993
6	180	5	10	3.838	0.7039

Note: * - the container is opened

The effect of oxygen in these conditions on the corrosion of steel was minimal. It is known that sodium bisulfite, like sodium sulfite, is an effective reagent for deoxidation of water [13]. In addition, in [14] it was reported that sodium sulfite significantly reduces the rate of corrosion of metals in mineralized media, even without the addition of oil. The results shown in Fig. 3, confirm this information.

As shown in Figure 3, the corrosion rate of steel, brass and copper is significantly reduced by the addition of sodium sulfite. Therefore, without a doubt, under anaerobic conditions in the technological processes of oil production, transportation and refining, corrosion processes are caused mainly by hydrogen depolarization. Based on this, measures should be identified to protect metals from corrosion.

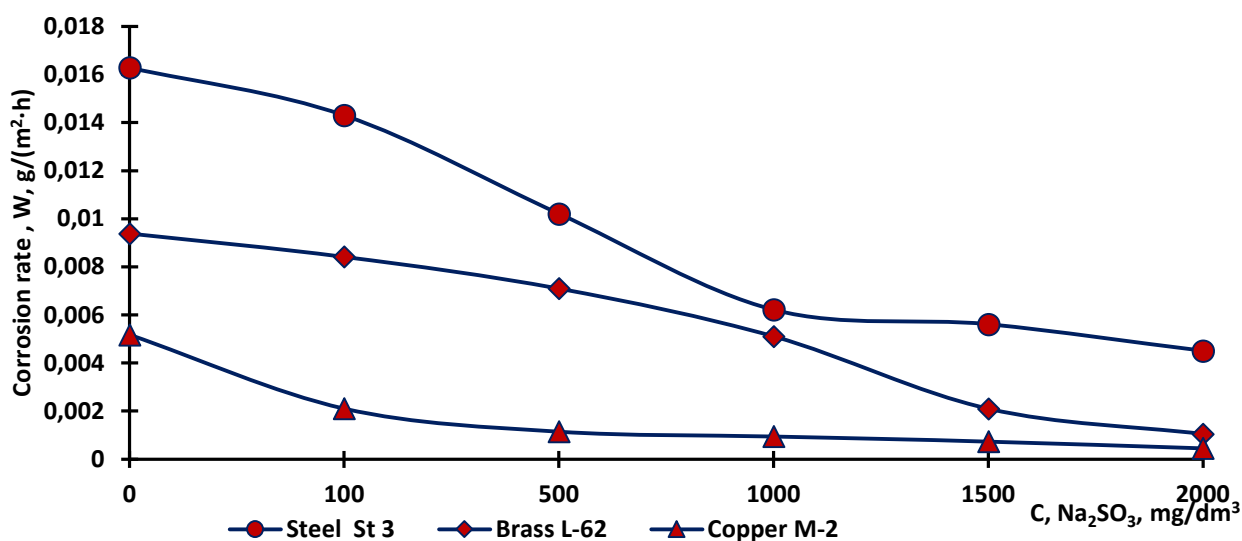


Fig. 3 – Dependence of corrosion rate of metals of steel ST 3, brass L-62 and copper M-2 on the concentration of sodium sulfite in 3% NaCl solution at temperature 20 °C

Conclusions. As a result of research on the study of corrosion of metals in water-oil mixtures depending on the level of water mineralization, the reaction of the environment, the ratio of volumes of oil and aqueous solutions at different temperatures:

- it is shown that with increasing NaCl concentrations up to 100 g/dm³ in neutral aqueous medium the corrosion rate of steel St 3 increases 1.8 times, and copper 18 times, due to increasing electrical conductivity and destruction of protective oxide film on metal surfaces.
- it was found that when acidifying water and aqueous solution of sodium chloride at a temperature of 20 °C, the corrosion rate of steel St 3 increases by 2.5–4.2 times. At a temperature of 80 °C the corrosion rate increases to 0.7920–1.0237 g/(m²·h) in a neutral environment, and at acidification to pH 2.84–3.15 the corrosion rate reaches 3.4860–4.5692 g/(m²·h).
- it was found that when using mixtures of 3 % sodium chloride solution at a temperature of 20 °C, the corrosion rate of steel St 3 at pH 6.25 reaches only 0.0123 g/(m²·h), and at pH 2.84–0.0488 g/(m²·h). At a temperature of 80 °C, these indicators at the appropriate pH were 0.2497 and 2.3707 g/(m²·h). Significant reduction of corrosion with the addition of oil due to reduced solubility of oxygen in water and hydrophobization of the steel surface. A similar effect of the pH of the medium and the addition of oil was observed on the corrosion processes of steel St 20, brass L-62 and copper M-2.
- the influence of sodium sulfite and metabisulfite on the corrosion of metals in aqueous media is determined.

It is shown that in a neutral environment the corrosion rate is reduced by deoxidation of water. When water is acidified, the corrosion rate increases significantly, even with deep deoxidation of water.

Prospects for further research. Further studies will examine the effect of inhibitors on the corrosion of metals in aqueous-petroleum mixtures in both aqueous media in the presence of oil and in petroleum media in the presence of aqueous solutions of salts with different pH values.

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ЗАЛЕЖНІСТЬ КОРОЗІЙНОЇ АКТИВНОСТІ ВОДНО-НАФТОВИХ СУМІШЕЙ ВІД ХАРАКТЕРИСТИК ВОДНИХ СЕРЕДОВИЩ

В роботі було досліджено процеси корозії металів у водно-нафтових сумішах в залежності від рівня мінералізації води, реакції середовища, співвідношення об'ємів нафти та водних розчинів за температур 20 °С та 80 °С. Показано, що агресивність середовища щодо металів зростає із підвищенням рівня мінералізації води, вмісту кислих реагентів при зниженні рН середовища та підвищенні температури. При цьому швидкість корозії металів суттєво знижується у присутності нафти. Підвищення вмісту нафти у водно-нафтових сумішах мало впливає на швидкість корозії металів. Збільшення жорсткості води у

присутності оцтової кислоти не знижує швидкість корозії металів, а в окремих випадках відмічено підвищення корозійної активності води із зростанням її жорсткості.

Ключові слова: *нафта, мінералізація, корозія металів, швидкість корозії, інгібітори.*

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