
Effect of nitrogen-containing inhibitors on the corrosion inhibition of low-carbon steel in solutions of mineral acids with various anionic compositions*

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Abstract

The corrosion of low-carbon steel in 2 M HCl, 2 M H_2SO_4 and 2 M H_3PO_4 (25°C) inhibited by a triazole derivative – IFKhAN-92 – and quaternary ammonium salts was studied by the mass loss of metal samples and by voltammetry of the steel electrode. The inhibition of electrode reactions on steel in these environments is due to the formation of polymolecular protective layers by the molecules of these nitrogen-containing compounds on the steel surface. This is a versatile mechanism of inhibitor action since it slows down the electrode reactions on steel, regardless of the acid composition of the corrosive environment. The strongest hindrance of electrode reactions on steel in all the environments studied and, hence, the strongest corrosion inhibition is provided by the IFKhAN-92 additive. This effect results from the ability of the molecules of this compound to form protective layers whose molecules are interconnected with each other in the layer bulk and with the surface of the metal being protected by chemical interactions owing to the presence of a triazole ring in their structure.

Keywords: *acid corrosion, corrosion inhibitors, triazole, quaternary ammonium salts, low carbon steel, hydrochloric acid, sulfuric acid, phosphoric acid.*

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Introduction

Important areas of industrial use of mineral acid solutions include the metallurgical industry, the oil and gas complex, as well as housing and communal services [1–9]. Acid solutions are used to treat oil- and gas-bearing strata in order to increase the yield of hydrocarbons, remove thermal scale from the surface of steel items, and clean the inner surfaces of steel equipment from mineral deposits and corrosion products. During operation, these corrosive

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solutions come into contact with the steel surfaces of process structures and metal items. If no special protective measures are used, this can lead to significant corrosion damage. The use of corrosion inhibitors (CIs) is an simple and popular method for steel protection in such process environments.

Organic nitrogen-containing compounds are the most popular industrial CIs for these environments as they hinder the corrosion of various steels in solutions of HCl [10–20], H_2SO_4 [21–25], H_3PO_4 [26–28], and HNO_3 [29]. Among such CIs, quaternary ammonium salts (QAS) deserve attention because many of them can slow down the corrosion of steels considerably [30–40]. Recently, organic compounds containing two or more quaternized nitrogen atoms in their structures have been studied extensively [41–47]. The protective effect of QAS can be enhanced by combining them with some anionic and molecular additives [48–51], which makes them a promising basis for creating mixed CIs. Triazole derivatives that have been extensively studied in recent decades [52–68] may become alternatives to QAS as CIs for steels in acid environments.

The corrosion of iron and low-carbon steels in solutions of mineral acids (so-called "non-oxidizing agents") is generally described by the overall reaction:

$$2H^+ + Fe = Fe^{2+} + H_2\,, \tag{1}$$

which is the result of four partial reactions, namely [69]:

anodic iron dissolution

$$Fe - 2e = Fe^{2+}\,, \tag{2}$$

cathodic iron deposition

$$Fe^{2+} + 2e = Fe\,, \tag{3}$$

cathodic hydrogen evolution

$$2H^+ + 2e = H_2\,, \tag{4}$$

and its anodic ionization

$$H_2 - 2e = 2H^+\,. \tag{5}$$

Corrosion predominantly results from reactions (2) and (4).

Cathodic hydrogen evolution (4) consists of several successive stages: H^+ delivery from the acid bulk to the metal surface (where $(H^+)_*$ is a hydrogen ion located at the closest distance from the metal surface)

$$H^+ \rightarrow (H^+)_*\,, \tag{6}$$

and charge transfer reaction (Volmer reaction)

$$(H^+)_* + e \rightarrow H_{ad}\,. \tag{7}$$

It is followed by a chemical stage (Tafel reaction)

$$H_{ad} + H_{ad} \rightarrow (H_2)_{ad} \tag{8}$$

or electrochemical recombination (Heyrovsky reaction)

$$H_{ad} + (H^+)_* + e \rightarrow (H_2)_{ad} \tag{9}$$

Several possible pathways of partial reaction (4) on metals are considered [70]. One of these involves slow discharge (7) and a fast stage of chemical recombination (8) or electrochemical recombination (9). The calculated Tafel slope (b_c) of cathodic polarization is 0.118 V.

The second pathway involves the limitation of the chemical recombination reaction (8) with a preceding fast discharge reaction (7) and is characterized by the calculated value of b_c=0.029 V.

In the third pathway, the limiting stage involves the electrochemical recombination (9) preceded by the fast discharge reaction (7) (b_c=0.040 V). Taking into account the inhomogeneity of the metal surface for the latter two processes, the values of b_c=0.11–0.12 V can be obtained. One can see that, most often, the value of b_c does not allow one to judge on the mechanism of hydrogen evolution. Analysis of the dependence of kinetic parameters of the cathodic reaction on the pH of the environment makes it possible to identify experimentally whether reaction (4) is limited by the discharge stage or recombination stage.

In HCl solutions, the evolution of hydrogen on iron and steels occurs at comparable rates of the discharge and recombination stages. In acid sulfate environments, the parameters of the electrode reaction are closer to those of a process limited by the discharge stage. The experimental values of b_c observed on an iron electrode in acid chloride and sulfate solutions are 0.115 and 0.110 V, respectively [70]. The limiting stage in acid phosphate solutions corresponds to recombination. The experimental value of b_c on Armco iron is 0.120 V and that on low-carbon steels is −0.130 V [71].

Thus, the mechanism of cathodic hydrogen evolution on iron or low carbon steel in a mineral acid solution depends on the chemical nature of the acid. The effect of the acid chemical nature on the stage of anodic ionization of iron (2), which occurs with joint participation of hydroxide anions and anions of acid residues, is more noticeable.

The reaction of iron (2) in HCl solutions is described using the Nobe–Chin mechanism [72]:

$$Fe + Cl^- + H_2O \leftrightarrow \left[FeCl(OH)\right]^-_{ads} + H^+ + e, \tag{10}$$

$$\left[FeCl(OH)\right]^-_{ads} \rightarrow \left[FeCl(OH)\right] + e, \tag{11}$$

$$\left[FeCl(OH)\right] + H^+ \rightarrow Fe^{2+} + Cl^- + H_2O, \tag{12}$$

where the calculated value of b_a is 0.070 V. In sulfuric acid environments, the Kolotyrkin–Florianovich mechanism is used [73]:

$$\mathrm{Fe} + \mathrm{H_2O} \leftrightarrow [\mathrm{FeOH}]^-_{\mathrm{ads}} + \mathrm{H}^+, \tag{13}$$

$$[\mathrm{FeOH}]^-_{\mathrm{ads}} \leftrightarrow [\mathrm{FeOH}]_{\mathrm{ads}} + \mathrm{e}, \tag{14}$$

$$[\mathrm{FeOH}]_{\mathrm{ads}} + \mathrm{HSO_4^-} \rightarrow \mathrm{FeSO_4} + \mathrm{H_2O} + \mathrm{e}, \tag{15}$$

or

$$[\mathrm{FeOH}]_{\mathrm{ads}} + \mathrm{SO_4^{2-}} \rightarrow \mathrm{FeSO_4} + \mathrm{OH^-} + \mathrm{e}, \tag{16}$$

$$\mathrm{FeSO_4} \leftrightarrow \mathrm{Fe^{2+}} + \mathrm{SO_4^{2-}}, \tag{17}$$

or

$$\mathrm{FeSO_4} + \mathrm{H_2O} \leftrightarrow \mathrm{FeHSO_4^+} + \mathrm{OH^-}. \tag{18}$$

The calculated b_a value is 0.035 V. In acid phosphate environments, Reshetnikov [71] considers two possible mechanisms of the anodic reaction. On Armco iron:

$$\mathrm{Fe} + \mathrm{H_2PO_4^-} \leftrightarrow [\mathrm{FeH_2PO_4}]_{\mathrm{ads}} + \mathrm{e}, \tag{19}$$

$$[\mathrm{FeH_2PO_4}]_{\mathrm{ads}} + \mathrm{H_2O} \leftrightarrow [\mathrm{FeOH}]_{\mathrm{ads}} + \mathrm{H_2PO_4^-} + \mathrm{H^+}, \tag{20}$$

$$[\mathrm{FeOH}]_{\mathrm{ads}} \rightarrow \mathrm{Fe^{2+}} + \mathrm{OH^-} + \mathrm{e}. \tag{21}$$

The observed b_a value is 0.035±0.005 V. On low carbon steels, a different mechanism is assumed in some cases:

$$\mathrm{Fe} + \mathrm{H_2PO_4^-} \leftrightarrow [\mathrm{FeH_2PO_4}]_{\mathrm{ads}} + \mathrm{e}, \tag{22}$$

$$[\mathrm{FeH_2PO_4}]_{\mathrm{ads}} + \mathrm{H_2O} \leftrightarrow [\mathrm{Fe(OH)(H_2PO_4)}]^-_{\mathrm{ads}} + \mathrm{H^+}, \tag{23}$$

$$[\mathrm{Fe(OH)(H_2PO_4)}]^-_{\mathrm{ads}} \rightarrow \mathrm{Fe^{2+}} + \mathrm{H_2PO_4^-} + \mathrm{OH^-} + \mathrm{e}, \tag{24}$$

where the observed b_a value is 0.040±0.005 V.

Despite the formal similarity of steel corrosion in different mineral acids, in each individual case there is a specific mechanism of partial electrode reactions, which can significantly affect the options available for inhibitory protection of the metal.

It seemed appropriate to perform a comparative study on the effect of nitrogen-containing compounds on the corrosion and electrode reactions of low carbon steel in solutions of mineral acids with various chemical compositions. We chose solutions of practically significant acids as corrosive environments, namely, HCl and H_2SO_4, as well as phosphoric acid that is promising for industrial use in many respects [26]. As nitrogen-containing CIs, we studied two products containing quaternary ammonium salts (benzyldimethyloctylammonium bromide and catamine AB), as well as a 3-substituted

1,2,4-triazole (IFKhAN-92 inhibitor). It should be noted that the goals of this study did not include the determination or clarification of the previously studied mechanisms of corrosion of steels in solutions of mineral acids with various chemical compositions. It was important for us to understand how significantly, other conditions being equal, the difference in the chemical composition of acid solutions can affect the protection of steel with CIs in these solutions.

Experimental

The corrosion rate of steel 3 (composition, mass%: 0.14–0.22 C; 0.15–0.33 Si; 0.40–0.65 Mn; up to 0.3 Cr; up to 0.30 Ni; up to 0.008 N; up to 0.30 Cu; up to 0.05 S; up to 0.04 P; up to 0.08 As; remainder Fe) in 2 M solutions of mineral acids was determined from the mass loss of samples (at least three samples per point) 50×20×3 mm in size. The amount of the acid solution was 50 ml per sample. Before an experiment, the samples were cleaned on an abrasive wheel (ISO 9001, grit 60) and degreased with acetone. The duration of the experiments was 2 hours. Corrosion tests were carried out in solutions with free access of air. Benzyldimethyloctylammonium bromide (BDMOA) and catamine AB, which is a mixture of alkylbenzyldimethylammonium chlorides ($[C_nH_{2n+1}N^+(CH_3)_2CH_2C_6H_5]Cl^-$, $n=10-18$), were used as the QAS. The IFKhAN-92 inhibitor (a derivative of 3-substituted 1,2,4-triazole) was also studied. The concentration of inhibitors in the solutions was 5 mM. Due to the low solubility of IFKhAN-92, it was introduced into mineral acids as a solution in ethanol, so that the ethanol concentration in the etching solution was 0.24 mol/L. The efficiency of inhibitors was estimated by the inhibition coefficients $\gamma=k_0/k_{in}$, where k_0 and k_{in} are the corrosion rates in the background solution and in the solution with the corresponding additive being studied.

Electrochemical measurements were carried out on a rotating disk electrode (460 rpm) made of steel 3, in 2 M solutions of mineral acids deaerated with hydrogen. The potentials of steel electrodes were measured against a saturated silver/silver chloride reference electrode and converted to the normal hydrogen scale. An electrode cleaned and degreased with acetone was kept in the test solution for 30 min, then the anodic and cathodic polarization curves were recorded using an EL-02.061 potentiostat at a dynamic potential scan rate of 0.5 mV/s. The effect of inhibitors on the electrode reactions of steel was studied in the range of $C_{in}=0.1-10$ mM.

All the studies were performed at a temperature of $t=25\pm2$°C.

Results and Discussion

Corrosion studies

According to the corrosion studies based on the mass loss of samples, the corrosivity of mineral acid solutions toward steel 3 decreases in the series: 2 M $H_2SO_4 > 2$ M $H_3PO_4 > 2$ M HCl (Table 1). The corrosion rate of steel 3 in H_2SO_4 and H_3PO_4 solutions are close to each other, while that in the HCl solution is 2 times smaller.

Table 1. Corrosion rates of steel 3 and corrosion inhibition coefficients of nitrogen-containing compounds (5.0 mM) in 2 M mineral acids.

Corrosion rates, g/(m^2·h)				**Corrosion inhibition coefficients**		
Without inhibitor	**IFKhAN-92**	**BDMOA**	**Catamine AB**	**IFKhAN-92**	**BDMOA**	**Catamine AB**
2 M H_2SO_4						
12	0.38	0.99	2.8	32	12	4.3
2 M HCl						
5.1	0.39	0.72	0.80	13	7.1	6.4
2 M H_3PO_4						
10	1.9	3.3	3.8	5.3	3.0	2.6

The strongest inhibition of steel corrosion in all of these environments is provided by IFKhAN-92 (Table 1). Compounds of the QAS class are inferior to it in this respect. The protective effect of nitrogen-containing CIs decreases in the series: IFKhAN-92 > BDMOA > catamin AB. With IFKhAN-92, the *k* values of steel decrease in the series $H_3PO_4 > HCl \approx H_2SO_4$. In the case of QAS, corrosion slows down in the series of inhibitors: $H_3PO_4 > H_2SO_4 > HCl$. In general, all other conditions being the same, substituted triazole – IFKhAN-92 – is the most efficient CI of steel in mineral acids; all the CIs studied show the weakest protective effect in the H_3PO_4 solution. For a more detailed understanding of the nature of the inhibiting action of the additives on the corrosion of steel in acid environments, their effect on its electrode reactions had to be studied.

Electrochemical studies

Hydrochloric acid

The corrosion of steel 3 in 2 M HCl occurs in the active dissolution region (Figure 1, Table 2). The slopes of the cathodic and anodic polarization curves are 0.10 and 0.12 V. The slope of cathodic polarization of steel (b_c) is close to the values predicted by the theory; in contrast, the slope of anodic polarization of steel (b_a) is significantly higher. We associate the reasons for the observed difference with the formation of a visually observable sludge layer on the steel surface during preliminary exposure in HCl solution and anodic polarization of the electrode. This layer creates additional diffusion limitations to the anodic reaction. On the anodic polarization curve, upon reaching a certain potential called the activation potential (E_{act}), non-polarizability of the steel electrode is observed ($b_a \rightarrow 0$) that is characterized by a sharp increase in the anodic current [74, 75]. The reasons for the anodic activation of a steel electrode in background acid solutions are associated with the removal of adsorbed hydrogen from its surface at E_{act}, which inhibits the anodic process [75–77].

Table 2. Corrosion and activation potentials (E_{cor} and E_{act}, V) of steel 3 in 2 M HCl, Tafel slopes b_c and b_a (V), cathodic and anodic current densities (i_c and i_a, A/m^2), and inhibition coefficients of cathodic and anodic reactions (γ_c and γ_a) at E=−0.30 and −0.05 V, respectively, and n=460 rpm.

C_{in}, mM	E_{cor}	b_c	i_c	γ_c	b_a	i_a	γ_a	E_{act}
Without inhibitor								
–	–0.22	0.10	35.4	–	0.12	481.5	–	–0.06
IFKhAN-92								
0.1	–0.17	i_{lim}*	5.4	6.6	0.12	15.4	31	–0.02
0.5	–0.19	i_{lim}	2.7	13	0.17	13.5	35	–0.01
1.0	–0.25	i_{lim}	0.8	44	0.18	11.5	42	0
5.0	–0.25	i_{lim}	0.8	44	0.20	9.7	50	0
10.0	–0.25	i_{lim}	0.8	44	0.21	3.8	127	0
BDMOA								
0.1	–0.15	i_{lim}	4.8	7.4	0.15	19.2	25	–0.02
0.5	–0.19	i_{lim}	2.0	18	0.17	17.2	28	–0.01
1.0	–0.21	i_{lim}	1.1	32	0.20	15.4	31	–0.005
5.0	–0.24	i_{lim}	0.9	39	0.21	9.8	49	0.01
10.0	–0.24	i_{lim}	0.9	39	0.24	5.8	83	0.02
Catamine AB								
0.1	–0.16	i_{lim}	5.2	6.8	0.13	28.8	17	0
0.5	–0.20	i_{lim}	2.8	13	0.14	17.3	28	0
1.0	–0.22	i_{lim}	2.6	14	0.15	13.8	35	0
5.0	–0.23	i_{lim}	2.6	14	0.15	13.8	35	0
10.0	–0.23	i_{lim}	2.6	14	0.15	13.8	35	0

*i_{lim} – limiting current.

All the nitrogen-containing CIs inhibit both electrode reactions on steel. The anodic process is hindered predominantly, as indicated by the shift of the corrosion potential (E_{cor}) towards more positive potentials in the presence of these additives in comparison with the E_{cor} value observed in the background environment (Table 2). In the selected range of CI concentrations (C_{in}=0.1–10 mM), both the cathodic and anodic reactions are hindered. At the maximum concentration studied, C_{in}=10 mM, the rate of the cathodic reaction at E=−0.30 V decreases 13-, 39-, and 43-fold in the presence of catamine AB, BDMOA, and IFKhAN-92. A similar picture is observed regarding the anodic reaction. The anodic reaction rate at $E=-0.05$ V decreases 35-, 83-, and 127-fold, respectively. In inhibited HCl solutions, the slopes of cathodic and anodic polarization of steel significantly exceed the background values. In the case of cathodic polarization of a steel electrode, this effect is so pronounced that a limiting cathodic current is observed in the presence of all the nitrogen-containing CIs. The observed effect is a consequence of the formation of protective layers on the steel surface in hydrochloric acid by the molecules of the compounds studied. This layer has a polymolecular structure and insulates the metal surface from the corrosive environment. As a result, the electrode reactions are controlled by the diffusion of the corrosive environment components to the electrode surface and by removal of reaction products through the protective layer of the CI. The formation of protective CI layer on the steel surface is also confirmed by the fact that the cathodic reaction rate at high concentrations, $C_{\text{in}}=1-10$ mM, nearly does not depend on their content in the corrosive environment. This effect is typical of a steel surface coated with a polymolecular protective layer of an inhibitor.

Like in the background HCl solution, the anodic polarization curves of steel in the presence of a CI contain a non-polarizability region. In acid solutions that contain CIs capable of forming protective films from CI molecules on steel surfaces, the anodic activation of a steel electrode is due to the removal of adsorbed hydrogen from the metal surface, partial desorption of a CI, and formation and growth of pits on the CI-free steel surface. In this case, E_{act} is the more positive, the higher the CI content in the corrosive environment [75]. Similar regularities are observed in the case being studied. Moreover, the value of E_{act} is 0.04−0.06 V higher in the presence of IFKhAN-92 than in the background. In the case of catamine AB and BDMOA, these values are 0.06 and 0.04−0.08 V, respectively.

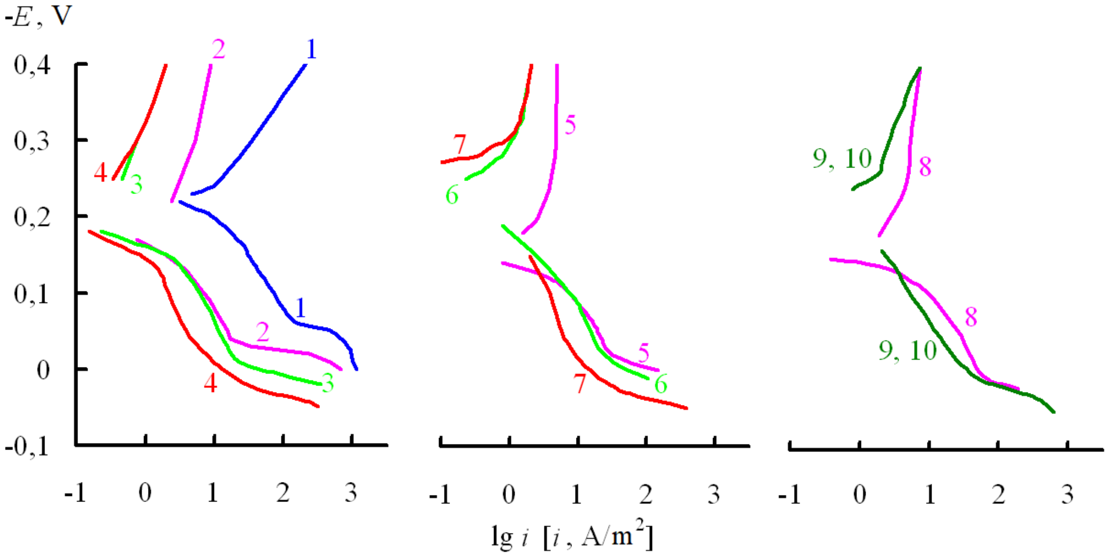

Figure 1. Polarization curves of steel 3 in 2 M HCl (1) in the presence of nitrogen-containing compounds (mM): 2, 5, 8 – 0.1; 3, 6, 9 – 1.0; 4, 7, 10 – 10.0. Curves 2–4 – IFKhAN-92, 5–7 – BDMOA, 8–10 – catamine AB.

Analysis of polarization data on steel in HCl solutions shows that all the CIs studied form protective layers on the steel surface in this environment. This mechanism of action of the CIs determines the strong simultaneous inhibition of the anodic and cathodic reactions on steel that are largely different in nature, which ultimately leads to efficient inhibition of steel corrosion. The effect of the studied CIs on the anodic and cathodic reactions on steel decreases in the series IFKhAN-92 > BDMOA > Catamin AB that fully agrees with the data on the hindrance of steel corrosion.

Sulfuric acid

The corrosion of steel 3 in 2 M H_2SO_4 occurs in the active dissolution region, and the slopes of the cathodic and anodic polarization curves are 0.10 and 0.07 V, respectively (Figure 2, Table 3). The higher slope b_a of steel in the H_2SO_4 solution also results from the formation of a sludge layer on the steel surface. In contrast to the HCl solution, no anodic non-polarizability of the steel is observed on the anodic polarization curve of steel in 2 M H_2SO_4.

Table 3. Corrosion potentials (E_{cor}, V) of steel 3 in 2 M H_2SO_4, Tafel slopes b_c and b_a (V), cathodic and anodic current densities (i_c and i_a, A/m^2), and inhibition coefficients of cathodic and anodic reactions (γ_c and γ_a) at $E = -0.30$ and -0.05 V, respectively, and $n = 460$ rpm.

$C_{in.}$, mM	E_{cor}	b_c	i_c	γ_c	b_a	i_a	γ_a
			Without inhibitor				
–	–0.20	0.10	29.2	–	0.07	1260	–
			IFKhAN-92				
0.1	–0.17	i_{lim}*	6.4	4.6	→ 0	1215	1.0
0.5	–0.13	i_{lim}	3.8	7.7	→ 0	977	1.3
1.0	–0.12	i_{lim}	1.3	23	→ 0	969	1.3
5.0	–0.08	i_{lim}	0.7	42	→ 0	385	3.3
10.0	–0.05	i_{lim}	0.4	73	→ 0	10**	126
			BDMOA				
0.1	–0.18	i_{lim}	7.7	3.8	0.07	1006	1.3
0.5	–0.16	i_{lim}	6.9	4.2	→ 0	982	1.3
1.0	–0.15	i_{lim}	5.4	5.4	→ 0	977	1.3
5.0	–0.10	i_{lim}	5.0	5.8	→ 0	700	1.8
10.0	–0.06	i_{lim}	1.5	20	→ 0	100**	13

$C_{in.}$, mM	E_{cor}	b_c	i_c	γ_c	b_a	i_a	γ_a
Catamine AB							
0.1	−0.14	i_{lim}	12.3	2.4	0.04	685	1.8
0.5	−0.14	i_{lim}	9.2	3.2	0.04	685	1.8
1.0	−0.14	i_{lim}	6.2	4.7	0.04	658	1.9
5.0	−0.11	i_{lim}	5.4	5.4	0.04	377	3.3
10.0	−0.09	i_{lim}	3.1	9.4	→ 0	292	4.3

*i_{lim} – limiting current.

**Found by extrapolation of the linear segment in the E *vs*. log i_a curve to $E=-0.05$ V.

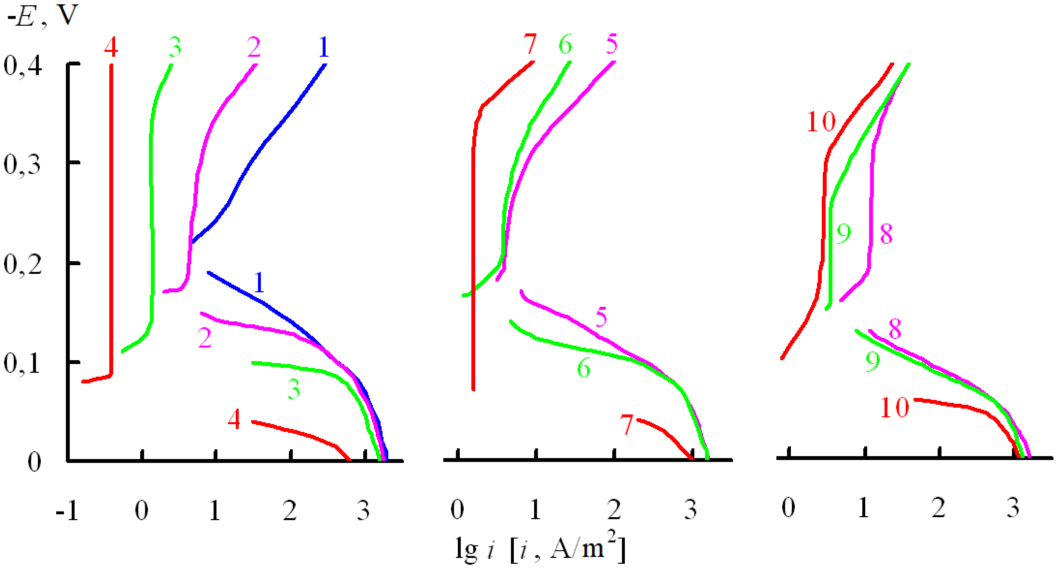

Figure 2. Polarization curves of steel 3 in 2 M H_2SO_4 (1) in the presence of nitrogen-containing compounds (mM): 2, 5, 8 – 0.1; 3, 6, 9 – 1.0; 4, 7, 10 – 10.0. Curves 2–4 – IFKhAN-92, 5–7 – BDMOA, 8–10 – catamine AB.

Addition of the CIs studied to 2 M H_2SO_4 significantly shift the E_{cor} value of steel towards positive values in comparison with the background environment. A significant shift of E_{cor} to the positive region indicates a strong predominant inhibition of the anodic reaction by the CIs studied. In the presence of 0.1−10 mM IFKhAN-92, 0.1−10 mM BDMOA, or 10 mM catamine AB, the shift of E_{cor} towards positive values is so significant that it reaches values close to the E_{act} of steel. This is indicated by the nature of the anodic polarization curves of steel that start from the non-polarizability region ($b_a \rightarrow 0$). It is interesting that in the presence of 0.1−5 mM catamine AB, the b_a slope is below the background value and is in full agreement with the values predicted by theory for steel in H_2SO_4 solutions. Apparently, this CI slows down the corrosion of steel so strongly that the formation of a sludge layer that considerably changes the parameters of the electrode reaction does not occur. At the same time, its own effect on the anodic reaction is insufficient to change the b_a value. In the presence of all the CIs studied, the cathodic reaction is characterized by a limiting current. It should indicate the formation of a protective film by these CIs on the metal surface, which creates diffusion limitations on the supply of environment components to the metal and (or) removal of the reaction products. IFKhAN-92 has the maximum hindering effect on both the cathodic and anodic processes, while the minimum effect is provided by catamine AB.

The result obtained shows that in the H_2SO_4 solution, IFKhAN-92 and BDMOA form a protective layer of CI molecules on the steel surface, which effectively slows down both of its electrode reactions. The inhibition of the anodic reaction is so significant that E_{cor} is shifted to the positive region up to E_{act}. Even on anodic polarization at steel potentials more positive than E_{act} where the CI is partially desorbed from the metal surface, the anodic current densities are smaller than the background values, which indicates that the CI effect persists. A similar picture is observed in a solution containing 10 mM of catamine AB. It is more difficult to interpret the results of electrochemical studies in solutions containing 0.1−5 mM catamine AB. On the one hand, the cathodic polarization curves characterized by a limiting current indicate the presence of a protective CI film on the metal surface. On the other hand, the slopes of the anodic polarization curves are close to the values predicted by theory, which is inconsistent with the presence of a film on steel. The combination of this contradiction with the weak effect of this IR on both electrode reactions allows us to conclude that the likelihood of the formation of its protective films on steel in this concentration range is low. Only at the high value $C_{in} = 10$ mM, one can conclude unambiguously that a protective film is formed.

Phosphoric acid

The corrosion of steel 3 in 2 M H_3PO_4 occurs in the active dissolution region (Figure 3, Table 4). The b_c slope is 0.12 V, which agrees with theory, but b_a is significantly larger than 0.04 V, which also indicates that a sludge layer exists on the steel surface in this environment. The effect of CIs on E_{cor} of steel is more complex in this acid. At relatively low C_{in}, it is more positive than in the background environment, while at higher C_{in}, it is more negative. This indicates that with an increase in C_{in}, predominant inhibition of the anodic reaction is replaced by inhibition of the cathodic reaction. In all the cases, the b_c slopes correspond to the limiting current, indicating the formation of a protective film by CI molecules on the steel surface. The slope is $b_a = 0.07$ V, which is higher than the value of $b_a = 0.04$ V predicted by the theory for the background environment, does not contradict the presence of a protective CI film on the metal surface, either. A protective layer in an inhibited environment can be formed from a mixture of sludge and the CI, but it is more likely that it is predominantly formed from IC molecules. Judging by the behavior of polarization curves in this environment, the CI film formed on steel slows down the cathodic reaction most strongly. The effect of the CI on the cathodic reaction on steel decreases in the series IFKhAN-92 > BDMOA > Catamin AB.

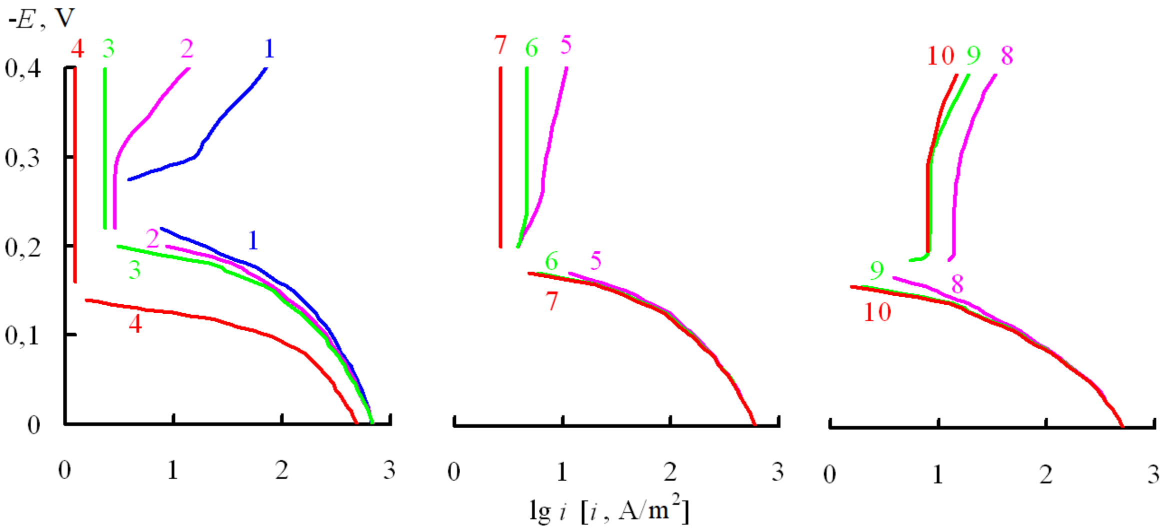

Figure 3. Polarization curves of steel 3 in 2 M H_3PO_4 (1) in the presence of nitrogen-containing compounds (mM): 2, 5, 8 – 0.1; 3, 6, 9 – 1.0; 4, 7, 10 – 10.0. Curves 2–4 – IFKhAN-92, 5–7 – BDMOA, 8–10 – catamine AB.

Table 4. Corrosion potentials (E_{cor}, V) of steel 3 in 2 M H_3PO_4, Tafel slopes b_c and b_a (V), cathodic and anodic current densities (i_c and i_a, A/m^2), and inhibition coefficients of cathodic and anodic reactions (γ_c and γ_a) at E=–0.30 and –0.05 V, respectively, and n = 460 rpm.

$C_{in.}$, mM	E_{cor}	b_c	i_c	γ_c	b_a	i_a	γ_a
			Without inhibitor				
–	–0.21	0.12	15.4	–	0.07	492	–
			IFKhAN-92				
0.1	–0.20	i_{lim}*	3.1	5.0	0.07	461	1.1
0.5	–0.20	i_{lim}	2.7	5.7	0.07	453	1.1
1.0	–0.20	i_{lim}	2.3	6.7	0.07	440	1.1
5.0	–0.17	i_{lim}	1.4	11	0.07	334	1.5
10.0	–0.15	i_{lim}	1.2	13	0.07	277	1.8
			BDMOA				
0.1	–0.19	i_{lim}	6.9	2.2	0.07	377	1.3
0.5	–0.19	i_{lim}	6.2	2.5	0.07	373	1.3
1.0	–0.19	i_{lim}	4.6	3.3	0.07	371	1.3
5.0	–0.19	i_{lim}	3.8	4.1	0.07	369	1.3
10.0	–0.19	i_{lim}	2.7	5.7	0.07	369	1.3

$C_{in.}$, mM	E_{cor}	b_c	i_c	γ_c	b_a	i_a	γ_a
Catamine AB							
0.1	–0.18	i_{lim}	15.3	1.0	0.07	265	1.9
0.5	–0.18	i_{lim}	12.7	1.2	0.07	262	1.9
1.0	–0.18	i_{lim}	8.5	1.8	0.07	260	1.9
5.0	–0.18	i_{lim}	8.1	1.9	0.07	258	1.9
10.0	–0.18	i_{lim}	8.1	1.9	0.07	258	1.9

*i_{lim} – limiting current.

* * *

A systematic study of the electrochemical behavior of steel in solutions of mineral acids containing QAS compounds and a triazole derivative has shown that their effect on the corrosion of this alloy is based on the formation of a polymolecular protective CI layer on the metal surface. This mechanism of affecting the corrosion process is quite versatile and does not depend on the composition of the corrosive environment. The experimental data obtained show that the protective layers formed by IFKhAN-92 protect steel in solutions of HCl, H_2SO_4 and H_3PO_4, despite the fact that the corrosion mechanism of steels in these environments is different. At the same time, the efficiency of steel corrosion inhibition by a polymolecular CI layer depends on its properties. The stronger it is bound to the metal surface and the stronger its structure, the more strongly it slows down the corrosion of steel. Of the compounds studied, IFKhAN-92 forms protective layers that reduce the corrosion of steel most efficiently. The data presented in literature [78–81] show that this CI is capable of chemisorption on the surface of steel from acid solutions by interacting with it via the triazole ring. Above the chemisorbed CI monolayer, its overlying layers are arranged that are bound to each other by chemical or physical forces. The chemical interaction between substituted triazole molecules inside and between CI monolayers can provide the formation of their polymeric complex with Fe(II) cations formed upon steel corrosion [82–84]. The molecules of the QASs studied do not contain fragments in the structure that could provide their chemical interaction with the steel surface and with each other. The possibility of weaker and more versatile physical interactions of their molecules still remains. The protective layers formed from them would be less efficient in inhibiting corrosion, as we actually observed.

Conclusions

1. The course of the anodic reaction of low-carbon steel in background solutions of mineral acids (HCl, H_2SO_4, and H_3PO_4) can be significantly altered by the formation of a sludge layer on its surface upon exposure at the corrosion potential and upon anodic polarization. The sludge formed on steel surface creates diffusion hindrance for the removal of reaction

products, which can significantly increase the slopes of anodic polarization in comparison to the values predicted by existing approaches to the description of the anodic reaction mechanism.

2. The mechanism of the inhibitory action of triazole and QAS derivatives on the cathodic and anodic reactions on steel is based on the formation of a polymolecular protective layer of CI molecules on the metal surface by these compounds. This is a versatile mechanism of CI action. Protective polymolecular CI layers slow down the cathodic and anodic reactions of steel in various solutions of mineral acids, despite the fact that the mechanism of electrode reactions changes depending on the nature of the acid present in the solution.
3. IFKhAN-92 forms the most stable protective layers on the steel surface in acid environments. The presence of a triazole ring in the structure of this compound enables the chemisorption interaction of its molecules with the steel surface in the course of the formation of a protective layer. Moreover, the chemical structure of the molecules allows them to be chemically bound within the bulk of the protective layer. QAS should form a protective layer through physical interactions of their molecules, which decreases their stability and, as a consequence, the efficiency of slowing down the electrode reactions.
4. Regardless of the composition of mineral acid solutions, the protective effect of the CIs studied decreases in the series IFKhAN-92 > BDMOA > catamin AB. The higher protective effects of IFKhAN-92 in steel inhibition in solutions of various acids in comparison with QAS are due to the maximum hindrance of electrode reactions by the former.

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