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EVALUATION OF THE EFFECTIVENESS OF AN AMIDO-CONTAINING CORROSION INHIBITOR IN MINERALIZED WATER-OIL ENVIRONMENTS

In this work, the effectiveness of inhibiting corrosion processes in mineralized water-oil media using a complexing amine-containing corrosion inhibitor was investigated in detail. The inhibition efficiency and corrosion rate were determined using the massometric method. According to the results of the experiments, it was found that the thiourea inhibitor was most effective in a medium with a NaCl concentration of 3 % and 10 % at pH 6–7 and doses of 10–50 mg/dm^3. It was under such conditions that a significant decrease in the corrosion rate was observed compared to control samples without an inhibitor.
In addition, the influence of temperature and acidity of the medium was an important factor. It was shown that at a weakly acidic pH and an elevated temperature of 80 °C, thiourea demonstrated increased inhibitory properties.
At the same time, the results obtained indicate that the inhibitor is effective at lower temperatures. In particular, some of the experimental data demonstrate a high level of metal protection in environments with a fixed pH value at a temperature of 20 °C. After the conducted studies, it can be stated that the amino-containing inhibitor thiourea is a promising corrosion inhibitor for use in mineralized water-oil systems.

***Keywords**: water-oil media, corrosion, inhibitor, temperature, pH, resource efficiency, closed water use systems*

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Statement of the problem. The destruction of metal equipment, mainly made of carbon steel, due to corrosion has become a serious problem that has attracted the attention of many researchers in recent decades. This is mainly due to the significant wear and tear of industrial and municipal equipment, environmental damage, which leads to significant economic losses. Global estimates show that the economic impact associated with corrosion can be from 3 to 6 % of the world's gross domestic product annually.

In many industrial sectors, carbon steel is the predominant base material due to its unique mechanical properties, making it suitable for the manufacture of equipment in a wide range of industries, particularly in water management systems. It is because of the versatility of carbon steel that there are a large number of corrosive environments and unique operating conditions that negatively affect the performance and service life of industrial equipment. This highlights the urgent need to develop effective strategies to reduce the effects of corrosion.

Analysis of previous research. Among various corrosion prevention methods, inhibitor protection has gained particular importance. This is primarily due to a number of advantages of using inhibitors, namely low effective doses, technologically simple application procedures, and cost-effectiveness.

Corrosion inhibitors are generally divided into two main categories depending on their chemical nature: inorganic inhibitors and synthetic organic inhibitors. The main commercial inorganic inhibitors include chromates, phosphates, phosphonates, nitrates and molybdates, which are most often used in neutral media. These inhibitors are known for their long-lasting anti-corrosion effect even at high temperatures and are inexpensive reagents. However, their high toxicity to ecosystems and lack of biodegradability have led to their ban in many countries. As a result, the proportion of organic inhibitors is increasing. Thiourea ($SC(NH_2)_2$) is an organic compound similar in structure to urea, in which the oxygen atom has been replaced by a sulfur atom. Its molecule has strong donor-acceptor properties due to the presence of nitrogen and sulfur atoms, which makes thiourea chemically active and suitable for the formation of coordination compounds with metals.

It is thanks to these properties that Thiourea slows down this process of corrosive destruction, forming a protective film on the metal surface, which prevents the metal from contacting aggressive agents, such as hydrogen or chloride ions. Studies show that the effectiveness of thiourea is particularly high in acidic environments, in particular in solutions of hydrochloric and sulfuric acids, where the corrosion rate is usually highest.

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The mechanism of action of thiourea as an inhibitor is based on its adsorption on the metal surface. Nitrogen and sulfur atoms have lone electron pairs that can form bonds with metal atoms, forming a dense layer that changes the surface potential and reduces the rate of anodic and cathodic reactions. The inhibition efficiency increases with increasing thiourea concentration, but in excessive amounts it can, on the contrary, destabilize the layer. Studies on carbon steel in 1 M HCl have shown that the protection efficiency reaches 80–95 % depending on temperature and concentration [1]; [2]. In a study of E-34 steel in sulfuric acid, it was found that the adsorption of thiourea obeys the Langmuir isotherm, which indicates the formation of a monomolecular layer with chemical interaction between the inhibitor molecules and the metal surface [4].

On the metal surface, thiourea can form complexes of the Fe–S or Fe–N type, which stabilize the passivation layer. In some cases, it acts primarily as an anodic inhibitor, reducing metal dissolution, in others, as a mixed inhibitor, inhibiting both parts of the corrosion process. Electrochemical studies have shown that the presence of thiourea increases the resistance to charge transfer and reduces the corrosion current density [4].

Despite its rather strong anti-corrosion properties, the use of thiourea has a number of limitations. It is a toxic compound, and its effect on organisms can be harmful with prolonged contact. There is evidence of possible carcinogenicity in high doses, although the International Agency for Research on Cancer (IARC) classifies it as a group 3 compound, i.e. "not classified as carcinogenic to humans" [5]. At elevated temperatures, thiourea can partially decompose, which reduces the effectiveness of protection. However, its availability on the Ukrainian market, even despite the full-scale invasion, and the relatively low cost of 1 m3 of treated water, at the level of 0.7–5 UAH, make its use promising both for water use systems and for equipment in the oil production and oil refining industries. However, a large number of corrosive environments and conditions of water use in industry and municipal services require a detailed study of the effect of thiourea on reducing the corrosion rate.

The aim is to establish the effect of the organic inhibitor thiourea on the intensity of metal corrosion processes in oil-containing media at different pH values and temperatures, as well as to determine the conditions under which the inhibition efficiency is maximum.

Working methodology. When conducting experiments to determine the effectiveness of corrosion inhibitors at different pH and temperatures, the degree of corrosion was determined using the massometric method. Samples in the form of rectangular plates were prepared for testing. Before testing, the samples were ground manually, then mechanically polished with a fine abrasive material until the scratches remaining from grinding were completely removed, marked and weighed. Degreasing was carried out with ethyl alcohol. At the end of the experiment, the metal samples were removed from the corrosive environment, dried and weighed again to determine the loss of metal mass [6].

The results obtained were calculated using formulas for determining the mass corrosion index, the corrosion rate reduction coefficient, and the degree of inhibitory protection against corrosion was determined for each individual sample.

The mass corrosion index was determined by the formula:

$$K_m = \frac{(Mn - Mk)}{S \cdot \tau} \quad (1)$$

where Mn – initial mass of the sample, g;
Mk – mass of the sample after the test, g;
S – sample area, m2;
τ – duration of the tests, h.

The speed reduction factor (j) is calculated by the formula:

$$j = Wx/Wi , \quad (2)$$

where Wi is the corrosion rate with inhibitor,
Wx is the corrosion rate in the blank sample.

The degree of corrosion protection (Z) was calculated based on the corrosion rate reduction coefficient, using the formula:

$$Z = \left(1 - \frac{1}{j}\right) \cdot 100\,\% \quad (3)$$

where j is the corrosion rate reduction coefficient.

Presentation of the main material. To determine the corrosion rate and the effectiveness of thiourea as a corrosion inhibitor, model solutions containing a water-mineral component (3–10 % sodium chloride solution) and an organic component – oil were used. The pH of the medium was changed using acetic acid and alkali. The model corrosion environments used in the work are typical not only for the oil production industry, but also for a wide range of industrial and municipal water systems, in particular, heating networks, circulating water supply systems, desalination plants and mining and enrichment processes. In the absence of oil products, the studied environment can be considered as a typical model for municipal and industrial water systems, while the introduction of oil allows extrapolating the obtained results to the operating conditions of the oil and gas complex equipment. This indicates the universal nature of the obtained results and the possibility of their implementation in the conditions of a circular economy and modernization of engineering infrastructure

Table 1 – Corrosion rate of St20 in solutions of different mineral composition at different temperatures

Composition of the aquatic environment	Oil concentration, cm^3/dm^3	pH	Temperature, °C	Corrosion rate, W, g/m^2h
3 % NaCl	50	9.0	20	0.0225
3 % NaCl	50	8.0	20	0.0287
3 % NaCl	50	7.0	20	0.0385
3 % NaCl	50	6.0	20	0.0458
3 % NaCl	50	5.0	20	0.0637
3 % NaCl	50	9.0	80	0.1124
3 % NaCl	50	8.0	80	0.1314
3 % NaCl	50	7.0	80	0.2409
3 % NaCl	50	6.0	80	0.2746
3 % NaCl	50	5.0	80	0.3551
10 % NaCl	50	9.0	20	0.034
10 % NaCl	50	8.0	20	0.0378
10 % NaCl	50	7.0	20	0.0465
10 % NaCl	50	6.0	20	0.0694
10 % NaCl	50	5.0	20	0.0825
10 % NaCl	50	9.0	80	0.2366
10 % NaCl	50	8.0	80	0.2475
10 % NaCl	50	7.0	80	0.3299
10 % NaCl	50	6.0	80	0.3929
10 % NaCl	50	5.0	80	0.4597

As can be seen from Table 1, for both NaCl concentrations and at both temperatures, a clear pattern of increasing corrosion rate with decreasing pH of the medium is observed. At a temperature of 20 °C in a 3 % NaCl medium, the corrosion rate increases from 0.0225 $g/m^2 \cdot h$ (pH 9) to 0.0637 $g/m^2 \cdot h$ (pH 5), i.e. almost 2.8 times. A similar trend is characteristic of a 10 % NaCl medium, where W increases from 0.034 to 0.0825 $g/m^2 \cdot h$, which corresponds to an increase of approximately 2.4 times. A decrease in pH leads to an increase in the concentration of H^+ ions, which activate the anodic process of metal dissolution and reduce the stability of protective corrosion films. An increase in temperature from 20 to 80 °C sharply intensifies corrosion processes regardless of the composition of the salt medium. For 3 % NaCl at pH 7, the corrosion rate increases from 0.0385 to 0.2409 $g/m^2 \cdot h$, i.e. more than 6 times. For 10 % NaCl at pH 7, the increase occurs from 0.0465 to 0.3299 $g/m^2 \cdot h$, i.e. approximately 7 times. This effect can be attributed to the acceleration of electrochemical reactions, increased diffusion of aggressive ions, and decreased stability of adsorption films on the metal surface at high temperatures. The presence of 50 cm^3/dm^3 of oil forms a multiphase environment in which corrosion processes are complicated by the formation of heterogeneous adsorption films. At moderate temperatures, these films can partially inhibit corrosion, but at elevated temperatures their protective properties decrease, which contributes to the localization of corrosion damage.

Based on the experimental results demonstrating a significant increase in corrosion activity with decreasing pH, increased mineralization (3–10 % NaCl) and increasing temperature to 80 °C, it is advisable to choose thiourea as one of the basic inhibitors for further studies. The nature of the thiourea molecule (S,N-atoms) ensures effective adsorption and formation of protective complexes with iron ions, which reduces anodic activity and slows down corrosion processes in high-salt and acidic environments.

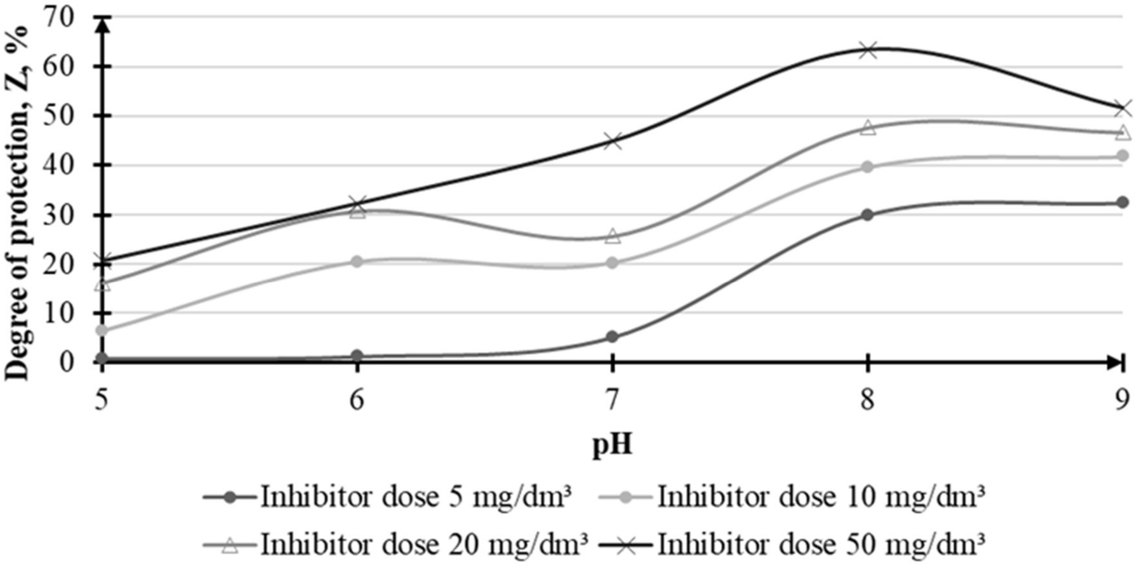

Fig. 1 – Corrosion protection efficiency when using thiourea inhibitor in 3 % NaCl solution containing oil at 20 °C

Figure 1 clearly shows that the maximum degree of corrosion protection, which is 63.7 %, is observed at a concentration of thiourea inhibitor of 50 mg/dm³ at pH 8. In general, analyzing the data of the graph, we can state the presence of a pronounced tendency to increase the degree of metal protection with increasing alkalinity of the environment and increasing the amount of inhibitor introduced. This indicates that a change in acid-base conditions is an important factor in regulating corrosion processes, and thiourea demonstrates the best parameters of action precisely in the range of weakly alkaline pH values.

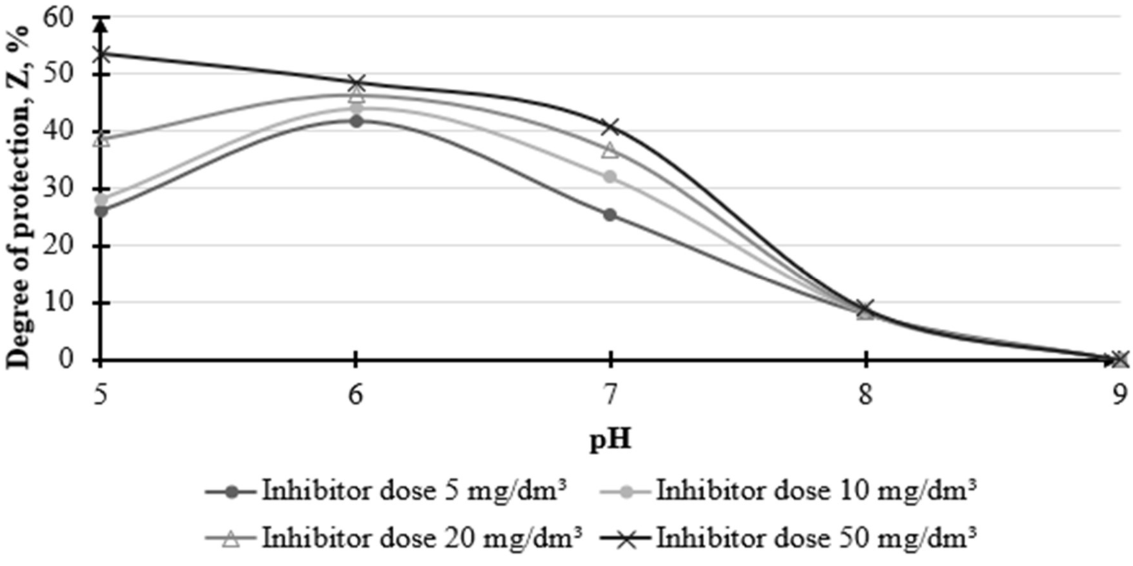

Fig. 2 – Corrosion protection efficiency when using thiourea inhibitor in 3% NaCl solution containing oil at 80 °C

According to the results presented in Figure 2, it can be determined that the most optimal indicators of the degree of corrosion protection were achieved at pH 6. The temperature regime of the experiment, which was 80 ° C, played a key role and significantly influenced the final values. With a further increase in pH, a decrease in the effectiveness of the inhibitory action is observed, which indicates the presence of an optimal pH limit, exceeding which leads to the opposite effect and a decrease in the protective ability of the system..

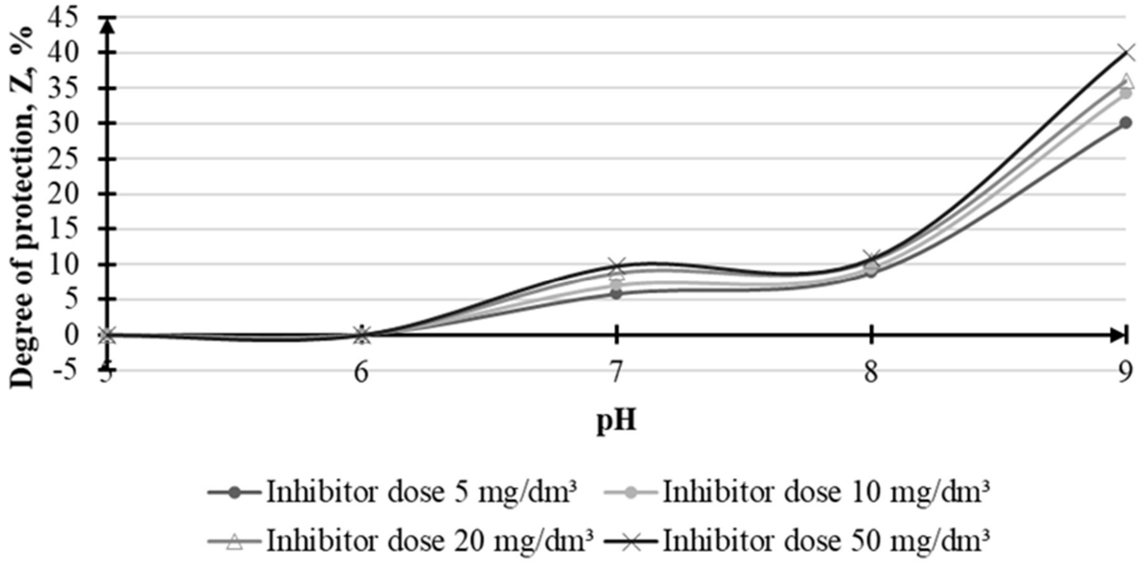

Fig. 3 – Corrosion protection efficiency when using thiourea inhibitor in 10% NaCl solution containing oil at 20 °C.

Figure 3 shows the nature of the increase in the effectiveness of the inhibitor thiourea in a 10 % NaCl solution. Based on the constructed dependence, it can be seen that optimally high values of the degree of protection are observed at pH 9. The highest value was achieved at a dose of inhibitor of 50 mg/dm³, where the degree of protection is 40 %. The obtained data confirm that an increase in the concentration of chlorides and an increase in the alkalinity of the medium can activate the mechanisms of metal passivation with the participation of the inhibitor.

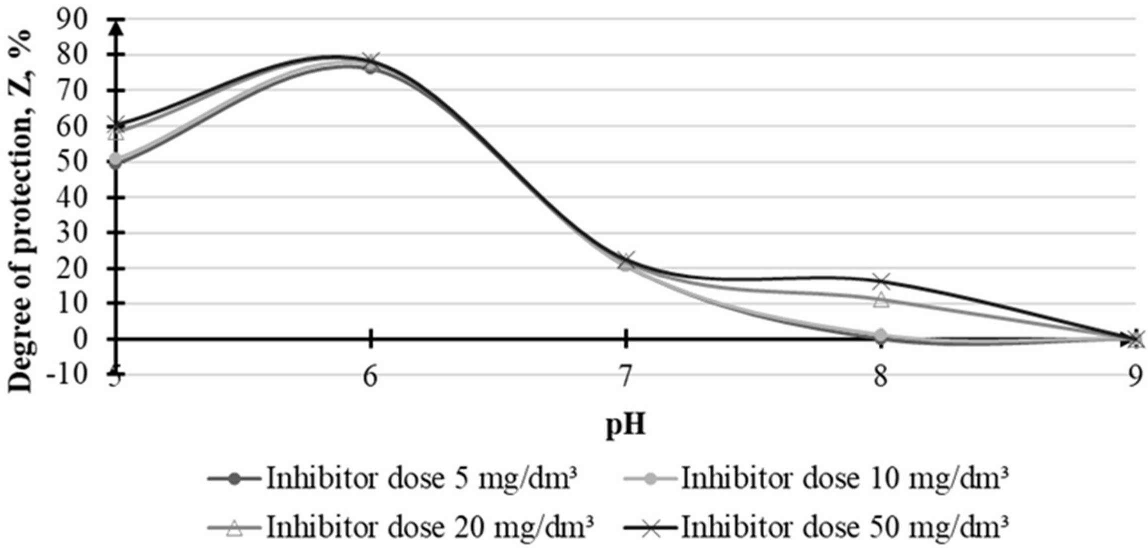

Fig. 4 – Corrosion protection efficiency when using thiourea inhibitor in 10 % NaCl solution containing oil at 80 °C

Figure 4 shows the opposite trend – the effectiveness of the thiourea inhibitor decreases significantly at pH 7, 8 and 9. This is significantly influenced by the elevated experimental temperature of 80 °C, which becomes the dominant factor determining the kinetics of the corrosion process. High temperature probably accelerates the diffusion processes and destruction of protective films, which reduces the effectiveness of the inhibitory effect even with increasing pH.

Conclusions. Analyzing the generalized results of the study of the influence of the thiourea inhibitor on the course of corrosion processes in aqueous-salt environments, it can be concluded that at a solution temperature of 20 °C, the degree of metal protection from corrosion increases with increasing pH level of the environment. In particular, in a 3 % NaCl solution, the highest degree of protection is 63.4 % and is observed at pH 8. For a 10 % NaCl solution, the optimal value is recorded at pH 9, where the maximum degree of protection reaches 40 %. In both cases, the best results were obtained at the same dose of the thiourea inhibitor – 50 mg/dm³, which indicates the presence of an optimal concentration of the reagent, which provides the most effective passivation of the metal surface under these conditions.

However, the results of experiments at a temperature of 80 °C demonstrate a significantly different pattern. At increased pH values (7, 8, 9), a decrease in the effectiveness of the inhibitory effect of thiourea is observed, which indicates a change in the mechanism of the corrosion process and possible destruction or instability of the formed protective film at high temperatures. Increased temperature can accelerate diffusion processes, activate the aggressiveness of chlorine ions and reduce the stability of inhibitory complexes on the metal surface.

At the same time, it is worth noting that at a temperature of 80 °C the inhibitor efficiency remains high in the region of lower pH values (5 and 6). The highest protection rates are 53.5 % for 3 % NaCl solution and 78.3 % for 10 % NaCl, which demonstrates potentially more favorable conditions for the use of thiourea precisely in the weakly acidic range at elevated temperatures. This suggests that the inhibitor exhibits different mechanisms of action depending on the temperature of the environment and the acid-base balance, which is an important criterion in the selection of optimal inhibitory protection parameters for industrial conditions.

Prospects for further research. Further research should focus on clarifying the mechanisms of thiourea inhibition under different temperature and pH conditions. In particular, additional studies are needed to determine the stability of the protective film at elevated temperatures, where a significant decrease in inhibitor efficiency is observed in alkaline environments. Understanding the adsorption behavior and structural changes of thiourea complexes on the metal surface will help optimize dosage and operating parameters.

It is also promising to investigate modified or combined inhibitor systems that could improve thermal stability and efficiency in highly aggressive chloride media. Testing thiourea-based formulations in real industrial water systems and varying salinity levels will provide practical validation of laboratory results and support the development of effective corrosion control technologies.

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ОЦІНКА ЕФЕКТИВНОСТІ АМІДОМІСНОГО ІНГІБІТОРА КОРОЗІЇ В МІНЕРАЛІЗОВАНИХ ВОДНО-НАФТОВИХ СЕРЕДОВИЩАХ

В даній роботі було детально досліджено ефективність інгібування корозійних процесів у мінералізованих водно-нафтових середовищах при використанні інгібітора тіосечовина. Актуальність дослідження обумовлена тим, що обладнання нафтогазової промисловості постійно працює в агресивних умовах, де присутні солі, кисень, сірководень, зміни температури та pH, що в комплексі значно прискорює руйнування металевих конструкцій. Використання ефективних і доступних інгібіторів корозії дозволяє подовжити експлуатаційний строк обладнання, знизити витрати на ремонт і технічне обслуговування, а також підвищити безпеку промислових процесів.

У дослідженні визначення ефективності інгібування та швидкості протікання корозії проводилося за допомогою масометричного методу. Цей метод, заснований на вимірюванні втрати маси металу після перебування у корозійному середовищі протягом певного періоду, дозволяє безпосередньо оцінити рівень захисної дії інгібітора. Дослідні зразки сталі піддавалися впливу середовищ з різною концентрацією хлориду натрію (NaCl), змінними показниками pH та температурою. Такий підхід відтворює умови, максимально наближені до експлуатаційних у свердловинах та трубопровідних системах.

За результатами проведених експериментів встановлено, що найефективніше інгібітор тіосечовина проявив себе у середовищі з концентрацією NaCl 3 % і 10 % при pH 6–7 та дозах 10–50 мг/дм^3. Саме за таких умов спостерігалося суттєве зниження швидкості корозії порівняно з контрольними зразками без інгібітора. Пояснення цього явища може бути пов'язане з формуванням щільної захисної плівки на поверхні металу, що перешкоджає контакту з агресивними іонами хлоридів.

Крім того, важливим фактором виявився вплив температури та кислотності середовища. Показано, що при слабкокислому pH та підвищеній температурі 80 °C тіосечовина демонструвала підвищені інгібуючі властивості. Це свідчить про її здатність активно взаємодіяти з поверхнею металу й стабілізувати захисний шар навіть в умовах високої кінетичної активності корозійних реакцій. У промислових умовах такі параметри є типовими, що робить результати дослідження особливо цінними для практики.

Водночас отримані результати вказують на те, що інгібітор ефективний і при нижчих температурах. Зокрема, частина експериментальних даних демонструє високий рівень захисту металу в середовищах із фіксованим значенням pH при температурі 20 °C. Це може бути важливим для систем з менш інтенсивним нагріванням або при транспортуванні нафти й води на поверхневих ділянках обладнання.

Загалом експеримент довів, що тіосечовина є перспективним інгібітором корозії для використання в мінералізованих водно-нафтових системах. Вона здатна забезпечити ефективний захист як при варіюванні температурних та кислотно-лужних умов, так і при різній концентрації солей у середовищі. Таким чином, використання тіосечовини як інгібітора корозії може суттєво підвищити надійність металевого обладнання в нафтогазовидобувній галузі й знизити технологічні ризики, пов'язані з руйнуванням матеріалів.

Ключові слова: *водно-нафтові середовища, корозія, інгібітор, температура, pH, ресурсоефективність, замкнуті системи водокористування*

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