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ORIGINAL RESEARCH PAPER

# NEW ENVIRONMENTALLY FRIENDLY COMPOSITIONS AS INHIBITORS OF METAL CORROSION

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**Abstract:** The development of effective and safe corrosion inhibitors of metal constructions remains an urgent problem. The aim of the present study is evaluation of inhibitory action of new compositions based on synthetic and biogenic surfactants, waste glycerol (a by-product of biodiesel) and low-toxic biocides-thiosulfonates, on the corrosion of St3 steel in different concentrations of sodium chloride aqueous solutions, at various temperatures. It was shown, that at 20 °C in medium NaCl (0.1 and 1.0 wt %) the best protective properties have mixtures of biosurfactant BS-1 with Tween-80 (1:5 and 1:3, w/w) – protection degree in 0.1 % NaCl was 69 and 61 %, respectively. With temperature increased to 50 °C, the protection degree decreased to 63 and 48 %. Waste glycerol (5 $g \cdot dm^{-3}$) in 1% NaCl also proved effectiveness, increasing St3 steel protection up to 86 %. When using biosurfactants and waste glycerol, the film on the metal surface formed due to hydrogen bonds between inhibitors functional groups and iron ions probably decreased the corrosion rate of steel. Compositions of biosurfactant BS-2 with thiosulfonates TS-1, TS-2 (ratio 0.5 and 0.25 $g \cdot dm^{-3}$) at 20 °C contribute to steel protection 99 %. Thus, action synergism was observed, therefore for effective steel protection it is advisable to use additional synergists into composition. The results indicate new prospects of compositions of biosurfactants, products of biotechnology, for new "green" corrosion inhibitors.

**Keywords:** *biosurfactants, corrosion inhibitors, rhamnolipids, thiosulfonates, trehalose lipids, waste glycerol*

## INTRODUCTION

Corrosion damage of industrial products and metal structures is an urgent problem in many industries, and sometimes the losses from them are enormous. Corrosion is considered to be one of the serious problems for most industrialized countries [1]. The corrosion process is spontaneous process that occurs in metal as a result of oxidation and occurs over a period of time at different operating temperatures. The type of corrosion mechanism and its rate of attack depend on the exact nature of the atmosphere in which the corrosion takes place. Corrosion processes are considered as the effect of high temperature, humidity, salt concentration, the acidity of environments, etc. on the metal part of the alloy [2, 3]. According to the ISO 8044 standard, corrosion can be defined as the deterioration of the state of metals due to the physical and chemical interaction between metals and the environment [4].

National Association of Corrosion Engineers (NACE) International has conducted a global study on corrosion costs and prevention strategies to estimate the cost of corrosion effects. These studies showed that the global estimated value of corrosion was 3.4 % of GDP (Gross Domestic Product) [5 – 7].

Various methods are used for corrosion prevention. Coatings are important widely used products that provide corrosion protection of metals and other substrates. Thus, paint coatings, which prevent diffusion and limit the access of an aggressive environment to the metal surface, have become widespread. When corrosion inhibitors (passivators) are added to paint materials, the protective effect of the coatings is enhanced. At present, up to 80 % of all protective coatings used in the world are paint ones; the addition of inhibitors significantly improves their barrier functions [8 – 10].

The preparation of the metal surface before applying the coating is also of great importance for the anti-corrosion characteristics. Usually, wet abrasive cleaning methods are the most effective in aqueous environments because cleaning of the metal surface creates roughness and removes invisible contaminants.

On the other hand, evaporation of water after blasting can cause rapid rust formation, and to avoid this, corrosion inhibitors are usually used in wet blasting [11].

The oil and gas industry is one of the largest users of water, especially in the case of low-yield wells. At the same time, the need to use reliable protection of metal structures of oil and gas production equipment is growing. In this regard, the development of innovative environmentally friendly corrosion inhibitors is an urgent and promising task.

The aim of our study is to evaluate the inhibitory effect of the compositions based on synthetic and biogenic surfactants, waste glycerol (a by-product of biodiesel), as well as low-toxic biocides-thiosulfonates, on the corrosion of St3 steel in different concentrations of sodium chloride aqueous solutions, at various temperatures.

## MATERIAL AND METHODS

The gravimetric method was used to quantitatively evaluate the protective ability of corrosion inhibitors in aqueous environments. The experiments were carried out according to the method described by Slobodyan *et al.* [12].

Weighed steel plates of 50x50x2 mm were completely immersed in beakers with 200 mL of compositions based on biogenic (rhamnolipids – BS-1, trehalose lipids – BS-2) and synthetic (Tween-80) surfactants, biocides-thiosulfonates (TS-1, TS-2), waste glycerol (WG), Corsol (commercial inhibitor), aqueous solutions of sodium chloride as control and kept for 7 days. Then the plates were washed, cleaned, and dried. The corrosion rate was determined gravimetrically and calculated according to the equation (1):

$$K_m = (m_1 - m_2) / S \cdot \tau \qquad (1)$$

where: $m_1$ - the initial weight of the sample [g];
$m_2$ - the sample weight after exposure to a corrosive environment and removal of corrosion products [g];
S - the total area of the sample [$cm^2$];
$\tau$ - exposure time of the sample in a corrosive environment [h].

The protective effect of the inhibitor (protection degree Z) was calculated according to the equation (2):

$$Z\ \% = (K_m – K_i / K_m) \cdot 100 \qquad (2)$$

where: $K_m$ and $K_i$ are the corrosion rates in non-inhibited and inhibited environments, respectively.

The corrosion depth index P was calculated according to the equation (3).
At least 3 repetitions were used for the tests.

$$P = (K_m / \rho) \cdot 10^{-3} \qquad (3)$$

*where*: $K_m$ is a corrosion rate [$g \cdot cm^{-2} \cdot h^{-1}$]
$\rho$ is a metal density [$g \cdot cm^{-3}$].

The development and use of environmentally safe inhibitors to combat corrosion remains an urgent issue for protecting metals and preserving the environment [13].
The objects of research were biosurfactants – products of microbial synthesis (rhamnolipids [14] and trehalose lipids [15]), obtained at the Department of Physical Chemistry of Fossil Fuels, L. M. Lytvynenko Institute of Physical Organic Chemistry and Coal Chemistry of the National Academy of Sciences of Ukraine; waste glycerol from biodiesel manufacturing process, thiosulfonates – synthetic analogs of garlic phytoncides synthesized at the Department of Biologically Active Compounds, Pharmacy and Biotechnology of Lviv Polytechnic National University. The commercial inhibitor Corsol was used for the comparison. This inhibitor in a concentration of 40 - 200 $mg \cdot L^{-1}$ is applied within the temperature range of 10 - 70 °C in the industry to prevent corrosion in water circulation systems. Corsol also has a biocidal effect on dinitrophic, sulfate-reducing, and other bacteria.

## RESULTS AND DISCUSSION

The anti-corrosion activity of substances of various chemical natures and their compositions on corrosion of steel St3 was evaluated at different temperatures in sodium chloride aqueous solutions of different concentrations.
Since the new, environmentally friendly inhibitors are designed to protect the metal surface of oil and gas production equipment operating in a natural environment (rivers, soils), where the sodium chloride content 0.1 and 1.0 %, we chose the mentioned

concentration range as a model. The choice of temperatures 20 and 50 °C was determined by the fact that 20 °C is the average daily temperature in summer in Ukraine, and 50 °C is a temperature that allows conducting research without significant evaporation of water; in addition, this is the temperature of the soil at a depth of 1000 meters.
Tables 1 - 6 show the results of tests of selected substances and compositions as corrosion inhibitors, in various variants at 20 and 50 °C, in medium with sodium chloride (0.1 and 1.0 %, respectively).

***Table 1.*** *Effect of surfactant mixtures on steel St3 corrosion, (20 °C, NaCl 0.1 %)*

| **№** | **Mixtures** | **Corrosion rate $K_m \times 10^6$ [$g \cdot cm^{-2} \cdot h^{-1}$]** | **Protection degree Z [%]** | **Corrosion depth index P [$mm \cdot year^{-1}$]** |
|---|---|---|---|---|
| 1 | Control | 4.17 | - | 0.0530 |
| 2 | BS-1 - 0.1 $g \cdot dm^{-3}$ | 1.85 | 55 | 0.0236 |
| 3 | Tween-80 - 1.0 $g \cdot dm^{-3}$ | 2.31 | 44 | 0.0293 |
| 4 | BS-1 - 0.1 $g \cdot dm^{-3}$+ Tween-80 - 0.5 $g \cdot dm^{-3}$ ratio 1:5 | 1.27 | 69 | 0.0161 |
| 5 | BS-1 - 0.1 $g \cdot dm^{-3}$+ Tween-80 - 0.3 $g \cdot dm^{-3}$ ratio 1:3 | 1.59 | 61 | 0.0203 |
| 6 | BS-1 - 0.1 $g \cdot dm^{-3}$+ Tween-80 - 0.15 $g \cdot dm^{-3}$ ratio 2:3 | 2.28 | 45 | 0.0289 |
| 7 | Corsol – 0.1 $g \cdot dm^{-3}$ | 0.92 | 78 | 0.0117 |
| 8 | Corsol - 0.1 $g \cdot dm^{-3}$+ BS-1 - 0.1 $g \cdot dm^{-3}$ ratio 1:1 | 1.05 | 75 | 0.0134 |

BS-1 – rhamnolipids

It was shown (Table 1) that the industrial inhibitor Corsol (0.1 $g \cdot L^{-1}$, 20 °C, NaCl – 0.1 %) exhibits a 78 % protection degree, and its composition with biosurfactant BS-1 (0.1 $g \cdot L^{-1}$) protects steel by 75 %. The mixtures of biosurfactant BS-1 with synthetic surfactant Tween-80 (1:5 and 1:3, w/w) have the best activity among the surfactant's compositions – the protection degree is 69 and 61 %, respectively.
The industrial inhibitor Corsol (0.1 $g \cdot L^{-1}$) has a 70 % protection degree at 20 °C and NaCl 1.0 % (Table 2), but its composition with BS-1 (0.1 $g \cdot L^{-1}$) protects steel only by 45 %. Among surfactants, mixtures of BS-1 with Tween-80 (1:5 and 1:3) have the best activity - the protection degree is 67 - 75 %; the composition of BS-1 with Tween-80 in a ratio of 2:3 has somewhat lower protection degree (34 %).
As the salt concentration increases, the protective properties of BS-1 compositions with Tween-80 composition (1:5 and 1:3) increase by 6 %.
The effect of surfactants on the anticorrosion protection of St3 steel at 50 °C and sodium chloride concentrations of 0.1 % and 1.0 % was also studied (Tables 3, 4).

***Table 2.*** *Effect of surfactants mixtures on steel St3 corrosion, (20 °C, NaCl, 1.0 %)*

| **№** | **Mixtures** | **Corrosion rate $K_m \times 10^6$ [$g \cdot cm^{-2} \cdot h^{-1}$]** | **Protection degree Z [%]** | **Corrosion depth index P [$mm \cdot year^{-1}$]** |
|---|---|---|---|---|
| 1 | Control | 2.72 | - | 0.0347 |
| 2 | BS-1 - 0.1 $g \cdot dm^{-3}$ | 1.28 | 53 | 0.0163 |
| 3 | Tween-80 - 1.0 $g \cdot dm^{-3}$ | 2.08 | 23 | 0.0265 |
| 4 | BS-1 - 0.1 $g \cdot dm^{-3}$ + Tween-80 - 0.5 $g \cdot dm^{-3}$ ratio 1:5 | 0.66 | 75 | 0.0084 |
| 5 | BS-1 - 0.1 $g \cdot dm^{-3}$ + Tween-80 - 0.3 $g \cdot dm^{-3}$ ratio 1:3 | 0.88 | 67 | 0.0112 |
| 6 | BS-1 - 0.1 $g \cdot dm^{-3}$ + Tween-80 - 0.15 $g \cdot dm^{-3}$ ratio 2:3 | 1.78 | 34 | 0.0227 |
| 7 | Corsol - 0.1 $g \cdot dm^{-3}$ | 0.81 | 70 | 0.0103 |
| 8 | Corsol - 0.1 $g \cdot dm^{-3}$ + BS 1 - 0.1 $g \cdot dm^{-3}$ ratio 1:1 | 1.50 | 45 | 0.0191 |

BS-1 – rhamnolipids

***Table 3.*** *Effect of surfactant mixtures on steel St3 corrosion, (50 °C, NaCl 0.1 %)*

| **№** | **Mixtures** | **Corrosion rate $K_m \times 10^6$ [$g \cdot cm^{-2} \cdot h^{-1}$]** | **Protection degree Z [%]** | **Corrosion depth index P [$mm \cdot year^{-1}$]** |
|---|---|---|---|---|
| 1 | Control | 7.13 | – | 0.0908 |
| 2 | BS-1 - 0.1 $g \cdot dm^{-3}$ | 2.02 | 71 | 0.0257 |
| 3 | Tween-80 - 1.0 $g \cdot dm^{-3}$ | 4.15 | 41 | 0.0528 |
| 4 | BS-1 - 0.1 $g \cdot dm^{-3}$+ Tween-80 - 0.5 $g \cdot dm^{-3}$ ratio 1:5 | 0.22 | 63 | 0.0028 |
| 5 | BS-1 - 0.1 $g \cdot dm^{-3}$ + Tween-80 - 0.3 $g \cdot dm^{-3}$ ratio 1:3 | 3.69 | 48 | 0.0469 |
| 6 | BS-1 - 0.1 $g \cdot dm^{-3}$ + Tween-80 - 0.15 $g \cdot dm^{-3}$ ratio 2:3 | 4.54 | 36 | 0.0578 |
| 7 | Corsol - 0.1 $g \cdot dm^{-3}$ | 2.82 | 60 | 0.0358 |
| 8 | Corsol - 0.1 $g \cdot dm^{-3}$ + BS 1 - 0.1 $g \cdot dm^{-3}$ ratio 1:1 | 4.36 | 38 | 0.0555 |

BS-1 – rhamnolipids

***Table 4.*** *Effect of surfactant mixtures on steel St3 corrosion, (50°C, NaCl 1.0 %)*

| **№** | **Mixtures** | **Corrosion rate $K_m \times 10^6$ [$g \cdot cm^{-2} \cdot h^{-1}$]** | **Protection degree Z [%]** | **Corrosion depth index P [$mm \cdot year^{-1}$]** |
|---|---|---|---|---|
| 1 | Control | 8.99 | – | 0.1140 |
| 2 | BS-1 - 0.1 $g \cdot dm^{-3}$ | 5.97 | 34 | 0.0759 |
| 3 | Tween-80 - 1.0 $g \cdot dm^{-3}$ | 7.49 | 17 | 0.0953 |
| 4 | BS-1 - 0.1 $g \cdot dm^{-3}$ + Tween-80 - 0.5 $g \cdot dm^{-3}$ ratio 1:5 | 4.28 | 52 | 0.0545 |
| 5 | BS-1 - 0.1 $g \cdot dm^{-3}$ + Tween-80 - 0.3 $g \cdot dm^{-3}$ ratio 1:3 | 4.49 | 50 | 0.0572 |
| 6 | BS-1 - 0.1 $g \cdot dm^{-3}$ + Tween-80 - 0.15 $g \cdot dm^{-3}$ ratio 2:3 | 4.53 | 49 | 0.0576 |
| 7 | Corsol - 0.1 $g \cdot dm^{-3}$ | 8.97 | 0.3 | 0.1140 |
| 8 | Corsol - 0.1 $g \cdot dm^{-3}$ + BS 1 - 0.1 $g \cdot dm^{-3}$ ratio 1:1 | 7.27 | 19 | 0.0925 |

BS-1 – rhamnolipids

As can be seen from Tables 3 and 4, when the temperature rises to 50 °C, with a salt content of 0.1 % and 1.0 %, the best values were obtained for the mixtures of BS-1 with Tween-80 (1:5 and 1:3). Thus, environmentally friendly compositions of biogenic surfactants and Tween-80 have the best corrosion protection: at temperatures of 20 and 50 °C and NaCl content 0.1 and 1.0 % mixtures of BS-1 with Tween-80 with ratios of 1:5 and 1:3. It is obvious that the simultaneous use of BS-1 and Tween-80 contributes to the formation of an adsorption film on the surface of the metal, which covers the electrochemically active areas of the surface.

The protection degree of the industrial inhibitor Corsol decreases from 78 % (20 °C) to 60 % (50 °C) at sodium chloride 0.1 % and from 70 to 0.3 % at sodium chloride 1.0 %.

Waste glycerol from biodiesel (5, 10, and 20 $g \cdot L^{-1}$) was also investigated as a steel corrosion inhibitor at NaCl concentration of 10 $g \cdot L^{-1}$. The metal plates were kept at a temperature of 20 °C (control and entries 2-4), as well as at 30 °C with and without stirring (entries 5 and 6, respectively, Table 5).

***Table 5.*** *Effect of waste glycerol from biodiesel production on steel St3 corrosion*

| **№** | **Mixtures** | **Corrosion rate $K_m \times 10^6$ [$g \cdot cm^{-2} \cdot h^{-1}$]** | **Protection degree Z [%]** | **Corrosion depth index P [$mm \cdot year^{-1}$]** |
|---|---|---|---|---|
| 1 | Control | 3.26 | – | 0.0368 |
| 2 | WG 5 $g \cdot dm^{-3}$ | 4.32 | 86 | 0.0048 |
| 3 | WG 10 $g \cdot dm^{-3}$ | 4.21 | 85 | 0.0058 |
| 4 | WG 20 $g \cdot dm^{-3}$ | 4.33 | 86 | 0.0048 |
| 5 | WG 20 $g \cdot dm^{-3}$, 30 ºC | 1.87 | 43 | 0.0208 |
| 6 | WG 20 $g \cdot dm^{-3}$, 30 ºC + stirring | 4.26 | – | 0.4757 |

WG – waste glycerol

At a temperature 20 °C, the protective degree of waste glycerol is 85 - 86 % regardless of its concentration (in the studied range of 5 - 20 $g \cdot L^{-1}$). When the temperature increases to 30 °C, the protection degree decreases twice and disappears after stirring. The increase in temperature and stirring of studied environment were found to have a negative influence on the inhibitory properties of waste glycerol. Obviously, this is due to the prevailing effect of the transport stage of cathodic depolarization: increased delivery of the oxygen depolarizer to the metal surface. Waste glycerol from biodiesel (5 $g \cdot dm^{-3}$) also confirmed its effectiveness in a 1 % NaCl solution – the protective properties increased to 86 %.

Compositions of trehalose lipids with biocides-thiosulfonates were also investigated as corrosion inhibitors at room temperature (Table 6).

It was shown that compositions of trehalose lipids with TS-1 and TS-2 (0.5:0.25 $g \cdot dm^{-3}$) protect St3 steel by 98 and 99 %, respectively. The obtained data (Table 6) indicate high degrees of protection, which indicates the effectiveness of the created low-toxic inhibitors. The protective effect of the obtained compositions can be explained by the synergistic interaction of their two components. As a result, structures are formed that are strongly bound to metal surfaces through oxygen heteroatoms and π-π interaction.

***Table 6.*** *Anticorrosive effect of compositions of trehalose lipids with thiosulfonates (25 °C, NaCl, 0.1 %)*

| № | Mixtures | Concentration [g·dm$^{-3}$] | Corrosion rate $K_m \times 10^6$ [g·cm$^{-2}$·h$^{-1}$] | Protection degree Z [%] | Corrosion depth index P [mm·year$^{-1}$] |
|---|---|---|---|---|---|
| 1 | Control | – | 5.0574 | 0 | 0.0643 |
| 2 | BS-2 + TS-1 | 0.5:0.5 | 0.8189 | 84 | 0.0104 |
| 3 | BS-2 + TS-2 | 0.5:0.5 | 0.3305 | 93 | 0.0042 |
| 4 | BS-2 + TS-1 | 0.5:0.25 | 0.1011 | 98 | 0.0012 |
| 5 | BS-2 + TS-2 | 0.5:0.25 | 0.0505 | 99 | 0.0006 |

TS-1 – thiosulfonate-1, TS-2 – thiosulfonate-2, BS-2 – trehalose lipids

Probably, the simultaneous introduction of BS-1 and Twin-80 into the solution leads to the formation of an adsorption film on the surface of the metal, which contributes to the suppression of the corrosion process. Biosurfactants – rhamnolipids and trehalolipids, produced by bacteria of the genera *Pseudomonas* and *Rhodococcus*, are typical biosurfactants. They have an amphiphilic character (hydrophilic and hydrophobic parts), significantly reduce the surface tension of solutions. Rhamnolipids (RL) and trehalolipids (TL) contain fragments of sugars (rhamnose, trehalose) with oxygen-containing groups. Their protective action is explained by the fact that oxygen atoms form a bond between the inhibitor molecule and the metal surface. An oxygen atom through two unpaired electrons can form two covalent bonds, respectively, and two pairs of electrons can enter into a donor-acceptor interaction with the metal surface (due to the impossibility of excitation). The donor-acceptor bond is quite strong, it can reach the strength of a covalent bond. At the same time, of two neutral particles in a donor-acceptor bond, one of them (the donor) is positively charged, and the other (the acceptor) is negatively charged, which leads to an electrostatic bond. Hydrocarbon radicals of biosurfactants, possessing hydrophobic properties aimed at an aggressive environment, repel water and corrosive-active particles, as well as additionally shield the metal surface and strengthen its blocking.

It was established that compositions of biogenic surfactants with synthetic ones, as well as with low-toxicity biocides or biodiesel waste can be environmentally friendly corrosion inhibitors with significant practical potential. Therefore, for effective protection of steel, it is advisable to introduce additional synergists into their composition. The results showed the perspectives of using biosurfactants – products of biotechnology – as components of "green" inhibitor compositions.

## CONCLUSION

The creation of ecologically safe and effective corrosion inhibitors of metal constructions remains an urgent problem. It was established that developed compositions of biogenic surfactants with synthetic ones, as well as with low-toxicity biocides-thiosulfonates or with biodiesel waste can be environmentally friendly corrosion inhibitors with significant practical potential. The mixtures of biosurfactant BS-1 and Tween-80 (1:5 and 1:3) are the most effective inhibitors at 20 and 50 °C and NaCl model solutions (0.1 and 1.0 %). This indicates synergism of action. It was established that at a temperature of 20 °C, the

composition of BS-1 with Tween-80 at concentrations 0.1:0.5 $g \cdot L^{-1}$ protects St3 steel by 69 % in 0.1% NaCl, and by 75 % – in 1.0 % NaCl.
Therefore, for effective protection of steel, it is advisable to introduce additional synergists into their composition. Obtained data confirm the prospects for further studies of the anticorrosion effect of new compositions and creation of practical recommendations for the use of environmentally friendly inhibitors in mineralized aqueous media in various technologies.
The results showed the perspectives of using biosurfactants – products of biotechnology – as components of "green" inhibitor compositions for various technological processes.

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