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## Metal corrosion inhibition by triazoles: A review

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### Abstract

Metal corrosion and the prospects of inhibiting this process have received much interest from society and scientific research. The annual global cost of corrosion is \$2.5 trillion, equivalent to roughly 3.4% of the world's gross domestic product. Implementing corrosion prevention best practices could result in global savings of 15–35% of that cost. Numerous studies were documented and dedicated on the triazole nucleus based compounds as fascinating corrosion inhibitors of various metals in hostile media. Their unique electronic structure possessing conjugated  $\pi$  and unshared pairs of electrons on the nitrogen atoms facilitates their adsorption on metal surfaces. Thus, physical and chemical interactions between the active centers of triazoles and  $d$ -orbitals of metallic materials occur to form a film on the surface. The nature of inhibitor activity is disclosed through polarization studies (cathodic, anodic or mixed-type). The range of various substituents on the triazole ring offers a vast array of inhibitory effects. temperature and inhibitor concentration effects must also be regarded when evaluating the corrosion activation and adsorption parameters supported further by the quantum chemical parameters such as DFT and molecular dynamics simulations. In this review, we looked through several instances of the use of distinct substituted triazole nucleus based compounds as significant corrosion inhibitors for different metals in various aggressive media.

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**Keywords:** corrosion, inhibition, metals, triazoles, DFT, simulation.

## 1. Introduction

Despite the availability of corrosion inhibitors, there is an urgent need for effective inhibitors for the protection of metals against corrosion in various corrosive environments [1–3]. A corrosion inhibitor is a compound that is added in low concentrations to a corrosive solution to reduce and/or minimize the corrosion rate [4, 5]. This effect is attributed to the adsorption of inhibitor particles on the metal surface resulting in the layer formation [6]. The effect of various organic and inorganic materials on the safeguards of the metal in various corrosive environments has been studied extensively [7, 8]. The most efficient corrosion inhibitors are functionalized organic compounds containing heteroatoms (such as N, S, P, O) linked by multiple bonds called heterocyclic compounds, especially those tethering  $-OH$ ,  $-COOH$ ,  $NH_2$  are reported as excellent corrosion inhibitors in acid and/or alkaline media. Among these, triazoles are aromatic nitrogen-heterocyclic five-membered compounds also known as pyrroldiazoles and having  $C_2H_3N_3$  molecular formula. They exhibit two positional isomers (Figure 1). The first unsubstituted triazole was originally synthesized by Fischer in 1878 [9].



**Figure 1.** The positional isomers of triazole.

Progress in chemistry over the past century has exerted great effect on the design, synthesis and investigation of new inhibitors that has triggered great changes in industry which impact all aspects of protection of metal in various environments. Nowadays, substituted triazoles have been designed and synthesized to achieve well-defined primary-order structures involving unsaturation, a sequence of nitrogen atoms, and functional groups. Nevertheless, the investigation of their inhibitor efficiency remains a challenging goal. Thus, substituted triazole cores have attracted great interest as tunable flexible ligands due to their versatile coordination behavior towards a wide range of metals.

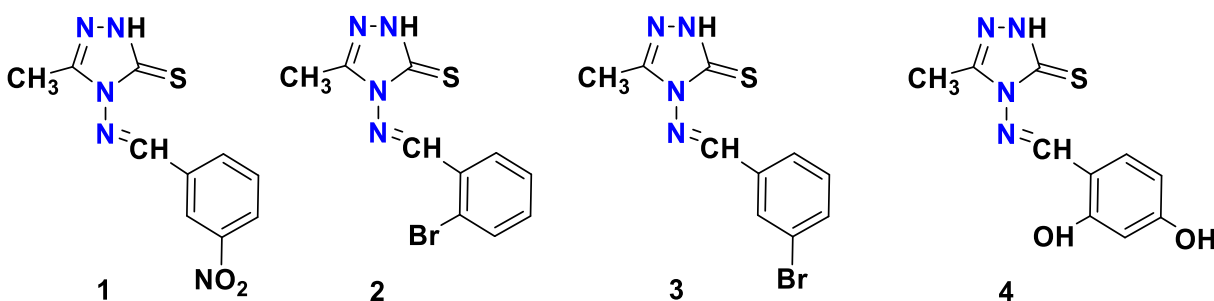
These lists are not extensive; for instance, triazoles demonstrated pharmacological importance [10–21], and the distinctive properties of the triazole core have attracted particular attention of researchers in developing new scaffold based triazole units with promising biological activities [22], which also demonstrated antifungal and antimicrobial activity [23]. They have been the subject of significant invention in relation to their use, such as the most researched SCO (Spin crossover) nanomaterials [24]. They are beneficial in a wide range of industrial applications. Numerous publications, mostly patents, showed their application in agriculture, medicines, dyes, photography, and polymers [25].

Due to the obvious aromaticity generated by the  $\pi$ -electrons, these tunable azole derivatives were frequently exploited as potential organic inhibitors. As a consequence, we

present herein a comprehensive review focusing on the use of organic compounds based on triazole moieties as fascinating organic corrosion inhibitors in a range of media, notably HCl, H<sub>2</sub>SO<sub>4</sub>, H<sub>3</sub>PO<sub>4</sub>, HNO<sub>3</sub>, and NaCl.

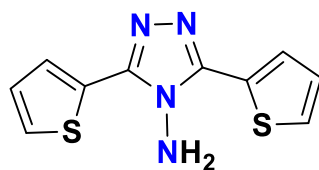
## 2. Corrosion inhibition by triazole derivatives in HCl media

Novel Schiff bases based on the 1,2,4-triazole core (**1–4**) were successfully synthesized from 1-amino-1,2,4-triazole-3-thione as the starting material (Figure 2). These triazoles were investigated for acid corrosion inhibition of mild steel using experimental and computational approaches. The results obtained showed that the targeted 1,2,3-triazole bearing the azomethine linkages revealed an excellent inhibition which was impacted by the concentration as well as the nature of the substituent in the order: **1** (92.27%) > **2** (83.66%) > **3** (83.66%) > **4** (83.66%) [26, 27]. The DFT and MD simulations were correlated and were in accordance with the experimental results [28]. The electrochemical studies showed that these materials acted as mixed-type inhibitors and their adsorption followed the Langmuir isotherm. The experimental results were in agreement with the DFT and MD simulations data.



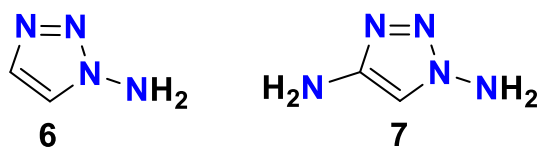
**Figure 2.** Structure of some triazoles carrying Schiff azomethine linkages (**1–4**) [26–28].

The corrosion inhibition of 3,5-bis(2-thienyl)-4-amino-1,2,4-triazole (**2-TAT**) (**5**) (Figure 3) was studied and reported [29]. The study was performed for carbon steel in 1 mol L<sup>-1</sup> HCl solution using gravimetric and electrochemical methods to explore the effectiveness of the inhibitor and its adsorption capacity on the metal. The experimental results revealed that the inhibition increased with increasing the material concentration reaching a maximum inhibition of 91.3% at 20 mg·L<sup>-1</sup>, following the Langmuir isotherm and appearing as a promising anticorrosion material.



**Figure 3.** Structure of 3,5-bis(2-thienyl)-4-amino-1,2,4-triazole (**5**) [29].

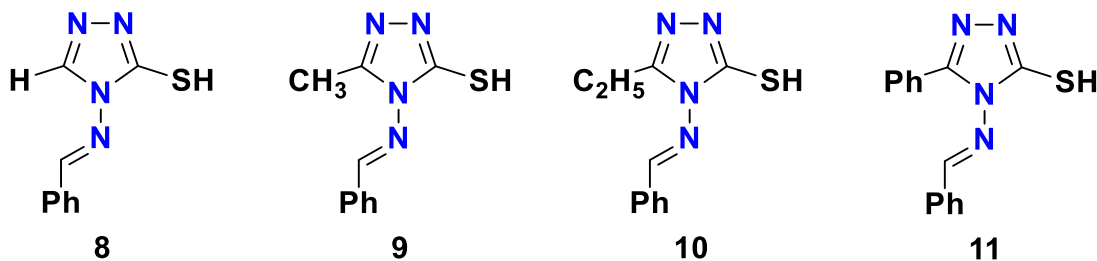
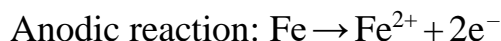
Moreover, the synthesis and the corrosion inhibition of copper in 0.5 M HCl medium was investigated using the synthesized 1-amino-1,2,3-triazole (**6**) (AT) and 1,4-diamino-1,2,3-triazole (**7**) (DAT) (Figure 4). The potentiodynamic polarization and weight loss measurements were adopted based on the effect of the concentration and showed good inhibitory efficacy and acted as mixed inhibitors obeying the Langmuir adsorption isotherm and supported by theoretical simulations [30].



**Figure 4.** Structure of 1,2,4-amino-1,2,3-triazole (**6**) (ATA) and 3,5-diamino-1,2,3-triazole (**7**) (DTA) [30].

A focused series of 5-substituted-2-mercapto-1,2,4-triazoles (**8–11**) (Figure 5) was synthesized as a new set of triazoles encompassing different substituents and heteroatoms and assessed for their corrosion and hydrogen permeation of mild steel in 1.0 M HCl based on the weight loss and electrochemical strategy exhibiting variable effectiveness (**8** > **9** > **10** > **11**). Polarization studies were also performed and showed that all investigated 1,2,4-triazoles **8–11** suppressed both anodic and cathodic corrosion reactions confirming their action as mixed type inhibitors and were in agreement with Langmuir adsorption isotherm [31].

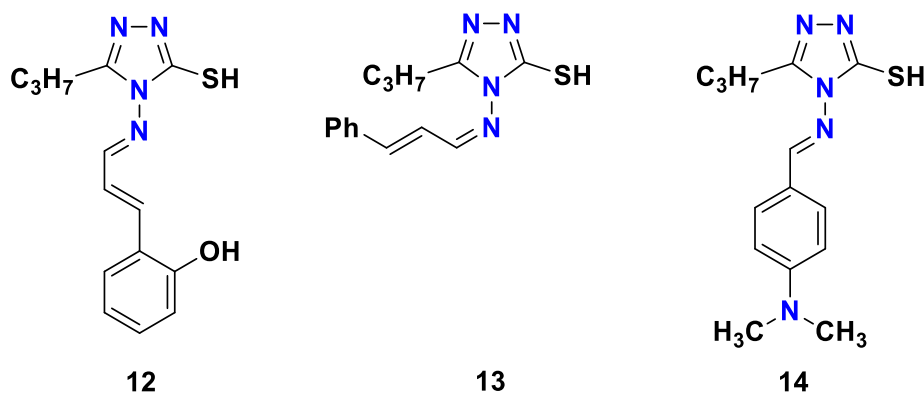
Anodic and cathodic corrosion reactions are:



**Figure 5.** Structures of 5-substituted-2-mercapto-1,2,4-triazoles (**8–11**) [31].

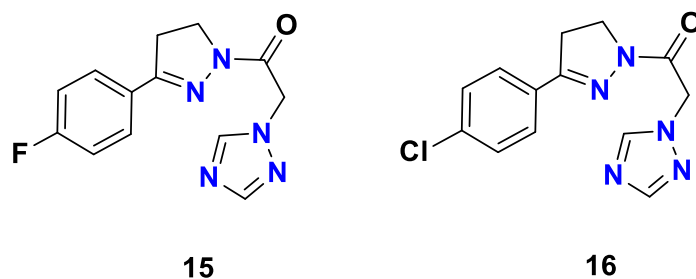
Three mercapto-1,2,4-triazole derivatives containing different azomethine linkages (**12**, **13**) presented in Figure 6 were designed, synthesized and reported as promising inhibitors with an efficiency more than 98%. The corrosion inhibition evaluation was carried out for mild steel under boiling conditions in 15% hydrochloric acid using weight loss and potentiodynamic polarization methods. The results confirmed that all the tested triazoles

(**12**, **13**) acted as mixed corrosion inhibitors by blocking the active sites on the metal surface and were in accordance with Temkin adsorption isotherm [32].



**Figure 6.** Structures of mercapto-1,2,4-triazole tethering azomethine linkage (**12–14**) [32].

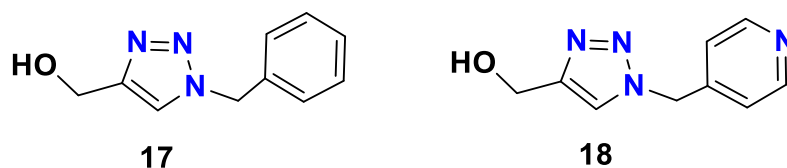
A weight loss measurement, potentiodynamic polarization and electrochemical impedance spectroscopy (EIS) investigation of substituted pyrazol-1,2,4-triazole molecular conjugates (**15**, **16**) (Figure 7) have been carried out for the evaluation of their corrosion inhibition in mild steel in 1 mol/l HCl solution. The reported 1,2,4-triazoles (**15**, **16**) were found as potent inhibitors with inhibition efficiency ranging from 91.7% to 94% at 298 K at  $10^{-3}$  M. The study reported that their inhibition increased with increasing concentration. The data also showed that their adsorption model followed the Langmuir isotherm, and the  $\Delta G^0$  values indicated that adsorption of the tested inhibitors was a spontaneous and chemisorption process [33].



**Figure 7.** Structure of substituted pyrazol-1,2,4-triazole molecular conjugates **15** and **16** [33].

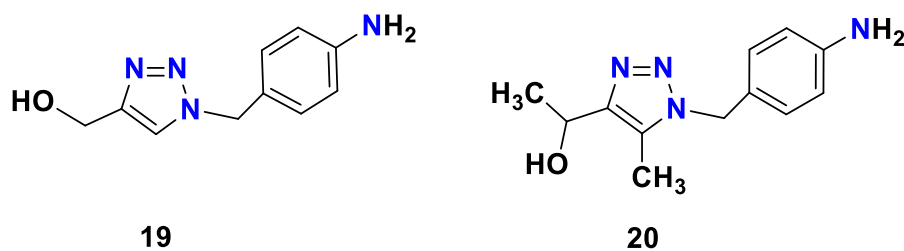
The impact of the synthesized (1-benzyl-1*H*-1,2,3-triazol-4-yl)methanol (BTM) **17** and (1-(pyridin-4-ylmethyl)-1*H*-1,2,3-triazol-4-yl)methanol (PTM) **18** presented in Figure 8 for the corrosion of C38 steel in hydrochloric acid solution was studied around 298 K by weight loss technique. The two triazole derivatives **17** and **18** exhibited inhibition efficiency values greater than 95% at 0.8 mM concentration and followed the Langmuir adsorption isotherm. As a result, the electrochemical impedance spectroscopy study revealed that PTM **18** exhibited greater corrosion inhibition efficiency than BTM **17** at the same concentration, which was in agreement with the weight loss measurements results. In order to understand

the difference in the inhibition efficiency given by the two investigated triazoles, BTM **17** and PTM **18**, and to clarify their interaction with the iron surface, density functional theory (DFT) was used for the theoretical study. This study was carried out using the Dmol3 module of the Materials Studio 6.0 software. The  $E_{\text{HOMO}}$ ,  $E_{\text{LUMO}}$ ,  $\Delta E_{\text{HOMO}}$ , and  $\mu$  dipole moment parameters were calculated and discussed to support the gravimetric and electrochemical results [34].



**Figure 8.** Structure of compounds **17**, **18** [34].

Moreover, the anticorrosive activity of 1,4-disubstituted-1,2,3-triazoles **19** and **20** for carbon steel was studied by the weight loss and electrochemical tests in acidic medium and showed considerable inhibitory efficacy (>90%) even at high temperatures (298–343 K) [35]. The inhibition efficacy increased with increasing triazoles concentration in this medium. It was noted that triazole **20** showed higher inhibition efficiency compared to compound **19**. Thus, the Langmuir isotherm proved the hypothesis supporting the chemical interaction occurring during the adsorption of inhibitors on the surface of carbon steel. However, the increase in activation energy in the presence of the inhibitors suggested a physisorption process for the interaction of the inhibitors with the surface of carbon steel.



**Figure 9.** Structure of 1,4-disubstituted-1,2,3-triazoles **19**, **20** [35].

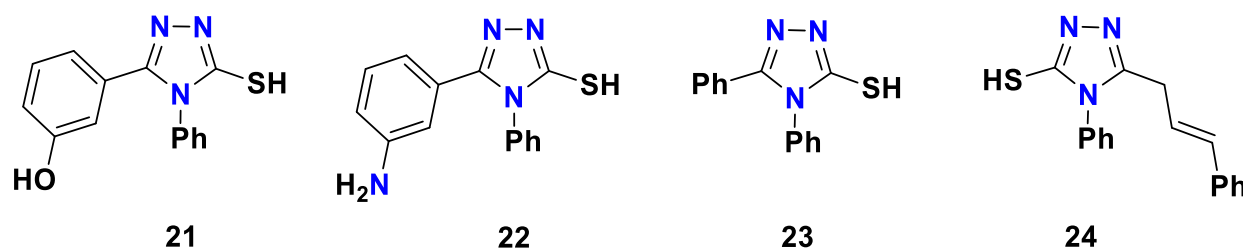
### 3. Corrosion inhibition by triazole derivatives in $\text{H}_2\text{SO}_4$ media

The copper electrode was made from a pure copper rod of 99.9 percent. Using epoxy resin to leave an exposed area of  $0.21 \text{ cm}^2$ , the rod specimens were enclosed in Teflon holders. The electrode was first mechanically polished with different grades of emery paper (to 1200 grit) before each experiment, and then ultrasonically cleaned in acetone for 2 min, followed by rinsing in double-distilled water. Reagent-grade  $\text{H}_2\text{SO}_4$  and  $\text{HCl}$  were used and double-distilled water was used for offensive solutions.

As previously reported, 3,5-bis(2-thienyl)-4-amino-1,2,4-triazole **5** [29] was synthesized and investigated for its electrochemical measurements using a classical three-

electrode cell [36]. Using the SCE, all potential values determined revealed that 2-TAT **5** was an effective inhibitor for copper in 1 M HCl and 0.5 M H<sub>2</sub>SO<sub>4</sub>, respectively [36]. Furthermore, the polarization results showed that 2-TAT **5** was a cathodic-type inhibitor in 1 M HCl and mixed-type in 0.5 M H<sub>2</sub>SO<sub>4</sub>, and that the efficiency of inhibition increased with an increase in the concentration of inhibitor **5** (Figure 3). It was reported that the addition of such material **5** enhanced the corrosion activation energy in 1 M HCl while possessing minimal influence on the activation energy in 0.5 M H<sub>2</sub>SO<sub>4</sub>.

A new series of substituted *N*-phenyl-mercapto-1,2,4-triazoles **21–24** (Figure 10) [37] was developed and implemented for their corrosion inhibition efficiency. All tested triazoles **21–24** were found to be powerful corrosion inhibitors in up to 3 N both HCl and H<sub>2</sub>SO<sub>4</sub> solutions. Their EI were proportional to their concentrations and played a great role in the prevention of mild steel corrosion *via* their adsorption on the surface of the metal, acting as filming (adsorption) inhibitors [37] according to Temkin's adsorption isotherm. In 1 N H<sub>2</sub>SO<sub>4</sub> solution, the EI of **22**, **23** and **24** derivatives did not change much as temperature was increased, whereas it increased for **21** derivative. Conversely, the EI of all derivatives **21–24** increased with increasing temperature when 1 N HCl solution was used as media. It was also observed that a slight shift in EI values for all tested materials **21–24** occurred in 1 N H<sub>2</sub>SO<sub>4</sub> solution. It was also observed that an increase in immersion time caused a slight shift in EI values for all tested materials **21–24** in 1 N H<sub>2</sub>SO<sub>4</sub> solution. While triazole **23** was the only derivative that demonstrated an increase in EI in 1 N HCl solution.



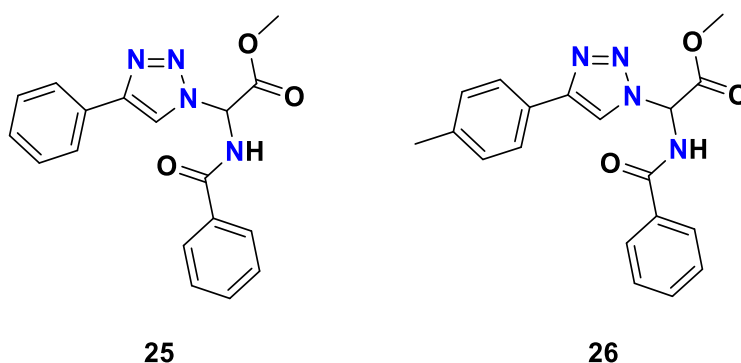
**Figure 10.** Structure of *N*-phenyl-mercapto-1,2,4-triazoles **21–24** [37].

On the other hand, two 1,4-disubstituted-1,2,3-triazolic compounds carrying a benzamido linkage and ester functionalities MBPTA **25** and MBTTA **26** were synthesized according to the reported procedure [38] (Figure 11). Assessment of the synthesized triazolic systems **25** and **26** against the corrosion of mild steel in 1 M H<sub>2</sub>SO<sub>4</sub> acidic solution at different concentrations following the Langmuir isotherm and the negative sign of  $\Delta G_{\text{ads}}^0$  showed that the compounds studied adsorbed spontaneously on steel surface, and this was supported by SEM.

The study confirmed the distinct efficacy of their corrosion inhibition, through their mixed action which was probably due to their adsorption on the mild steel surface and the charge transfer resistance ( $R_{\text{ct}}$ ) increases. The study also included the effects of weight loss, EIS and potentiodynamic polarization of the same pattern. The adsorption of such materials

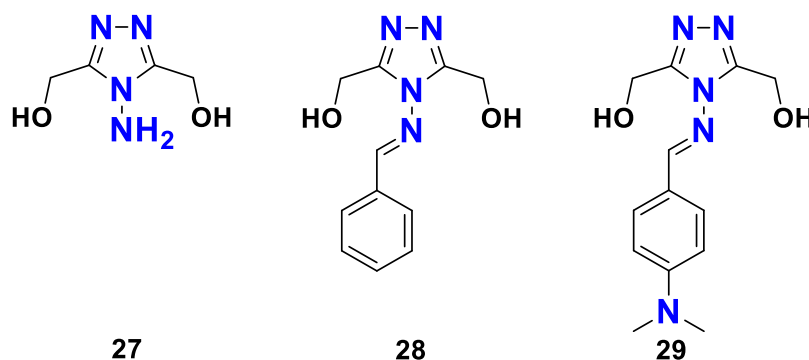


was evidenced by the theoretical outcome, and it was reported that simulation was in strong agreement with the experimental inhibition efficiencies [39].



**Figure 11.** Structure of 2-benzamido-1,2,3-triazole acetate derivatives **25** and **26** [39].

Two 1,2,4-triazole based Schiff bases **28** and **29** (Figure 12) derived from 4-amino-4*H*-1,2,4-triazole-3,5-dimethanol (**27**) as starting material were effectively constructed and synthesized for further corrosion inhibition for copper in aerated 1 N H<sub>2</sub>SO<sub>4</sub> [40]. The results showed that such scaffolds acted as good inhibitors of mixed type but at high concentrations only, and their efficiency decreased at 300 K with longer exposure periods. The inhibition occurred by their adsorption on the surface of the metal and blocking the reaction sites. All of these findings were validated by theoretical modeling, which demonstrated that triazole **29** had a greater contact with copper metal in 1 N sulfuric acid than derivatives **27** and **28**, which was most likely owing to the dimethyl amino substituent in its structure [40].

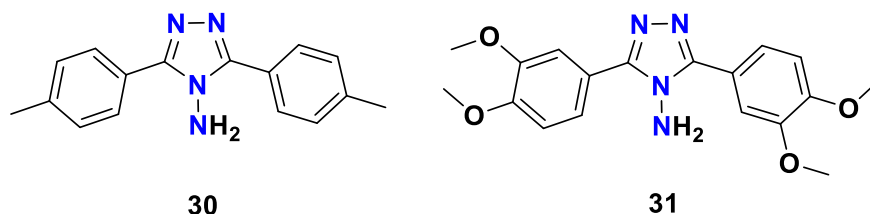


**Figure 12.** Structure of 1,2,4-triazole-3,5-dimethanol derivatives **27–29** [39].

#### 4. Inhibition of corrosion by triazole derivatives in H<sub>3</sub>PO<sub>4</sub> media

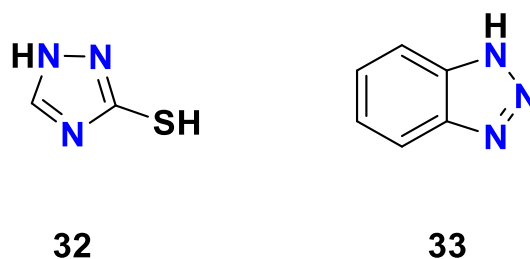
The inhibition properties of the synthesized 3,5-*bis*(disubstituted)-4-amino-1,2,4-triazole derivatives **30** and **31** for mild steel corrosion in 2 M H<sub>3</sub>PO<sub>4</sub> were reported [39] (Figure 13). The experimental results revealed the high inhibition efficiency which increased with increasing inhibitor concentration and decreased with temperature rise. The inhibitor **31** was found to be more effective than its analogue amino-triazole **30**. Furthermore,

potentiodynamic polarization data revealed that the frameworks **30** and **31** hindered the metal dissolution and hydrogen evolution processes, indicating that the tested materials are mixed-type inhibitors involving both physisorption and chemisorption in 2 M H<sub>3</sub>PO<sub>4</sub> solution and followed the Langmuir adsorption isotherm. The DFT calculations agreed with the reported results [41].



**Figure 13.** Structure of 5-bis(disubstituted)-4-amino-1,2,4-triazole derivatives **30** and **31** [39].

The inhibitory behavior of 3-mercapto-1,2,4-triazole (MT) (**32**) and benzo-1,2,3-triazole (BT) (**33**) for copper in 0.5 M H<sub>3</sub>PO<sub>4</sub> solution was studied using the mass-loss strategy [42]. In addition, gravimetric assays were applied to quantify the experimental performance of MT **32** and BT **33** triazoles. Figure 14 discloses that benzotriazole **33** was reported to be a better inhibitor than triazole **32**. Although DFT and MP2-based quantum chemical investigations provide additional results, the former has several advantages and is probably more suited for practical uses. Electronic chemical potential, molecular softness, charge transfer, and induced charge on the metal surface are some of the parameters measured that have a close relationship with the test findings for the heterocyclic scaffolds investigated here [42].



**Figure 14.** Structure of 3-mercapto-1,2,4-triazole (MT) (**32**) and benzo-1,2,3-triazole (BT) (**33**) [42].

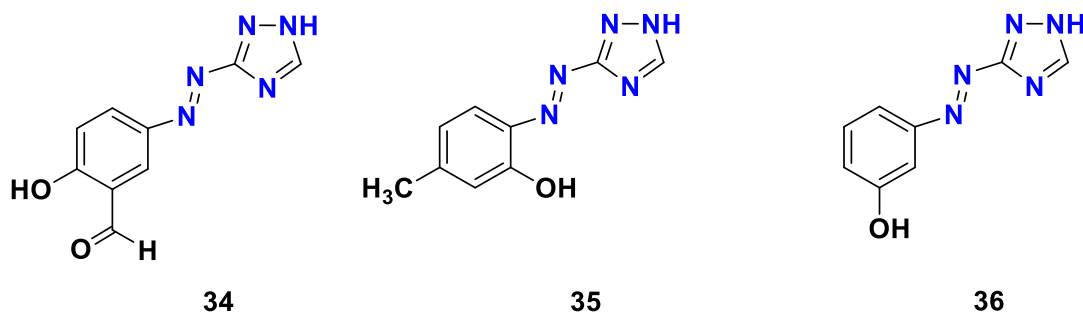
## 5. Inhibition of corrosion by triazole derivatives in HNO<sub>3</sub> media

As extension of the previous study focused on the inhibitory behavior of triazole (MT) **32** and benzotriazole (BT) **33** for copper in 0.5 M H<sub>3</sub>PO<sub>4</sub>, the same materials **32** and **33** were also tested in HNO<sub>3</sub> media.

Novel 3-arylaazo-1,2,4-triazole derivatives (AATR) **34–36** (Figure 15) were designed and developed for the assessment of their corrosion prevention efficacy for copper in aggressive HNO<sub>3</sub> solutions utilizing a dilution method (1 × 10<sup>-5</sup> to 5 × 10<sup>-4</sup> M in 0.5 M HNO<sub>3</sub>)

and free acid solution as the blank solution. The experimental findings revealed that the synthesized triazoles **34–36** showed outstanding iron corrosion inhibition activity in aerated 0.5 M HNO<sub>3</sub> (**36** ≈ **35** > **34**), which increased as the inhibitor concentration increased and reduced as temperature grew following by the modified Langmuir isothermal adsorption model and thermodynamic/kinetic El-Awady's adsorption model. Potentiodynamic polarization curves demonstrated that **34–36** substances worked as mixed type inhibitors. While, the kinetic parameters of thermodynamic adsorption and activation ( $K_{\text{ads}}$ ,  $\Delta G_{\text{ads}}^0$ ,  $A$ ,  $E_a$ ,  $\Delta H_a^0$  and  $\Delta S_a^0$ ) showed that both physisorption and chemisorption are implicated in the adsorption process.

The corrosion ability of 3-arylo-1,2,4-triazole (AATR) derivatives **34–36** was investigated and proven by the DFT quantum-chemical computations which were in agreement with the experimental results and disclosed that a relationship could be created between factors relevant to the electronic structure. The establishment of copper chelation between Cu<sup>2+</sup> and the AATRs [Cu(II)–AATR] complex in such media (HNO<sub>3</sub>) was validated by cyclic voltammetry and UV spectroscopy experiments. The calculations demonstrated that the inhibitors **34–36** adsorb on the surface of copper in appropriate order (**36** > **35** > **34**); this order accorded with the experimental technique [43].



**Figure 15.** Structure of 3-arylo-1,2,4-triazole derivatives (AATR) **34–36** [43].

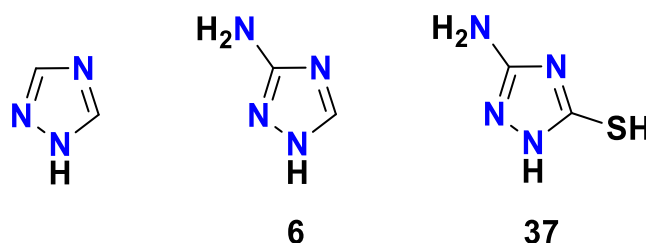
The use of new synthesized triazole compounds received nowadays more attention as corrosion inhibitors. The percentage inhibition of 1-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)ethanone (MPTE) reached 95.10% at the concentration of 0.5 mM and 303 K. The inhibitory action was explained by the presence of both nitrogen and oxygen atoms, and benzene and triazole rings that are joined together with a carbon–nitrogen single bond and  $\alpha,\beta$ -unsaturated carbonyl in conjugation with triazole rings [44]. Kuznetsov summarized in his review the use of triazoles as a class of multifunctional corrosion inhibitors [45, 46].

## 6. Inhibition of corrosion by triazole derivatives in NaCl and alkaline media

The triazole derivatives with different substituents were also investigated for their inhibitor potency in alkaline media. Thus, unsubstituted-1,2,4-triazole (TA), 3-amino-1,2,4-triazole (ATA) (**6**), and 5-amino-3-mercapto-1,2,4-triazole (AMTA) (**37**) were used as corrosion inhibitors for the copper–nickel alloy CuNi 90/10 in 3.5 wt.% NaCl solution and

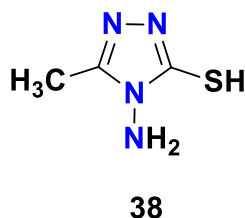
demonstrated excellent corrosion inhibition capability on the copper–nickel alloy surface through physisorption. In the concentration ranges investigated (Figure 16), 5-amino-3-mercapto-1,2,4-triazole (**37**) proved to be the most efficient chemical; the achieved effectiveness was more than 98% at  $10^{-3}$  M [47].

Whenever it comes to the use of triazole derivatives as copper alloy inhibitors in simulated sea water, the literature is extensive. As example, the synthesized 3-amino-1,2,4-triazole-5-thiol (ATT) (**37**) was assayed for corrosion inhibition of 60Cu–40Zn alloy in an aqueous solution containing 3% NaCl as a solution similar to sea water. The experimental polarization curve results supported the use of such material as good inhibitors as it acted as a mixed inhibitor, while the inhibition efficiency reaches a value of 97% (1 mM). The EIS technique results revealed that the inhibitor concentrations affected the value of the charge transfer resistance, consequently the inhibitory efficiency increased and reached a maximum value of 99% in the presence of 1 mM of ATT [48]. The tested ATT **37** also demonstrated a high inhibitory effect on copper corrosion in aerated acidic chloride pickling solution (0.5 M HCl) [49].



**Figure 16.** Structure of 1,2,4-triazole, 3-amino-1,2,4-triazole (**6**) and 3-amino-5-mercapto-1,2,4-triazole (**37**).

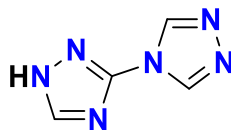
On the other hand and based on the potentiodynamic polarization and electrochemical impedance procedures, the synthesized 3-methyl-4-amino-5-mercapto-1,2,4-triazole (MAMT) (**38**) (Figure 17) was used to prevent the corrosion of 6061/Al-15(vol.%) SiC(p) composite in 0.5 M NaOH [50]. The efficiency of the used inhibitor **38** did not exceed 70% at 50 ppm.



**Figure 17.** Structure of 3-amino-5-mercapto-1,2,4-triazole (MAMT) **38**.

A novel framework named 3,4'-bi-1,2,4-triazole (bTA) (**39**) was constructed and evaluated for the corrosion ability (Figure 18). The experimental results demonstrated that bTA **39** was an effective copper corrosion inhibitor in neutral aqueous sodium chloride

solutions. In addition, the polarization analysis confirmed that bTA acted as a mixed inhibitor, and the inhibition effectiveness was reported to be high [51].

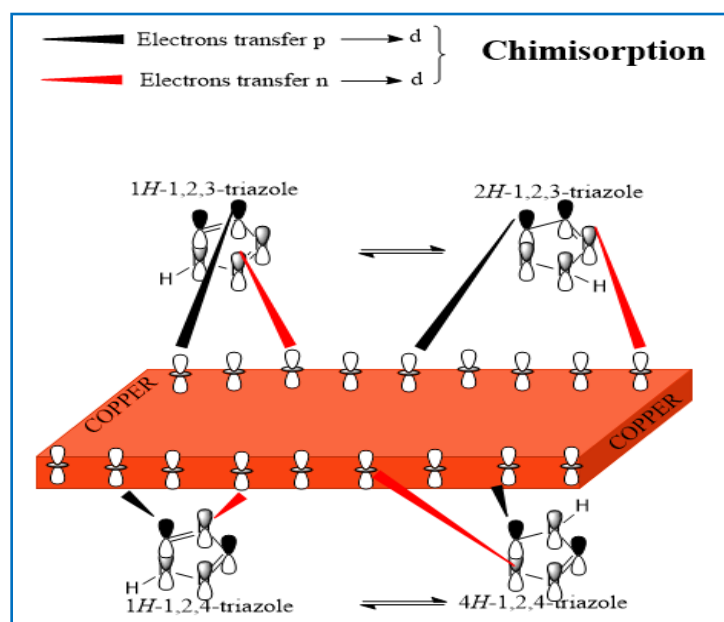


39

**Figure 18.** Structure of 3,4'-bi-1,2,4-triazole (bTA) (39).

## 7. Mechanism of the protective action of triazole derivatives

The mechanism of the inhibitory action of triazoles is generally regarded *via* the presence of three nitrogen atoms as well as  $\pi$  bonds and also the presence of various substituents and functional groups to increase or decrease the interaction of inhibitor with the *d*-orbitals of metals. Figure 19 illustrates the possible interactions to engage coordination bonds and/or Van der Waals interactions. A survey of the literature reveals that triazole derivatives can form complexes with metallic ions on the surface and consequently secure the materials against aggressive attacks [52–57].



**Figure 19.** Proposition of interactions of 1,2,3- and 1,2,4-triazoles.

## Conclusion

In the review, we emphasize the importance of using triazole compounds as organic inhibitors to retard/stop corrosion of different metallic materials in aggressive media. The molecular structure of triazole compounds plays a dominant role in interactions between

corrosion inhibitor and metal's surface. Electron-rich  $\pi$ -system aromaticity and heteroatoms (N, O, S ...) facilitate the formation of an adsorption film to play a barrier against the arrival of ions or molecules such as  $H^+$ ,  $O_2$  to destroy the metals.

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