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Azole-Based Compounds as Corrosion Inhibitors for Metallic Materials

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Abstract

To face against metallic corrosion and its corresponding undesirable consequences, the implementation of corrosion inhibitor compounds is a well-known method. In this regard, a wide range of organic heterocyclic molecules has been employed as anti-corrosion agents for several metal/medium systems. Azole-based compounds, namely, N-azole, N&S-azole (i.e., thiazole), and N and O-azole (i.e., oxazole) molecules, as well as their derivatives, have shown an excellent ability to act as efficient corrosion inhibitors for different metals and alloys in various corrosive media. For this purpose, we aim in the current chapter to discuss the application of these compounds as retarders of metallic corrosion as well as related highlighted outcomes in recent years.

Keywords: azole, oxazole, thiazole, heterocycle, corrosion, metal, inhibitor, organic, electrochemical, surface

1. Introduction

Corrosion is an undesirable natural (i.e., spontaneous) phenomenon that involves the degradation of material via its electrochemical and/or chemical reactions with the components of the adjacent aggressive environment. Metals and their alloys are known as the most susceptible materials for corrosion phenomena, which are the subject of the current chapter. This spontaneous process results in significant economic and safety losses in many industrial fields, as well as in non-industrial ones [1]. According to the recent NACE's study [2], the financial loss due to the corrosion is around 2.5 trillion \$ (USD), which is about 4.2% of the total gross domestic product. In the aim to face against metallic corrosion and corresponding outcomes, the implementation of corrosion inhibitor compounds is a well-known method due to its economic rentability, high efficiency, and simple utilization. By definition, a corrosion inhibitor is a chemical compound that is added, at a lower concentration, into the aggressive medium to prevent or retard (to an acceptable level) the corrosion of considered metallic material [3, 4].

There is a broad agreement in the corrosion literature that the inhibitor compounds protect metal against corrosion via their adsorption, namely, through chemical or/and physical adsorptions process, into the metal surface, which forms protective film upon the surface. Afterward, the formed compact film acts as a protective barrier on metal against aggressive species existing in the surrounding

environment [5, 6]. Chemical adsorption involves the sharing of electrons between inhibitor molecules and the atoms of metal surface that leads to form coordination bonds, whereas physical bonding involves the electrostatic and/or *van der Waals* interactions between the inhibitor molecules and metal surface [7].

Among employed corrosion inhibitors in the industrial area, organic inhibitors are the most used ones, which are employed mainly in acidic media during the acid pickling, acid descaling, and acid cleaning processes of metallic materials [8, 9]. These organic compounds are characterized by the presence of lone pair electrons of heteroatoms (i.e., O, N, P, and S), functional groups (e.g., alcohols, acids, and amines), and/or multiple bonds on their molecular skeletons, which act as the favorable sites of adsorption during the inhibitor-metal interactions [10]. The adsorption process of inhibitors, hence their protection ability, is related to many factors like chemical composition and charge nature of the metal surface, electronic and molecular structures of considered inhibitor, solution's pH, temperature, inhibited solution/metal contact time, hydrodynamic conditions, and so on [11].

A wide range of organic heterocyclic molecules have been used as anti-corrosion compounds for many metal/medium systems, and others are still being explored by several researchers over the world. Especially, heterocyclic molecules containing nitrogen, oxygen, and/or sulfur atoms, such as azole, oxazole, and thiazole compounds or their derivatives, have shown remarkable protection effectiveness against metallic corrosion in several aggressive media. **Figure 1** shows the molecular structure of azole moieties, which are used as corrosion inhibitors for various metallic materials. These compounds are five-atom aromatic ring molecules that contain a nitrogen atom and at least one other nitrogen, oxygen, or sulfur atom as part of the ring [12]. The azole-based compounds can be divided into three major classes, namely, N-, N&O-, and N&S-containing azole sets. In addition to their attractive molecular structures, i.e., presence of heteroatoms, double bonds, and their planar structure, azole-based compounds are soluble in almost any polar aggressive environments, particularly in acidic media.

In this context, the inhibition of metallic corrosion by using these compounds is a well-studied academic and industrial topic. **Figure 2(a)** illustrates the number of produced publications over this topic in the last 50 years. As can be seen from this histogram, the increase of publication number demonstrates an exponential behavior, which reveals that the current topic is an active one. According to available corrosion literature (**Figure 2(b)**), nitrogen-azole derivatives (N-azoles) are extensively studied and reported as corrosion inhibitors in comparison with thiazole (N&S-azoles) and oxazole (N&O-azoles) ones. It is important to outline that recently considerable attention is directed toward the synthesizing of new azole,

Figure 1.
Molecular structures of the core rings of some azole-based compounds used as corrosion inhibitors.

Figure 2.
(a) The number of produced publications per each year from 1969 to 2020 and (b) its corresponding percentage repartition for each azole-based compounds set (i.e., N-azole, N&O-azole, and N&S-azole derivatives) according to the Scopus® database.

thiazole, and oxazole substituted derivatives with higher prevention capacities and stability for different metal/medium combinations.

To quantify the prevention ability (i.e., inhibition efficiency) and/or to characterize the inhibition behavior/mechanism of azole-based compounds toward metallic corrosion, direct and indirect experimental techniques are used. Regarding direct techniques, they include weight loss (WL), the volume of liberated hydrogen gas (VG), and temperature variations (TV) [13–16]. Among them, the WL method is widely used because it can be employed in either concentrated or diluted corrosive solutions contrary to VG or TV ones. Besides, the indirect techniques include some direct current (DC) and alternating current (AC) electrochemical techniques, especially potentiodynamic polarization (PDP), electrochemical impedance spectroscopy (EIS), and electrochemical frequency modulation (EFM). In recent works, several researchers have limited their experimental investigations in the use of electrochemical techniques due to their high precision, the possibility to understand the action mechanism, minimal time, and material consumptions [17–19]. The inhibition efficiency of an examined inhibitor compounds can be calculated using Eq. (1) in which v^0 and v denote the corrosion rate of considered metal without and with the addition of inhibitor compound, respectively.

$$IE (\%) = \left(\frac{v^0 - v}{v^0} \right) \times 100 \quad (1)$$

In the present chapter, we aim to present the application of azole-based compounds as anti-corrosion agents for metals and their alloys in the corrosive aqueous media, as well as related highlighted outcomes in recent years. For this purpose, the current chapter will be divided into three sections. We begin by the application of N-azoles as corrosion inhibitors. Afterward, we move to illustrate the main findings in the case of N&S-azoles (i.e., thiazole derivatives). Finally, we end the present chapter by their N&O-azoles (i.e., oxazole derivatives).

2. Using N-containing azole compounds as corrosion inhibitors for metallic materials

Among available suggestions for metal inhibition against its corrosion, N-azole compounds have shown a remarkable ability to prevent metallic degradation in different corrosive environments. For example, good inhibition effectiveness was

outlined in the case of iron and copper as well as their alloys in almost any mineral acid, saline, and alkaline solutions. In this context, numerous corrosion inhibitors containing different N-azole nucleus structures (**Figure 1**) were tested and reported in the literature [20–24]. **Figure 3** displays the produced publications dealing with the inhibition of metal corrosion using these inhibitors in the latest 50 years. It is evident from this chart that triazole-based compounds are widely served as inhibitors compared to imidazole, pyrazole, and tetrazole ones, respectively. Subsequently, we will discuss the property of triazole- and imidazole-based compounds to retard the corrosion of metallic materials.

Triazole moiety can be found in numerous compounds that are used in a wide application range, especially in the medical field as antimicrobial, anti-inflammatory, anticancer, and antifungal drugs [20, 21]. On the other hand, the existence of three nitrogen atoms in the same molecule with a planar geometry has attracted the attention of many corrosion scientists to evaluate the protective effect of triazole molecules against metallic corrosion. Good inhibition property of either 1,2,4- or 1,2,3-triazole molecules is noted in the case of various metals, e.g., copper, iron, and its alloys in acid and non-acid media [22–24]. Recently, more attention has been focused on the development of new stable anti-corrosion compounds containing triazole core rings [25, 26]. As a result, these compounds have shown a remarkable affinity toward metallic surfaces, leading to the formation of a protective organic film on the surface of the protected metal. Furthermore, in most cases, the inhibition efficiency of these compounds increases by increasing their concentration. The role of triazole-based compounds as corrosion inhibitors for copper, iron, aluminum, zinc, and its alloys has been outlined in many corrosive media [27–31]. Among considered media, there are H_2SO_4 , HCl , HNO_3 , H_3PO_4 , and NaOH solutions at different concentrations, as well as natural/artificial seawater, sulfate, and chloride environments [32]. **Figure 4** shows the molecular structures of some 1,2,4-triazole derivatives used as effective corrosion inhibitors.

It was found that the nature of side substitutions of the triazole moiety has strongly influenced its ability to prevent corrosion phenomena. For instance, Resende et al. [33] have evaluated the inhibition capacity of three newly synthesized 1,2,3-triazole derivatives (**C-1**, **C-2**, and **C-3**, **Figure 5**) through click chemistry reaction against carbon steel corrosion in acid media. They observed that the recorded inhibition efficiency of these heterocyclic molecules depends on the substituent nature, which is ranked as **C-2** (96%) > **C-1** (92%) > **C-3** (72%) at 250 mg L^{-1} of inhibitors after 24 hours of immersion. Moreover, **C-2** and **C-1** inhibitors exhibited an excellent inhibition trend in comparison with a commercial inhibitor as reported by the authors. In another study, additionally to O&N

Figure 3.

Distribution of produced publications related to the use of N-azole family corrosion inhibitors according to the Scopus® database.

Figure 4.
 Some 1,2,4-triazole-based compounds used as corrosion inhibitors.

Figure 5.
 Some 1,2,3-triazole derivatives used as corrosion inhibitors.

heteroatoms and phenyl rings characterizing **C-1** and **C-2** compounds, the introduction of phosphorus atom (P) was done to synthesis two new ecologically 1,2,3-triazole derivatives (**C-4** and **C-5**, **Figure 5**). The corrosion assays demonstrated that the addition of dimethylamino ($-\text{N}(\text{CH}_3)_2$) functional group in the side phenyl ring has improved the prevention efficiency of newly examined inhibitors from 91 to 94% at 1 mM for mild steel in 1 M HCl solution. An inhibition efficiency over 80% is also achieved by using other 1,2,3-triazole derivatives, e.g., **C-6**, **C-7**, **C-8**, **C-9**, and **C-10** in **Figure 5** [34–37].

It is well-known for more than 60 years that the combination of triazole core ring with benzene one, the so-called benzotriazole (**C-1** in **Figure 6**), as well as their derivatives can act as efficient and stable corrosion inhibitors during long contact time for several metal/solution systems, especially for copper and its alloys [38]. For instance, this bicyclic aromatic molecule behaves as a useful inhibitor for pure copper, $\text{Cu}_{90}\text{Zn}_{10}$, and $\text{Cu}_{60}\text{Zn}_{40}$ alloys in chloride environments such as 3.5% NaCl solutions and artificial seawater [39, 40]. The good corrosion prevention capacity was also obtained both for dynamic and stagnate conditions at lower concentrations. Nonetheless, lesser inhibition efficiencies of benzotriazole and its derivatives are gained in acidic media than the base and near-neutral ones, which is due to the dissolution of formed protective film on the metal surface in acid media [38, 41]. A literature examination discloses that benzotriazole showed a particular ability to control the corrosion of AA2024 aluminum alloy in 5 mM NaCl solution as compared to 1,2,4-triazole and amino-1,2,4-triazole, and in its presence both anodic and cathodic dissolutions were reduced [27]. Additionally, in sulfide-polluted 3.5% NaCl solution, an excellent inhibition performance of 93% is obtained for carbon steel at 5 mM of benzotriazole [42].

As the main way to enhance the capability of benzotriazole to control metallic corrosion, there is the chemical modification of its molecular structure. This strategy aims to introduce more adsorption sites within the benzotriazole skeleton by adding functional groups and conjugated systems. In this regard, various benzotriazole-based derivatives were synthesized and tested as corrosion inhibitors. For instance, a new heterocyclic derivative consisting of two benzotriazole molecules and 1,3,4-thiadiazole moiety (**C-2**, **Figure 6**) exhibited good inhibition efficiency for copper in chloride environments both at acidic and near-neutral pH, 79

Figure 6.
Some benzotriazole family corrosion inhibitors.

and 87% at 1 mM, respectively [41]. Recently, two structural benzotriazole derivatives (**C-3** and **C-4**, **Figure 6**) have been reported as useful anti-corrosion compounds against the degradation of brass alloy in an artificial seawater. For example, at 150 ppm the inhibitors offer 82 and 92% as corrosion reduction percentages for **C-4** and **C-3**, respectively [39]. Furthermore, Ravichandran et al. [43] have carried out a comparative study on three benzotriazole-based inhibitors, namely, **C-1**, **C-5**, and **C-6** as depicted in **Figure 6**, for brass alloy corrosion in 3% NaCl solution. The associated outcomes of this study reveal that all tested heterocyclic molecules behave as efficient corrosion inhibitors and the inhibition efficiency increases as follows: **C-1** (77%) < **C-5** (90%) < **C-6** (93%) at lower concentration (150 ppm). The observed protection is attributed to the formation of inhibitor Cu(I) complexes on the metal surface, which isolate the surface from aggressive agents in the solution. Many other novel benzotriazole derivatives with more or less complex molecular structures have been reported in the literature as potent anti-corrosion compounds such as **C-7**, **C-8**, **C-9**, **C-10**, and **C-11** derivatives in **Figure 6** [44–46].

It is important to specify that the introduction of further functional groups into the benzotriazole skeleton has not usually improved its inhibition performance. For instance, it was outlined that the alcohol-benzotriazole derivative (**C-12**, **Figure 6**) exhibited reduced inhibition efficiency compared to simple benzotriazole for pure copper immersed in 3% NaCl medium [47]. Besides, without performed additional chemical modifications on the benzotriazole molecular skeleton, the improvement of its inhibition performance can be also done via the synergism effect with other additive species, e.g., halide and metallic ions and organic and inorganic compounds [48, 49]. As reported by Bokati et al. [50], the addition of phosphate (Na_3PO_4) and molybdate (Na_2MoO_4) compounds into corrosive solution (natural seawater) have enhanced the inhibition efficiency of benzotriazole, particularly for copper, as compared to mild steel alloy. Additionally, the mixture of benzotriazole/ Ce^{3+} was proven to have greater synergistic inhibition effect for zinc/iron and aluminum/copper model galvanic couples in NaCl solution [51, 52].

An additional N-containing azole variety compound that has also received sufficient attention is imidazole and its derivatives as well. Such attention is due to its non-toxicity and appropriate molecular and electronic structures to act as a corrosion inhibitor: the compound is planar and aromatic and contains 2 N heteroatoms. Its mechanism of action as an inhibitor is the same as stated for other reported azole compounds. An increase of concentration leads to an enhancement of its protection capacity, while in many cases the temperature has shown an undesirable effect: its increase can imply a reduction of observed inhibition property of imidazole-based

inhibitors. The tendency of imidazole heterocyclic molecules to inhibit metal corrosion, especially for copper, has been extended to synthesis novel derivatives having excellent inhibition efficiency for a longer time. The latter extension aimed to introduce additional favorable centers of adsorption via some functional or non-functional groups such as —SH, —NH₂, —COH, —OCH₃, —SCH₂Phe, and —Phe [53–57]. **Figure 7** illustrates the chemical structure of some substituted imidazole moieties used as corrosion inhibitors. It was outlined that imidazole-based compounds showed interesting activity to act as anti-corrosion agents in several corrosive environments like HNO₃, HCl, H₂SO₄, NaCl, and NaOH media, with the higher prevention efficiencies noted in chloride and in sulfuric acid solutions. **Table 1** shows the inhibition data related to the application of some imidazole derivative (**Figure 7**) as retarder compounds against copper corrosion in various media [58–64]. On the other hand, the synergism effect has also been used to improve further the attained inhibition efficiency, which was performed by adding supplementary additives, e.g., halide ions, into the inhibited solution [60].

Another common anti-corrosion compound among imidazole-based derivatives is benzimidazole, which is a heterocyclic aromatic molecule with planar geometry consisting of an imidazole and a benzene moiety (**C-1**, **Figure 8**). It was discovered for the first time by Hoebrecker as a part of vitamin B₁₂ [65]. In the last decades, benzimidazole, as well as its derivatives, has been reported as effective anti-corrosion agents for many metallic materials such as mild and carbon steels [66, 67]. The property of benzimidazole-based inhibitor to retard corrosion rate was attributed to the formation of an adsorbed protective film on the metal surface, which can consist of metal-benzimidazole complex or adsorbed benzimidazole molecules [68, 69]. As stated for benzotriazole, numerous benzimidazole derivatives with different structural compositions have been synthesized and then used as corrosion inhibitors. In this regard, simple benzimidazole derivatives showed potent inhibition effect, and in order to obtain them the chemical modification of benzimidazole core is carried out by the insertion of different functional groups. Among introduced groups, there are —SH, —NH₂, —OH, —SCH₃, —CH₂NH₂, —CH₂OH, —Cl, —Br, and carbon chain with different lengths [70–78]. **Figure 8** summarizes the chemical structures of some benzimidazole-based derivatives employed as corrosion inhibitors and their corresponding inhibition data.

Figure 7.
Molecular structures of some substituted imidazole derivatives used as corrosion inhibitors.

Inhibitor	Media	IE ([inh.])
Imidazole	0.5 M H ₂ SO ₄ /3% NaCl/0.1 M NaOH	55% (0.5 M)/50% (0.1 mM)/46% (2 mM)
C-1	1 M H ₂ SO ₄ /1 M HCl/3% NaCl	70% (10 mM)/90% (10 mM)/61% (10 mM)
C-7	0.5 M H ₂ SO ₄ /3% NaCl	93% (0.5 M)/94% (5 mM)
C-8	0.5 M 2SO ₄ /0.5 M HCl/3%NaCl	88% (0.05 M)/54% (0.1 M)/93% (0.7 mM)

[inh.]: inhibitor concentration.

Table 1.
The inhibition efficiency (IE) of imidazole and some of its derivatives (see **Figure 7**) against copper corrosion.

Figure 8.

Molecular structures of some reported benzimidazole-based derivatives as corrosion inhibitors, as well as corresponding inhibition data, which are presented as “inhibition efficiency, % (inhibitor concentration, mM)/corrosive medium, M/metal.” Abbreviations: CS, carbon steel; MS, mild steel; Fe, pure iron; Cu, copper; Zn, zinc; Al, aluminum.

An additional strategy to enhance the performance of benzimidazole to inhibit metallic corrosion is the combination of the latter heterocyclic molecule with other aromatic systems like benzene or triazole core rings without and with further substituent groups. Under this view, various hybrid benzimidazole/aromatic ring-based derivatives have been reported as anti-corrosion molecules [79–86]. **Figure 9** summarizes some benzimidazole/aromatic ring class inhibitors, as well as corresponding inhibition data. Even in very corroding media, benzimidazole/aromatic ring derivatives have shown excellent ability to protect metallic materials against corrosion in these media. For instance, it was found that **C-9** and **C-10** (**Figure 9**) derivatives could offer good protection against mild steel corrosion in a 15% HCl solution. The maximum corrosion retardation of 91% was pointed out for **C-9** derivative with —OCH₃ side phenyl substituent at 200 ppm concentration [87, 88].

On the other hand, several simple and complex bridged benzimidazole derivatives (i.e., bis-benzimidazoles) were employed as potent corrosion inhibitors in which different chain bridges are implemented as linear carbon chains without and with heteroatoms. **Figure 10** presents some bis-benzimidazole corrosion retarders. For instance, 1,4-bis-benzimidazolyl-butane (**C-1** in **Figure 10**) exhibited an efficiency of 98% at 0.68 mM inhibitor for mild steel in acid media [89], while at lower concentration (0.10 mM) the insertion of a nitrogen atom in the carbon bridge (**C-2**, **Figure 10**) provided good inhibition efficiency of 89% [71]. Ahamad et al. [90] reported the connection of two benzimidazoles via di-sulfur-bridge for the synthesis of the novel derivative (**C-3**, **Figure 10**). The corrosion tests reveal the excellent property of bridged benzimidazole inhibitors to control mild steel corrosion both in hydrochloric and in sulfuric acid media, with the attained inhibition efficiencies around 98%. Furthermore, it was found that some bis-benzimidazole derivatives can offer higher inhibition prevention for prolonged immersion time as

Figure 9.
Molecular structures of some reported benzimidazole/aromatic ring derivatives set as corrosion inhibitors as well as corresponding inhibition data.

Figure 10.
Molecular structures of some used bis-benzimidazole corrosion retarder's type.

reported by Dutta et al. [91] for C-4, C-5, C-6, and C-7 compounds, with the lower recorded efficiency 88% after 4-day immersion of mild steel in 1 M HCl solution. The length of the carbon chain of the benzimidazole bridge has influenced the ability of these derivatives to retard corrosion. In this context, three bis-benzimidazole derivatives (C-8, C-9 and C-10, **Figure 10**) exhibited a significant

tendency to reduce mild steel corrosion in acid environment, with an inhibition percentage up to 94% obtained at 0.1 mM for the derivative with longer carbon chain (i.e., **C-10**).

To understand the action mechanism of an inhibitor compound at an atomic scale, the calculation of some electronic and molecular parameters using a chemical computational approach corresponding to the adsorption process is required. In this context, Kokalj's team and other groups have studied in-depth the role of the molecular and electronic structures of many N-azole inhibitor molecules for their inhibition property for various metallic materials [92–97]. Density functional theory (DFT)-based calculations have been employed by these scientists to quantify the interaction magnitude of considered inhibitor molecules with the chosen metal surfaces, as well as their adsorption configuration onto these surfaces through qualitative analysis.

3. Using N&S-containing azole compounds (thiazoles) as corrosion inhibitors for metallic materials

Referring to previous works [98–100], heterocycle-based inhibitors with both sulfur and nitrogen atoms in their structure were offered outstanding prevention activities in comparison with those containing only sulfur or nitrogen atoms. In this regard, several N&S-containing azole compounds (**Figure 1**), like thiazole and thiadiazole derivatives, have been attested to be operational inhibitors against the corrosion of many metallic materials in a wide variety of corrosive media. Based on the available corrosion literature, special attention is devoted to thiadiazole-based compounds compared to thiazole ones. Such attention trend is based on the fact that the presence of further heteroatoms (N atoms) on those heterocyclic molecules can raise their adsorption onto the metal surface and consequently enhance their inhibition effectiveness.

In addition to the potent affinity of pre-existing heteroatoms (i.e., N and S atoms) in the 1,3-thiazole ring to interact with the metal surface during the inhibition process, the attachment of the latest ring with further substituents to improve its inhibition efficiency was recently reported. In this view, many 1,3-thiazole-based derivatives are developed via different synthesizing reaction procedures. Confering to obtained results, these new derivatives were shown to have a great tendency to reduce the dissolution of various metallic substrates. For instance, Raviprabha and Bhat [101] have evaluated the anti-corrosion property of ethyl-2-amino-4-methyl-1,3-thiazole-5-carboxylate derivative (**C-1**, **Figure 11**) for AA6061 aluminum alloy in 0.05 M HCl medium. Based on the calculated thermodynamic parameters corresponding to the adsorption process of **C-1** molecules, the chemisorption process of derivative molecules is proposed as a potential mechanism of inhibition. Moreover, it was disclosed that an increase in temperature level implies an elevation of inhibition activity of evaluated 1,3-thiazole derivative, with the prevention percentage of 93% at 333 K and 100 ppm of **C-1**. Another similar 1,3-thiazole derivative (**C-2**, **Figure 11**) with pyridinium ring also showed a good capacity to regulate copper dissolution in molar HCl solution, with a maximum of 94% as prevention efficiency achieved at 10^{-3} M.

The nature and position of added substituents in a 1,3-thiazole ring-based inhibitor can considerably influence its inhibition performance. Recently, two mono-substituted 1,3-thiazole derivatives (**C-3** and **C-4**, **Figure 11**) have revealed this behavior, which were used to protect X65 steel alloy largely employed in pipelines for natural gas transportation purposes. The ethenone-substituted 1,3-thiazole derivative (**C-4**) exhibited superior performance to control X65 steel

dissolution than isobutyl one (C-3), in which the recorded prevention efficiency at 5×10^{-3} M is being around 90 and 70%, for C-3 and C-4, respectively [102].

In addition to lateral substituents, which contain supplementary electron-donating centers (e.g., functional groups, aromatic and azole rings), the inhibition performance of 1,3-thiazole-based derivatives is also improved by increasing their electron-donating capability via attachment with a benzene ring. In this regard, Chugh et al. [103] have synthesized four new derivatives based on benzo[d]thiazole core structure (C-5, Figure 11), which exhibited an increased anti-corrosion property by replacing hydrogen atom (IE = 79%) of R substituent (on the lateral benzene ring) by chlorine atom (IE = 85%), methyl group (IE = 88%), and finally —NH_2 functional group (IE = 90%). In the same way, the combination of benzo[d]thiazole bi-rings with imidazoline ring (C-6, Figure 11) is found to act as an efficient corrosion inhibitor in the water-glycol medium [104]. More complex 1,3-thiazole derivative molecules were evaluated and reported as good corrosion inhibitors at lower concentrations, e.g., ceftriaxone 1,3-thiazole derivative (C-7, Figure 11) demonstrated an inhibition percentage of 95% at 400 ppm for mild steel in acidic environment [105]. Table 2 illustrates the relevant outcomes on the use of two other 1,3-thiazole-based compounds as corrosion inhibitors [106, 107].

1,3,4-Thiadiazoles, another class of thiazole heterocyclic molecules, have been widely examined for their uses in numerous fields such as agrochemical and pharmaceutical areas. For example, sulfamethoxazole and methazolamide are market drugs that contain a 1,3,4-thiadiazole ring [108, 109]. On the other hand, the use of 1,3,4-thiadiazole-based compounds as inhibitor additives also reduced the degradation of metals caused by the surrounding aggressive environment. Many 1,3,4-thiadiazole derivatives were reported to act as potent anti-corrosion agents in different operating conditions. The molecular structure of this five-atom ring type is characterized by the incorporation of an additional nitrogen atom into the 1,3-thiazole ring in 4 position. The presence of further heteroatoms in conjugated 1,3,4-thiadiazole-based molecules plays a curious role in their protection activities. The latest feature is due to the highest tendency of heteroatoms with the conjoint multi-bonds to facilitate the adsorption of these compounds onto the metal surface, and subsequently formed protective film isolates the substrate from solution components.

Several 1,3,4-thiadiazole derivatives with different attached hydrocarbon chains were synthesized and evaluated as corrosion inhibitors. It was found that the size and shape of inserted substituents, as well as their chemical properties, can influence the performance of developed 1,3,4-thiadiazole derivatives to retard metal dissolution. For instance, the substitution of mercapto groups at 2 and 5 positions of the thiadiazole nucleus by ethyldisulfanyl (C-1, Figure 12) augmented the achieved

Figure 11.
 Molecular structures of some 1,3-thiazole-based derivatives used as anti-corrosion agents.

inhibition efficiency from 82.4 to 88.1% at 0.4 mM of inhibitors toward copper corrosion in PAO base oil environment. Concerning the protection activity of these compounds, it was attributed to their physical adsorption on copper oxide surface as theoretically expected and experimentally verified [110]. Moreover, a series of 2,5-dimercapto-1,3,4-thiadiazole derivatives was also reported as anti-corrosion compounds by Wei and Gemmill et al. [111, 112]. Molecular structures of some reported derivatives are summarized in **Figure 13**.

In recent years, microwave irradiation heating has been used as a convenient green method for the synthesis of different heterocyclic inhibitors [113] from which we mention 2-amino-5-alkyl-1,3,4-thiadiazole derivatives, with the corresponding synthesizing scheme displayed in **Figure 14**. The length of the side alkyl chain impacted their capacity to control the dissolution of mild steel in 1 M H₂SO₄ solution, with the inhibition effectiveness increase with rising chain length, except for —C₁₃H₂₇ alkyl case for which the prevention efficiency rapidly decreases [114]. Additionally, the replacement of the alkyl chain of 2-amino-5-alkyl-1,3,4-thiadiazole by mercapto substituent (—SH) was led to a perfect protection efficiency of 99.3% [115].

On the other hand, four novel 1,3,4-thiadiazole-thiosemicarbazones derivatives and their cobalt(II) ion complexes (**C-2**, **Figure 12**) have been found to play the important role as anti-corrosion agents for carbon steel in acid media. However, the tests revealed that the molecular structure of these compounds has a little effect on the obtained inhibition efficiencies, which are around 90% in the presence of 500 ppm inhibitors [17]. Based on 1,3,4-thiadiazol-2-amine, new heterocyclic scaffold derivative (**C-3**, **Figure 12**) was synthesized and reported as an excellent inhibitor (IE = 91% at 0.5 mM) against mild steel corrosion in the molar hydrochloric acid medium [116, 117]. Another derivative of 1,3,4-thiadiazol-2-amine (**C-4**, **Figure 12**) has been also reported to act as a useful inhibitor for copper in de-aerated, aerated, and oxygenated 3% NaCl solutions, with a maximum efficiency of 94% obtained at 5 mM of the inhibitor [118]. Besides, 1,3,4-thiadiazol-containing organic inhibitors also served to improve the anti-corrosion property of

1,3-Thiazole derivative	Metal	Medium	Inhibition efficiency (IE)
Mild steel	1 M HCl	IE = 96% at 298 K and 5 mg kg ⁻¹
R = -H, -CH ₃ , -OCH ₃ , -Br, -NO ₂	304 L stainless steel	3 M HCl	IE: OCH ₃ (93%) > CH ₃ (91%) > H (89%) > Br (87%) > NO ₂ (86%). IE was enhanced by synergistic combination with 1 mM KSCN at 303 K and 11 × 10 ⁻³ mM

Table 2.
Relevant data related to the application of some 1,3-thiazole-based compounds as corrosion inhibitors.

Figure 12.
Molecular structures of some 1,3,4-thiadiazole-based derivatives used as anti-corrosion agents.

Figure 13.
Some 2,5-dimercapto-1,3,4-thiadiazole anti-corrosion compounds.

Figure 14.
Synthesis route of 2-amino-5-alkyl-1,3,4-thiadiazole derivatives under microwave irradiations.

some coatings. For instance, 2-acetyl-amino-5-mercapto-1,3,4-thiadiazole (C-5, **Figure 12**) has shown a good ability to improve the protective quality of chitosan coatings on zinc, for which the protection efficiency passed from 64 to 91% in the presence of C-5 derivative [119].

4. Using N&O-containing azole compounds (oxazoles) as corrosion inhibitors for metallic materials

In addition to N&S-containing azole corrosion inhibitors, oxazole-based compounds (i.e., N&O-containing azoles) have gained considerable attention in recent years in this regard. Oxadiazole molecule consists of a five-membered heterocyclic ring with at least one nitrogen and an oxygen atom. These N&O-containing heterocycles are interesting molecules that exist in wide biological-based compounds like diuretics, anxiolytics, and local anesthetics. Moreover, oxazole shows an antimycotic activity and can be used as anti-inflammatory agents as well as antibacterial toward pneumoniae, micrococcus, and *Staphylococcus aureus* [120, 121].

Numerous N&O-containing azole heterocyclic molecules have been studied and reported as efficient anti-corrosion agents for various metallic materials, especially in acidic media [122–126]. Such beneficial effects are related to their special affinity to adsorb on the metallic surfaces. Moreover, these compounds possess lone pair electrons on the oxygen and nitrogen atoms, which can interact favorably with the vacant orbitals of metal, leading to formation of protective barrier film [127].

Figure 15 shows the produced publications related to the inhibition of metal corrosion employing oxazole-based inhibitors in the last 50 years. It is clear from this figure that among available N&O-containing azole compounds, the oxazole, isoxazole, 1,2,4-oxadiazole, and 1,3,4-oxadiazole ones (**Figure 1**) are frequently used for corrosion inhibition purposes. Noticeable attention is focused on 1,2,4- and 1,3,4-oxadiazole inhibitors, mainly due to the presence of several nitrogen atoms on their five-membered heterocycle in comparison to other N&O-azoles.

Due to its excellent descaling properties, sulfamic acid (NH_2HSO_3) is used in a large variety of industrial applications such as cleaning of heat exchangers and cooling

water systems. As compared to other acids like hydrochloric acid, sulfamic acid shows a lower corrosion rate of stainless steel (SS) without the problem of chloride-induced stress corrosion cracking of SS. In order to reduce further this corrosion, the addition of inhibitor compounds into sulfamic media is mandatory. As effective inhibitor candidates, four new synthesized oxazole derivatives have been reported as good corrosion inhibitors for 316 L-type SS in 0.6 M NH_2HSO_3 solution by Fouda et al. [122]. The molecular structures of reported oxazole derivatives are presented in **Figure 16** (C-1, C-2, C-3, and C-4). According to weight loss experiments and electrochemical tests via different techniques, a good prevention ability around 90% is recorded at lower concentration (i.e., 2×10^{-4} M) of investigated derivatives after a moderate immersion time (3 h), especially for the fourth derivative (C-4). In addition to the presence of benzene ring and nitrogen and oxygen atoms, the good inhibition property of C-4 derivative as compared to the other ones is attributed to the existence of four aromatic rings as substituents, which results in its larger molecular size and planar geometry, leading to highest coverage of the metal surface area by adsorbed C-4 molecule. Based on this study, it can be outlined that the substitution of oxazole core ring by biggest lateral substituents can effectively improve the inhibition property of oxazole derivatives at lower concentrations.

The protection activity of other oxazole derivatives set has been reported in recently published work [123]. The authors of this work have synthesized a series of three 2-phenyl oxazole derivatives with different substitutions at the carbon five of the oxazole ring (C-5, C-6, and C-7, **Figure 16**). A significant reduction of mild steel dissolution rate in molar hydrochloric acid solution is observed in the presence of these derivatives. The protective effect of synthesized oxazole compounds can be clearly revealed in **Figure 17**, in which i-E curve decrease is shown in the presence of these compounds as their concentrations rise (colored lines) compared to the blank solution (black line). Accordingly, the order of corrosion inhibition is as follows: C-6 (94.7%) > C-7 (85.9%) > C-5 (78.6%) at 10^{-3} M concentration. Using quantum chemical computations via the DFT-B3LYP/6-31G(d,p) method, the highest inhibition activity of the C-6 oxazole derivative is attributed to its great reactivity with the metal surface, which is induced by the benzene-1-sulfonate substituent. The presence of sulfur atom can cause the elevation of oxazole compounds adsorption process onto the metal surface, which reflects the good prevention capacity of these compounds.

In order to get great protection performance, the synthesis of new oxazole derivatives in which other azole-based core rings are incorporated has been reported. In this context, three new benzoimidazole/1,3,4-oxadiazole derivatives (C-1, C-2, and C-3, **Figure 18**) were reported as efficient organic inhibitors for mild steel dissolution

Figure 15.

Distribution of produced publication percentage related to the corrosion inhibition using oxazole-based compounds according to the Scopus® database.

Figure 16.
Chemical structures of newly synthesized benzo and 2-phenyl oxazole derivatives.

Figure 17.
Potentiodynamic curves of mild steel in 1 M HCl without and with synthesized 2-phenyl oxazole derivatives (C-6, C-7, and C-8, Figure 16) at different concentrations [112].

in acidic solutions [124–126]. These compounds exhibited an interesting effect in both sulfuric and hydrochloric acid solutions, which are largely used for the metal cleaning process in several industrial fields. It should be kept in mind that the higher reduction of corrosion rate caused by adding these inhibitors is obtained in hydrochloric acid than the sulfuric one, which reveals the possible effect of aggressive

Figure 18.
Chemical structures of examined 1,3,4-oxadiazole derivatives.

Figure 19.
SEM images of mild steel samples (a) before and after immersion in 0.5 M HCl solution, (b) without and (c) with C-2 benzoimidazole/1,3,4-oxadiazole derivative [115].

Figure 20.
Chemical structures of 1,2,4-oxadiazole derivatives with pyridinium substituents.

Figure 21.
Chemical structures of isoxazole derivatives.

media on the inhibition activity of used benzoimidazole/1,3,4-oxadiazole inhibitors. Moreover, the nature of considered acid can influence also the trend of recorded inhibition efficiencies, e.g., in HCl solution; the order is **C-1** ($\approx 92\%$) > **C-2**, while in H_2SO_4 one is **C-2** (75%) > **C-1** > **C-3** at the same concentration. Such conclusions are in good agreement with those of Bentiss et al. [128], which used other 1,3,4-oxadiazole derivatives (**C-4** and **C-5**, **Figure 18**). This means that the corrosive environments can influence the inhibition efficiency of oxadiazole compounds [90]. On the other hand, the substitution of a small carbon chain (e.g., ethyl in the case of **C-2**, **Figure 18**) by another one with the bigger size (e.g., propyl in the case of **C-3**, **Figure 18**) cannot usually induce an enhancement of the inhibition ability of oxazole-based inhibitors. The SEM images of mild steel surface in **Figure 19** confirm the efficacy of **C-2** derivative as an effective corrosion inhibitor.

A novel synthesizing procedure of 3,5-disubstituted 1,2,4-oxadiazole molecule was proposed by Outirite et al. [129]. By means of this procedure, three new 1,2,4-oxadiazole derivatives with pyridinium substituents (**Figure 20**) have been synthesized and reported as excellent corrosion inhibitors for C38 carbon steel in 1 M hydrochloric acid solution [130]. It is well-known that an increase of inhibitor concentration in the corrosive medium mainly leads to an enhancement of its prevention activity. Under this fact, the inhibition capacity of the latest listed derivatives was elevated by raising their amount in considered corrosive solution. On the other hand, the position of nitrogen atoms in pyridine substituents was shown not to have a notable influence on the anti-corrosion property of evaluated compounds. Nevertheless, a remarkable inhibition efficiency of around 95% was obtained at 8×10^{-4} M of synthesized 1,2,4-oxadiazole derivatives.

Several isoxazole-based molecules have also demonstrated noticeable protective performance for various metallic materials, such as $\text{Cu}_{90}\text{Ni}_{10}$ alloy and galvanized and mild steels, under different operating conditions. For instance, two new 5-phenylisoxazole derivatives have been developed and evaluated by Dominguez-Crespo et al. (**C-1** and **C-2**, **Figure 21**) [131]. According to experimental tests, 5-phenylisoxazole compounds exhibited great prevention effectiveness toward the degradation of galvanized steel and copper/nickel alloy. At 5 ppm as inhibitor concentration, the recorded prevention percentages are 100 and 93% for **C-2** and **C-1** in the case of galvanized steel, while in the case of $\text{Cu}_{90}\text{Ni}_{10}$ alloy they are 88 and 68% for **C-2** and **C-1** compounds. It is interesting to underline that achieved protection efficiencies are comparable to those of the commercial inhibitors (working under the same conditions). Another isoxazole derivative has been found to be an adequate inhibitor for mild steel in 1 M HCl aggressive solution [132]. The

Oxazole derivative	Metal	Medium	Inhibition efficiency (IE)
Mild steel	0.1 M H ₂ SO ₄	IE = 94%, IE was enhanced by synergistic combination with halides at 303 K and 12 × 10 ⁻⁴ M
C-1	Mild steel	1.0 M H ₂ SO ₄	IE: C-1 (78%) < C-2 (86%) < C-3 (89%) in 1.0 M H ₂ SO ₄ solution, IE: C-1 (71%) < C-2 (77%) < C-3 (83%) in 1.0 M HCl solution at 301 ± 2 K and 500 ppm
C-2		and 1.0 M HCl
C-3
Mild steel	1.0 M HCl and 0.5 M H ₂ SO ₄	IE = 93% in 1.0 M HCl solution, IE = 82% in 0.5 M H ₂ SO ₄ solution at 298 ± 1 K and 5 × 10 ⁻³ M

Table 3.
Relevant data related to the application of some oxazole-based compounds as corrosion inhibitors for mild steel.

molecular structure of the new synthesized derivative (C-3) is depicted in **Figure 21**. Both experimental and theoretical approaches pointed out that evaluated C-3 derivative acts as an effective corrosion inhibitor, in which its inhibition performance reaches 93% at 10⁻³ M.

Rather than employing oxazole derivatives, another novel strategy to enhance the anti-corrosion activity of these compounds is the use of its metal complexes. In the recent work, Najeeb [133] has reported the good performance of some metal complexes of a 1,3,4-oxadiazole derivative (C-6, **Figure 18**) against the corrosion of mild steel in 1 M HNO₃ medium. As core metal ions, Najeeb has tested Co(II), Ni(II), and Cu(II) ions. As a major outcome of this work, an increase of inhibition efficiency was observed via the metallic complexing process, and the following order of the inhibition efficiency is outlined: Co(II)-oxadiazole > Ni(II)-oxadiazole > Cu(II)-oxadiazole > oxadiazole. Moreover, the inhibition performance of these heterocyclic oxygen/nitrogen compounds can be synergistically enhanced by adding halide ions into the inhibition systems [134].

As was revealed in the literature, many other oxazole-based derivatives have recently stated as good anti-corrosion compounds for several metallic materials that immersed in different corrosive environments. **Table 3** illustrates the relevant data related to the use of some oxazole-based compounds as corrosion inhibitors [134–136].

5. Conclusion

In the current chapter, we focused on the application of azole-based compounds as inhibitor agents against metallic corrosion. Almost N-, N&S-, and N&O-azole-containing compounds were found to provide good protection property for numerous metal (or alloy)/medium systems. In this context, three main strategies were

adopted to enhance the capability of these compounds to inhibit the corrosion. The first one is based on the synergistic effect, in which supplementary additives (e.g., halide ions) are added into the corrosive media containing azole-based compound, while the chemical modification of azole molecular structures is the second strategy. The latest one is widely used and aimed to introduce further active sites of adsorption within these heterocyclic molecules. Recently, the metallic complex of azole compounds was also reported as an effective strategy to improve their prevention capacity. It is important to outline that N-azole compounds are extensively studied and reported as inhibitors for many metal/medium combinations in comparison with N&S- and N&O-azole ones. Consequently, more attention should be directed to examine the latest two-azole classes, especially oxazole-based compounds.

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