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A brief review of the performance of azole-type organic corrosion inhibitors

Milad Edraki[1], Milad Sheydaei[2,*], Davood Zaarei[1]

[1] *Department of Chemical and Polymer Engineering, South Tehran Branch, Islamic Azad University, P.O. Box 19585-466, Tehran, Iran*
[2] *Faculty of Polymer Engineering, Sahand University of Technology, P.O. Box 51335-1996, Tabriz, Iran*

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ABSTRACT

Whenever metals and metal-based materials interact with their environment (chemically, biochemically, or electrochemically), surface loss occurs, a process termed as corrosion. Corrosion is one of the important challenges which leads to the reduction of product life. In recent years, due to the growing interest of the world in protecting the environment and the harmful effects of using chemicals on ecological balance, the traditional approach with respect to corrosion inhibitors has gradually changed. Azole compounds have received a lot of attention in the oil, gas, and petrochemical industries since their structures prevent corrosion in acidic and alkaline environments. Also, due to having elements such as nitrogen and sulfur in their structure, they have been used widely as anti-fungal and anti-bacterial materials in the pharmaceutical industry as well. In this study, azole organic compounds, the encapsulation methods, and the mechanism of action of these compounds on the corrosion behavior of metallic surfaces have been investigated.

1. Introduction

Corrosion is a degradation process that occurs when metal and metal-based materials are exposed to moisture and oxygen [1]. Corrosion is one of the serious problems of metals which leads to a reduction in product life [2]. Metal-based materials are widely used in various industries such as shipbuilding, marine platforms, and jetties [3]. Therefore, corrosion inhibition is very important and can prevent significant losses. For example, in 2013, the loss due to corrosion was estimated at 2.5 trillion US$ [4]. Corrosion depends on several factors such as humidity, electrolytes, temperature, and pH, and can also accelerate the degradation process by combining chemical, biological, and mechanical factors [5,6]. Corrosion can have destructive effects on the environment and a direct impact on human health [7,8]. When a structure is placed in a corrosive environment, it tends to experience localized corrosion of crevice, pitting, and microbial-induced corrosion [9,10]. Coatings are one of the best approaches for corrosion control [11,12]. However, most of the current coatings are based on volatile organic compounds and have adverse consequences for the environment and human health [13]. Nanomaterials have been investigated for reducing corrosion as they enhance thermal, mechanical, physical, and optical capabilities, subsequently tuning the surface and interaction areas [14]. Corrosion additives enhance the performance of coatings, but since the discovery of their toxicity, efforts have been made to find their replacement with environmentally friendly properties [15]. For this reason, the use of natural additives such as plant extracts has received much attention [16,17]. In this study, azole-type organic corrosion inhibitors and the research done in this field are discussed.

2. Corrosion inhibitors

A corrosion inhibitor is a chemical compound that can reduce the rate of corrosion after being in an environment with a low concentration. Many factors are important for the use of an inhibitor such as availability, price, and toxicity, hence many inhibitors are contraindicated [11,18]. Inhibitors are classified and one of them is used depending on the environment used and the purpose, in general it can be said that they

* Corresponding author. Tel.: +989211868265; e-mail: mi_sheydaei@sut.ac.ir, m.sheydaei@yahoo.com

are classified into five classes: oxidizing, non-oxidizing, organic, metal cations, and pigments [18]. Although they can also be considered based on whether the inhibitor reduces which half of the reaction (anodic, cathodic, or both) [11,16]. When using anodic inhibitors, an important issue that should be noted is critical concentration. If the concentration of anodic inhibitor is low, it causes a passivation film to be created that is not able to cover the anodic surface, as a result, more local attacks occur on the uncoated surface [19]. This event will not happen in the case of using cathodic inhibitors, even if the partial covering of cathodic sites leads to less corrosion rate and as a result, less metal dissolution in the anode (positive pole) [18]. Some coatings are self-healing, such as polysulfide polymers. In fact, this is due to the relatively labile and dynamic covalent characteristics of disulfide, which induces self-healing properties for polysulfide polymers [20-29]. Figure 1 shows the performance of a green self-healing compound as well as corrosion inhibitors.

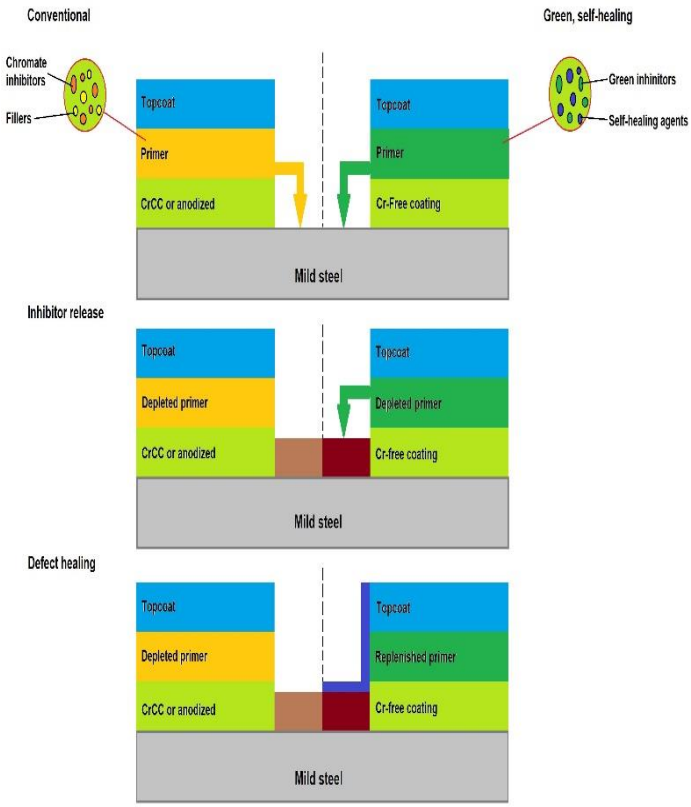

Figure 1. Schematic illustration of healing in common coatings (left) and green coatings (right).

In a normal coating, the first layer is a conversion layer [30]. The inhibitor is present in the upper primer and is released when a defect occurs and protects the metal surface [11,12,30]. Many plants such as matcha and ginger have anticorrosion and antimicrobial properties due to their bioactive components (see Figure 2) [11,31-40].

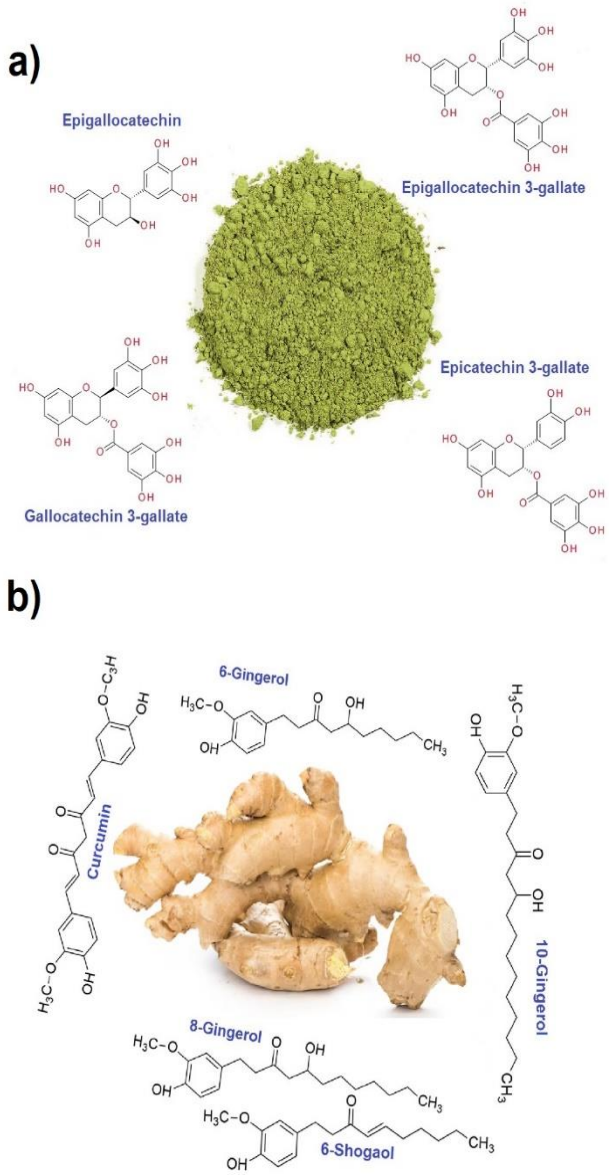

Figure 2. The most important compounds in matcha (a) and ginger (b).

The primer can be modified, in fact, self-healing agents and corrosion inhibitors are added to the primer. In case of defects in the coating, not all agents are always released, some remain and can protect the coating in the event of a defect occurring again [30]. The protective performance of the inhibitor depends on the content of the inhibitor reaching the uncovered areas, so the release is very important [41]. Release depends on how the inhibitor is placed in the coating and how it leaches. Also, homogeneous distribution is one of the important factors so that the inhibition properties are uniform. Moreover, the speed of leaching should be furthermore considered. Although the rapid release is important in providing protection, it is ineffective in long-term corrosion prevention [41]. The

release of the inhibitor in the coating can occur through water contact with the coating. Encapsulation is one of the methods used to release the inhibitor, in fact, in this method, the release can be controlled [41,42]. The change in pH is a very important factor that can lead to a change in the solubility of the inhibitor in the coating, and as a result, a change in the release is observed [41]. The thickness of the coatings used to protect metals is usually several micrometers, so the use of capsules with micro and nano dimensions is very practical and possible [43]. Different methods can be discussed for adding inhibitors in coating formulations, but usually, encapsulation methods for including inhibitors with low solubility in coatings are suggested [44]. For example, we can refer to urea-formaldehyde microcapsules that are spheres and contain corrosion-inhibiting compounds [44,45]. When any scratch or other defect damages the coating, the microcapsule breaks and the inhibitor is released [45]. Another method is using double-layer hydroxide, which is composed of metal hydroxide layers. The entry of metal cations with valence electrons into these layers causes them to be charged, and as a result, the created charge is compensated by the anionic compounds inserted between the channels of these layers [44-46]. After that, anionic inhibitors can be inserted between double-layer hydroxide layers. Another way of nano-encapsulation is using silica nanocapsules [47]. Silica nano capsules covered layer by layer were synthesized by poly electrolyte layers and inhibitor layers. These protectors release corrosion inhibitors in case of pH changes. Also, the inhibitor can be incorporated into silica nanoparticles using *in situ* synthesis of porous silica shells starting from an oil-in-water emulsion [47,48]. In this method, the inhibitor is dissolved in the organic phase, and the synthesis of nanocapsules and the addition of the inhibitor are done in one step. The inhibitor is encased in a porous nanosphere, actually slowing the release of the inhibitor so that long-term corrosion protection is provided [47].

3. Organic azole corrosion inhibitors

One of the important compounds of organic molecules is heterocyclic, that have many applications in different fields and has a special place in different sciences. Due to the different frameworks of these compounds in natural products as well as their wide applications in materials science, medicinal chemistry, and agrochemicals, they have a special place [49]. Among them, azoles are very important. In fact, azoles are a broad class of five-membered heterocyclic aromatic compounds whose framework contains one to five nitrogen atoms, although it can contain at least one S or O atom as part of the azole-conjugated ring [50]. Oxadiazoles, oxazoles, and isoxazoles are examples of various oxygen-containing azoles that have been widely studied in the field of biological activities [49-51]. Azole derivatives contain valuable properties such as anti-fungal, anti-diabetic, immunosuppressive, anti-inflammatory, anti-viral, anti-tuberculosis, and anti-cancer activities, and also have excellent oral bioavailability and chemical stability [52-56]. In addition to the properties of azoles, they are used as scaffolds for various fields including materials, energy, and catalysis [57]. Organic heterocyclic molecules have anti-corrosion compounds for many metal systems, especially heterocyclic molecules that contain nitrogen, oxygen, and sulfur atoms [52,57]. Figure 3 shows the molecular structure of some of them that are used as corrosion inhibitors [58]. Mahdavian *et al*. [18] investigated the corrosion inhibition performance of 2-mercaptobenzoimidazole and 2-mercaptobenzoxazole compounds for mild steel in a 1 molar hydrochloric acid solution. The results showed that by increasing the concentration of inhibitors to 1 mM, the double-layer capacitance and corrosion current density decreased compared to mild steel without inhibitors, but the charge transfer resistance and corrosion potential increased. Also, 2-mercaptobenzoimidazole had better results than 2-mercaptobenzoxazole.

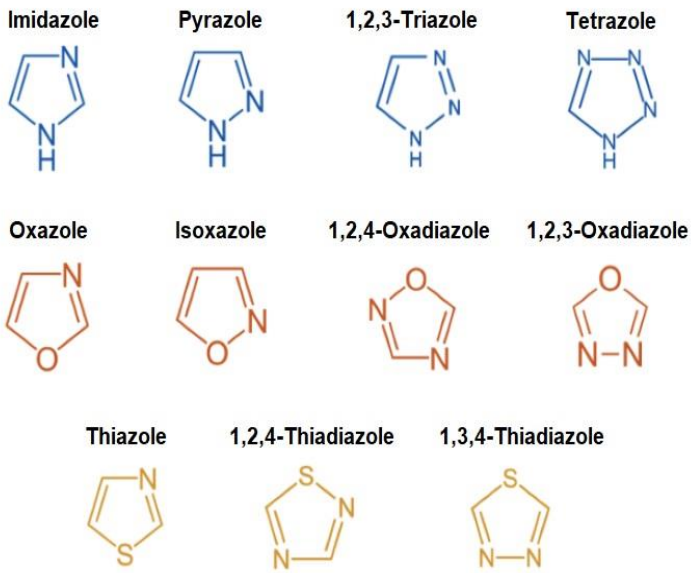

Figure 3. Molecular structures of the core rings of some azole-based compounds.

Wang *et al*. [59] investigated the corrosion behavior of mild steel in a 0.5 molar hydrochloric acid environment using 1,4-bis-benzimidazolyl-butane (BBB). The results showed that by increasing the concentration of BBB inhibitor, the corrosion rate decreased significantly. Solehudin *et al*. [60] assessed the inhibition performance of benzotriazole with 0.1-5 mM/L concentrations on the corrosion behavior of carbon steel in 3.5 *wt*.% of sodium chloride solution. The results showed that the carbon steel without inhibitor showed a corrosion rate of 1.58 mm/th, a

corrosion current density of 136.52 μA/cm^2, and a polarization resistance of 6.12 Ω, while with the increase of the inhibitor, the corrosion rate and corrosion current density decreased to 0.1 mm/th and reached 9.02 μA/cm^2. In the end, the polarization resistance and the inhibition efficiency were increased, both reaching 98.28 Ω and 93%.

Jafari *et al*. [61] examined the inhibition effect of 2-mercaptobenzothiazole and 2-aminobenzothiazole compounds on carbon steel in a 1 molar hydrochloric acid solution. They observed that both of these compounds increased the corrosion resistance of the samples as the immersion time increased from 15 to 300 min, and at the same immersion time, 2-mercaptobenzothiazole had better corrosion inhibition compared to 2-aminobenzothiazole. Marathe *et al*. [62] loaded 2-mercaptobenzoimidazole and 2-mercaptobenzothiazole in urea-formaldehyde microcapsules by using in-situ polymerization and dispersed it as an anti-corrosive pigment in polyurethane (PU). The corrosion resistance of PU coating against 0.5 molar hydrochloric acid solution was evaluated. The results showed that the PU coating containing 2-mercaptobenzothiazole microcapsules has better inhibition efficiency than the PU coating containing 2-mercaptobenzoimidazole microcapsules. Hegazy [63] investigated the effect of a cationic surfactant based on tolyltriazole on the corrosion behavior of carbon steel in a 7-molar phosphoric acid solution. The results showed that with increasing inhibitor concentration, the corrosion rate decreases and inhibition efficiency increases. Singh [64] investigated the corrosion behavior of 304 stainless steel in a 1.0 molar sulfuric acid solution by using 2-mercaptobenzothiazole (organic inhibitor) with concentrations of 100 to 400 ppm. The results showed that by increasing the inhibitor concentration to 400 ppm (at 20 °C), the inhibition efficiency value reached 97%. But, as the temperature increased to 40 °C, the inhibitory efficiency decreased to 80.7% at the same concentration.

Kermannezhad *et al*. [65] modified amino propyl-functionalized mesoporous nanosilica with 2-mercaptobenzoxazole and evaluated its effects on the corrosion behavior of steel and copper in a 3.5 *wt*.% of sodium chloride solution. The results showed that by increasing the concentration of this pigment to 1.32 mM, the corrosion rate and corrosion current density of steel and copper reduced and their corrosion potential increased. Also, inhibition efficiency for copper and steel reached 85.6 and 51%, respectively. Radovanovic *et al*. [66] investigated the effect of 2-Mercapto-1-methylimidazole inhibitors on brass in a 0.1 molar di-sodium tetraborate over the span of 1-180 min. The results showed that by increasing the concentration of inhibitor to 1×10-2 mol/dm^3 and after 180 min, the corrosion current density reduced and reached 0.3 μA/cm^2, and inhibition potential increased to -0.167 V. Finally, the inhibition efficiency was 94.2%.

Edraki *et al*. [67] used sodium montmorillonite clay nanoparticles as a carrier of 2-mercaptobenzimidazole and 2-mercaptobenzothiazole organic corrosion inhibitors and evaluated their anti-corrosion performance on mild steel in the salt solution phase. The results showed that the nanoparticle containing 2-mercaptobenzothiazole had higher anti-corrosion performance.

Edraki *et al*. [68] modified sodium montmorillonite clay nanoparticles by organic inhibitors 2-mercaptobenzimidazole and 2-mercaptobenzothiazole and investigated them as anti-corrosion nanopigments in epoxy disperse resin and then on mild steel. The results showed that the epoxy coating containing 3 *wt*.% of 2-mercaptobenzothiazole compared to the epoxy coating containing 3 *wt*.% of 2-mercaptobenzimidazole had higher anti-corrosion performance and also the presence of nanopigments increased the hydrophobicity of the surface.

Bahrani *et al*. [69] studied the effect of loading organic imidazole and inorganic zinc cation inhibitors in the layered talc pigment structure on the corrosion protection performance of epoxy ester coating. The results showed that the cheap and available talc pigment, which does not have much anti-corrosion effects, can create active corrosion protection effects in the coating by loading the corrosion inhibitor.

Kashani *et al*. [70] synthesized nanocapsules based on PU as the shell and organic corrosion inhibitor 2-mercaptobenzimidazole as the core and then investigated it in dispersed silane hybrid coatings and on aluminum surfaces. The results showed that the coating containing 0.1 g of the aforementioned synthesized nanocapsules showed the best anti-corrosion and self-healing performance.

4. Conclusion

To sum up, organic azole compounds are used as inhibitors and sometimes act as anodes, cathodes, or both. Therefore, as a general rule, cathodic and anodic inhibitors act through the process of adsorption and create a layer. Naturally, the occurrence of molecules that have a strong tendency towards metal surface compounds is indicative of good inhibition efficiency and low environmental risk. These absorbed inhibitor molecules create a protective hydrophobic layer on the metal surface which causes a barrier against the dissolution of metal in electrolyte and increased the resistance against corrosion.

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