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ORGANIC CORROSION INHIBITORS

SYNTHESIS, CHARACTERIZATION,
MECHANISM, AND APPLICATIONS

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Organic Corrosion Inhibitors

Organic Corrosion Inhibitors

Synthesis, Characterization,
Mechanism, and Applications

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Preface

Corrosion is a highly dangerous phenomenon that causes huge economic and safety problems. Various methods of corrosion monitoring, including cathodic protection, painting and coatings, alloying and dealloying (reduction in metal impurities), surface treatments, and use of corrosion inhibitors have been developed depending upon the nature of metal and environment. Application of organic compounds, especially heterocyclic compounds, is one of the most common, practical, easy, and economic methods of corrosion mitigations. Obviously, these compounds become effective by adsorbing on the metallic surface using electron-rich centers including multiple bonds and polar functional groups. These electron-rich centers act as adsorption sites during their interaction with the metallic surface. Along with acting as adsorption sites, the polar functional groups such as $-OH$ (hydroxyl), $-NH_2$ (amino), $-OMe$ (methoxy), $-COOH$ (carboxyl), $-NO_2$ (nitro), $-CN$ (nitrile), and so on also enhance solubility of organic compounds in polar electrolytes. Present book describes the collection of major advancements in using organic compounds as corrosion inhibitors including their synthesis, characterization, and corrosion inhibition mechanism.

Through this book it can be seen that use of organic compounds serves as one of the most effective, economic, and ease methods of corrosion monitoring. Using previously developed methods, 15% (US \$375) to 35% (US \$875) of cost of corrosion can be minimized. Different series of organic compounds, including heterocyclic compounds, are effectively used as corrosion inhibitors for different metals and alloys in various environments. Because of the increasing ecological awareness and strict environmental regulations, various classes of environmental-friendly alternatives to the traditional toxic corrosion inhibitors have been developed and being implemented. These series of compounds mostly include carbohydrates, natural polymers and amino acids (AAs), and their derivatives. Corrosion scientists and engineers strongly believe that these environmental-friendly alternatives will be capable to replace, in the near future, the toxic marketable products that are still being used via many worldwide industries.

A book covering the recent developments on using organic compounds as corrosion inhibitors is broadly overdue. It has been addressed by Drs. Verma, Hussain, and Ebenso in this book which attends to fundamental characteristics of organic corrosion inhibitors, their synthesis and characterization, chronological growths, and their industrial applications. The corrosion inhibition using organic compounds, especially heterocyclic compounds, is broad ranging. This book is divided into five sections, where each section contains several chapters. Section 1 “Basics of corrosion and prevention” describes the basic of corrosion, experimental and computational testing of corrosion, and a comparison between organic and inorganic corrosion inhibitors. Section 2 “Heterocyclic and non-heterocyclic corrosion inhibitors” describes the collection of different series of heterocyclic and non-heterocyclic corrosion inhibitors such as amines, imidazole, quinoline, pyridine, indole, and their derivatives. This section also includes organic compounds as corrosion inhibitors for oil and gas industries.

Section 3 “Organic green corrosion inhibitors” entirely focuses on green corrosion inhibitors. This section describes the corrosion inhibition characteristics of carbohydrates, amino acids (AAs), oleochemicals, chemical medicines, ionic liquids (ILs), and their derivatives. Section 4 “Organic compounds based nanomaterials as corrosion inhibitors” describes the corrosion inhibition properties of carbon nanotubes (CNTs: SWCNTs and MWCNTs), graphene oxide (GO), and their composites. In the end, Section 5 “Organic polymers as corrosion inhibitors” gives a description on natural and synthetic polymers as corrosion inhibitors.

Overall, this book is written for scholars in academia and industry, working corrosion engineering, materials science students, and applied chemistry. The editors and contributors are well-known researchers, scientists, and true professionals from academia and industry. On behalf of Wiley, we are very thankful to authors of all chapters for their amazing and passionate efforts in making of this book. Special thanks to Prof. M. A. Quraishi, who guided us continuously in drafting of this book. Special thanks to Michael Leventhal (acquisitions editor) and Katrina Maceda (managing editor) for their dedicated support and help during this project. In the end, all thanks to Wiley for publishing the book.

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SciVal Insights Report (2010–2015), he has a citation impact 10% above world average: second most prolific author in the field of corrosion inhibition worldwide and fifth most downloads of his publications globally in the field of corrosion inhibition. His Google Scholar Citations since 2013 is over 8000 with an H-index of 64 and i10-index of 216. His RESEARCHERID account shows H-index of 44 with total citations of 5779 and average citation per article of 24.78. He is also a B3 NRF Rated Scientist in Chemistry (South African National Research Foundation). INTERPRETATION – B3: Most of the reviewers are convinced that he enjoys considerable international recognition for the high quality and impact of his recent research outputs. He is a member of International Society of Electrochemistry, South African Chemical Institute (M.S.A. Chem. I.), South African Council for Natural Scientific Professions (SACNASP) (Pri. Sci. Nat.), Academy of Science of South Africa (ASSAf), and a fellow of the Royal Society of Chemistry, UK (FRSC).

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4

Organic and Inorganic Corrosion Inhibitors: A Comparison

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4.1 Introduction

Corrosion is one of the important challenges in the contemporary world, especially, after the introduction of technology into our lives following the industrial revolution, though it has been known and some techniques used to prevent it since the ancient Greek period. Furthermore, following the rapid development of informatics, aviation, and aerospace technology for the last 70 years, it is very important to design the materials used according to suitable environmental conditions, as well as to protect the materials used and to extend their durability periods. In the civilizing world, it is not enough to determine the most effective method, condition, or anticorrosion materials in order to prevent or delay corrosion, but also it is getting important that they are less harmful to the environment, with lower toxicity, and can be obtained from renewable resources, instead of existing natural resources. In this context, organic corrosion inhibitor material design has been very promising in many production areas besides the classical inorganic corrosion inhibitors.

Metal corrosion is known as metal erosion because of chemical or mostly electrochemical reactions when a metal comes into contact with surrounding materials. It is also known that the natural corrosion process is generally an electrochemical phenomenon and in this type of corrosion, it is known that the oxidation process is facilitated by the presence of an electron acceptor known as a suitable depolarizer according to the equation $M \rightarrow M^{n+} + ne^{-}$. In addition, although there are many types of metal corrosion and classification of them [1], the main types of metal corrosion [2] can be given as follows:

1. Uniform (general) corrosion
2. Galvanic (two-metal) corrosion
3. Thermogalvanic corrosion
4. Crevice corrosion (including deposit corrosion)
5. Pitting corrosion
6. Selective attack, selective leaching (de-alloying)
7. Intergranular corrosion (including exfoliation)
8. Erosion corrosion
9. Cavitation corrosion
10. Fretting corrosion
11. Stress corrosion cracking
12. Corrosion fatigue

In the contemporary world, considering the environmental and economic damages caused by corrosion, it is very important to determine the source of corrosion and to take measures to prevent or at least slow it down: the main ways to slow down and/or preventing corrosion can be summarized as follows.

1. Metal type
2. Protective coating
3. Environmental measures
4. Sacrificial coatings
5. Corrosion inhibitors
6. Design modification

Among these protecting ways, the material selection and design is the simplest way to control the corrosion but in cases where there are not always many options for material selection and micro design, different methods are known to be applied to prevent or slow down corrosion. However, in the liquid phase and atmosphere, the corrosion inhibitors can be often enforced to improve the heat-exchange efficiencies to reduce the corrosion. In this case, the electrochemical methods – anodic, cathodic, or both – have provide a wide profit in terms of the economics and saving the natural sources [3]. Material scientists have commonly exploring the usefulness of the alloys in addition to heat treatment regimes and protective coatings to reduce or prevent the corrosion [3].

Besides, the surface treatments, which are reactive, applied, and biofilm coatings in addition to the anodization, are used to slow down and protect the corrosion. For instance, the use of painting or enamel protects the material from corrosion by creating a corrosion-resistant barrier between the damaging environment and the structural material. In this case, the use of chemicals that form an electrically insulating or chemically impervious coating on exposed metal surfaces to suppress electrochemical reactions make the system less susceptible to scrapes or imperfections in the coating because extra inhibitors can be present wherever the metal is exposed.

Nowadays, organic and inorganic corrosion inhibitors, whether natural or artificial, are increasingly important. Because of the great importance of them, in this section, the organic and inorganic corrosion inhibitors are presented considering the molecular structures, main characteristics, and environmental effects.

4.2 Corrosion Inhibitors

Corrosion inhibitors are known to be used in countless fields of both production and commercial areas such as pipelines, cooling systems, refinery units, water treatment, painting, oil refinery units, and so on [4]. A basic classification of corrosion inhibitors is given in [Figure 4.1](#).

4.2.1 Organic Corrosion Inhibitors

Organic corrosion inhibitors reduce and protect metal or alloy metal dissolution in aggressive environment by forming a thin film layer with adsorption process on the target surface. The adsorption of the compounds on the target surface can be physical, chemical, or both ways based on the type and properties of both the organic compound and target surface in addition to the environmental conditions of the corrosion happened in [5, 6]. The kinetic and/or thermodynamic searches on adsorption mechanism of the corrosion inhibition of the organic compounds have showed that the performance of the corrosion inhibitor have been mostly related to the surface coverage of the compounds depending on the structural properties [7]. It is also well known that heterocyclic compounds containing N, S, O, and P atoms, as well as compounds containing π -bonds and/or polar group that will allow electron delocalization, they provide protection of metals or alloys from corrosion. In the literature, it has been reported in the experimental studies

that heterocyclic compounds such as azoles, indoles, and aromatic rings, as well as open-chain organic-based compounds such as epoxy and polymeric systems [8–10] can be successfully used against metal and/or alloy corrosion. In this context, it is important to address the major groups of organic compounds that provide corrosion protection and/or retarding properties.

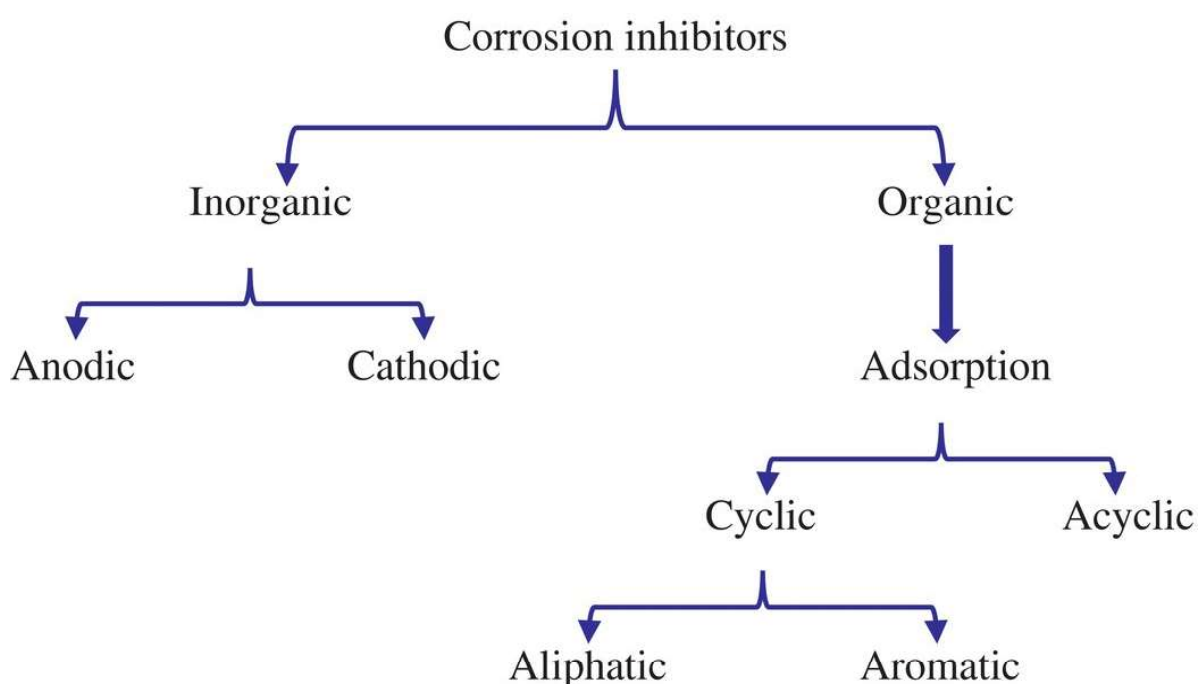


Figure 4.1 A basic classification of the corrosion inhibitors.

4.2.1.1 Azoles

Azoles are a class of five-membered aromatic heterocyclic compounds containing at least one nitrogen atom and have an aromatic structure. They are named according to the number of nitrogen atoms in the aromatic ring and their position. In addition, there are other heteroatom-containing azo compounds [11–13] such as O and S in their structure, and the main structures are shown in the [Figure 4.2](#). The aromatic structures of azo compounds and the heteroatoms they contain cause an increase in polarity of these

compounds due to electron delocalization in compounds, which make these compounds very useful materials for inhibition of corrosion. Srivastava et al. [14] have recently reported the efficiency of a benzo[d]imidazole derivative green corrosion inhibitor has a very good inhibition capability (>98%) on the carbon steel at optimum dosage conditions and continued the strong inhibition adsorption (>95%) even at 333 K. The inhibition ability of pyridine thiazole compound on copper corrosion in acidic medium has been investigated by a series of experiments techniques (EIS, SEM, AFM), and shown by the EIS technique that the maximum inhibition capacity is reached at 94% at 1 mM concentration [15]. Recently, the inhibition capabilities of a series of imidazole derivatives on the corrosion of mild steel in acidic conditions have been investigated by electrochemical, thermodynamic, and also computational techniques; one of the most important results of this investigation is that the imidazole derivatives provide the anodic protection of mild steel and promote cathodic hydrogen reactions as well [16]. Besides, phenanthroimidazole derivatives [17] and bis-benzothiazoles derivatives [18] have a very high inhibitory potency for mild steel and showed a mixed type inhibitor capacity by TAFEL diagrams, but the priority for the cathode.

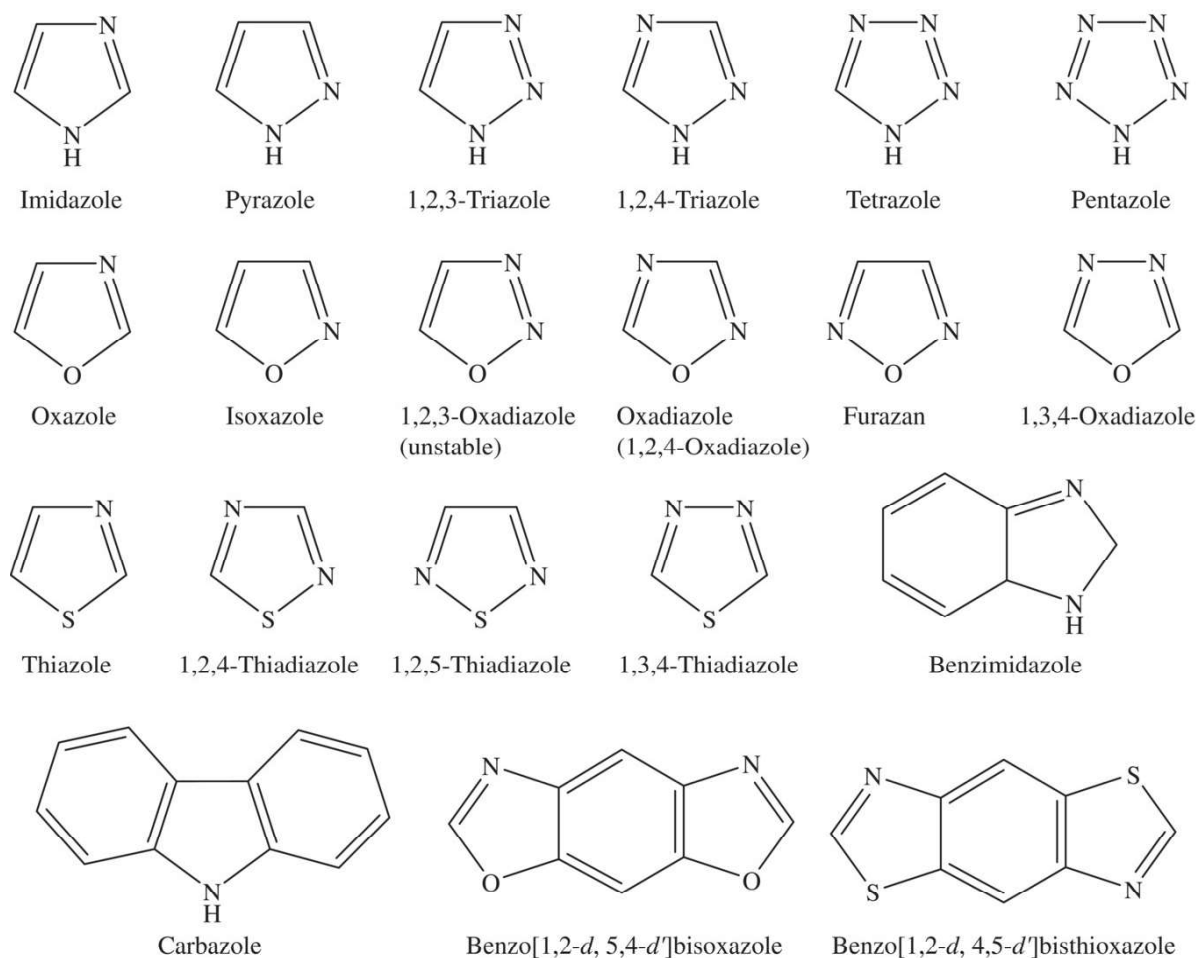


Figure 4.2 The chemical structures of the main azole compounds.

4.2.1.2 Azepines

Azepines with seven-membered as a subgroup of heterocyclic organic compounds and their effects are generally well known in medicinal fields [19] (Figure 4.3). Potential use of waste drugs as corrosion inhibitors has received increasing attention in recent years. In 2009, Arslan et al. suggested that drug molecules with quantum chemical methods can show inhibitory properties for mild steel in an acidic environment [20]. It has also been reported that different groups of heterocyclic compounds used in many medical fields are promising as

environmentally friendly corrosion inhibitors [21]. For a series of triazoloazepine derivatives, the corrosion and protection constants on steel 45 with a concentration of 1 M have been evaluated and suggested that Cl atom substitutions make the main structure more potent against corrosion among all substituents (H, F, Cl, I, CH₃O, and CH₃ on different position of the molecule) [22]. In addition, the inhibitory characteristics of acetonitrile and secondary amines containing triazoloazepine ring for carbon steel corrosion in HCl and hydrogen sulfate have been investigated and reported that the reciprocal effects of triazoloazepine and p-tolyl groups in the secondary amines provide a maximum synergetic effect during inhibition of steel corrosion in the HCl [23]. In the past, the corrosion inhibition properties of the carbamazepine unused drug on steel in different solvent environments have been examined, and the inhibition efficiency of the carbamazepine has been determined as 85% [24]. Besides, carbamazepine has effective role in both anodic processes and cathodic hydrogen evaluation, and thus the amount of carbamazepine does not affect the corrosion potency [24].

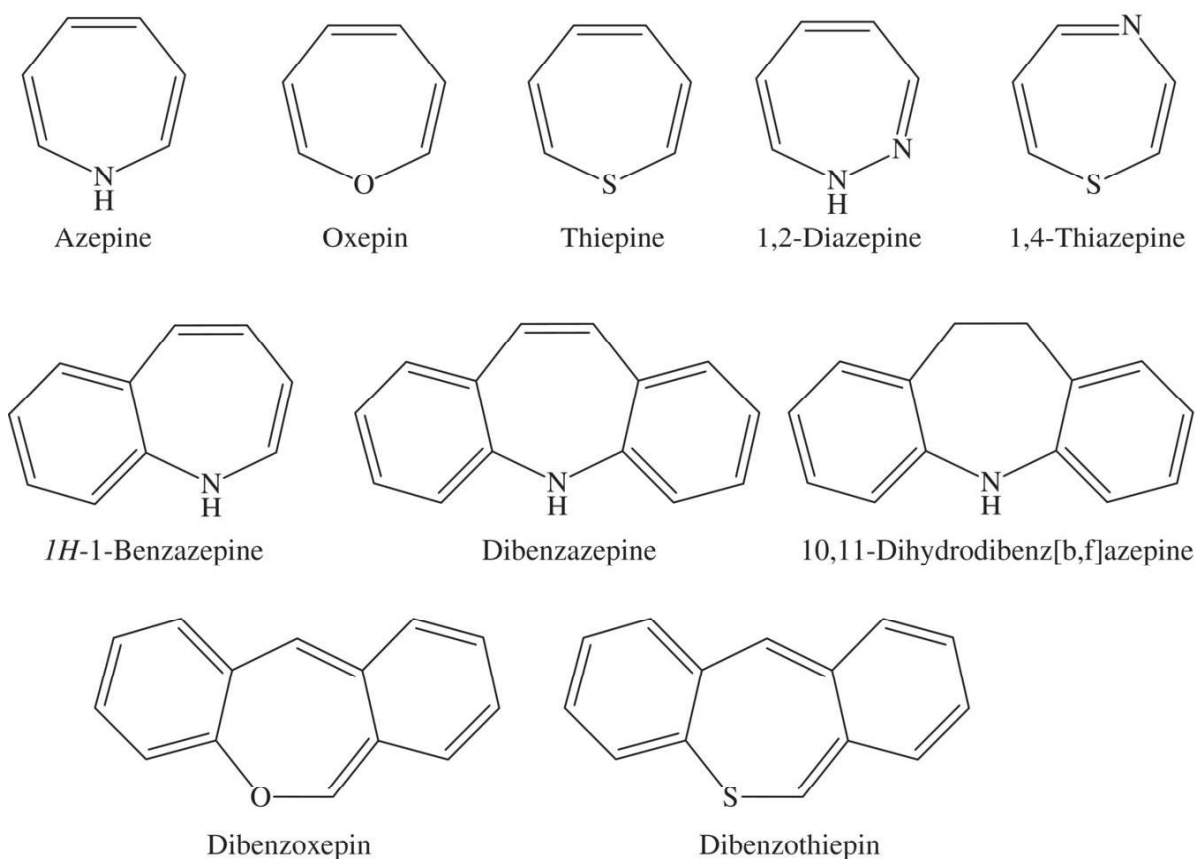
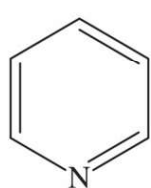


Figure 4.3 The chemical structures of the main azepine compounds.

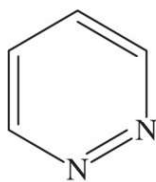
4.2.1.3 Pyridine and Azines

Pyridine is a heterocyclic organic compound shown by the simple structural formula C_5H_5N and is widely used in many production areas due to its aromatic structure [25] (Figure 4.4). The lone pair of the nitrogen atom in the structure has sp^2 hybridization but does not contribute to the aromaticity of the compound as it lies outside the plane of the ring like sigma bonds and facilitates bond formation with an external system through an electrophilic attack. On the other hand, Azines are an organic compound group with the functional group $RR'C = N-N = CRR'$, obtained by the condensation of hydrazine with ketones and aldehydes [26]. In the past, many papers have been reported on

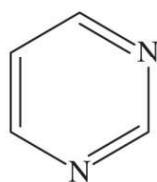
pyridine and its derivatives and suggested that they have sufficient potency for inhibition of iron, aluminum, carbon steel, N80 steel, mild steel, and zinc in different acidic media [27-34]. Furthermore, Kurmakova and coworkers [35], with empirical and theoretical support, have evaluated the effects of quaternary pyridinium, imidazopyridinium, and imidazoazepinium salts for the inhibition of biocorrosion of steel; they have suggested that the observed inhibition capability of the salts have been largely proportional to both the polarizability and energy gap values of the salts. In addition, a lot of papers also have reported that azine derivatives have been potential corrosion inhibitors for mild steel [36-39], carbon steel [40], iron [41] and copper [42], an acidic environment. In a recent study, it has been suggested that the newly synthesized polybenzoxazine compound is promising as a highly corrosion-resistant material for mild steel and similar alloys under different conditions, including marine conditions [43].



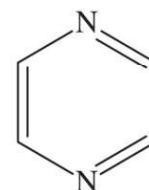
Pyridine



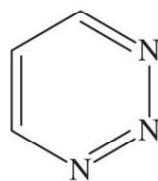
Pyridazine (1,2-diazine)



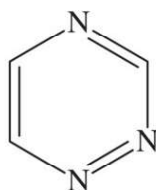
Pyrimidine (1,3-diazine)



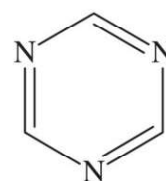
Pyrazine (1,4-diazine)



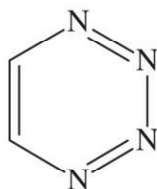
1,2,3-Triazine



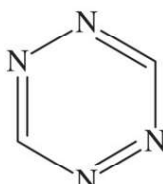
1,2,4-Triazine



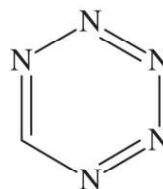
1,3,5-Triazine



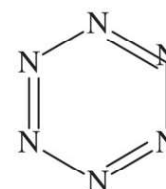
1,2,3,4-Tetrazine



1,2,4,5-Tetrazine

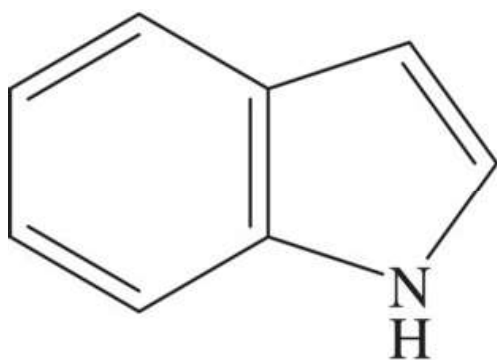


Pentazine

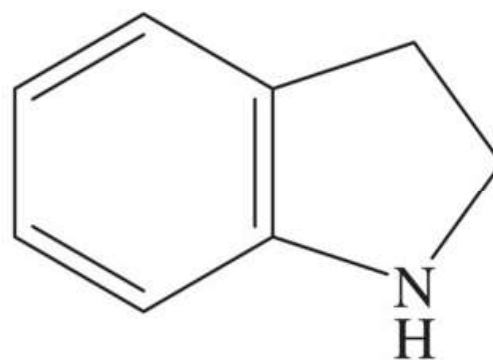


Hexazine

Figure 4.4 The chemical structures of the main pyridine and azine compounds.



Indole



Indoline

Figure 4.5 The chemical structures of the main indole compounds.

4.2.1.4 Indoles

Indoles having a formula C_8H_7N has a bicyclic structure, consisting of a six-membered benzene ring fused to a five-membered pyrrole ring, which provides the usefulness in many production and manufacturing sectors, as well as in the medicinal importance of them [44, 45] (Figure 4.5). In the past, the inhibition capabilities of the indole-based melatonin compound at 30 and 60 °C with a concentration of 10 mM on the steel surface have been determined [46] as 98.3 and 88.6%. In addition, the corrosion efficiencies of 12-(2,3-dioxindolin-1-yl)-N, N, N-trimethyldodecan-1-ammonium bromide [47] and 1-(2-hydroxyethyl)-2-imidazolidinone [48] compounds on mild steel have been determined as 95.9% (in 1 M HCl) at 298 K and 85.4% (in 0.5 M HCl) at 293 K, respectively. In addition, the inhibition efficiency of 4-((1H-indol-3-yl) methyl) phenol (IMP) and 4-(di (1H-indol-3-yl) methyl) phenol compounds to protect copper corrosion at 2 mM concentration has been determined [49] to be 99.3 and 97.5%, respectively. In another study, the inhibition potency of new synthesized 3,3-((4-(methylthio)phenyl)methylene)bis(1H-indole) compound on copper corrosion has been examined; it has been suggested to have a good corrosion inhibition capability based on the electrochemical techniques; the adsorption process of the compound on copper surface has been contributed by both physisorption and chemisorption [50].

4.2.1.5 Quinolines

Quinoline is a compound with the formula C_9H_7N , usually soluble in organic solvents. Although it has little application area with its simple form, its derivatives have a wide range of applications in the literature, which from basic sciences to industrial engineering [51–56] (Figure 4.6). Besides, quinoline derivatives containing highly polarizable groups such as -OH, -OMe, -NH₂, and so on in addition to having

with both nitrogen atom and π -electronic system, it facilitates interaction with the metal surface, making this group of compounds very important among anticorrosive materials, especially in green corrosion inhibitors [57]. Recently, the anticorrosive behavior or corrosion inhibition potency of “oxoquinolinecarbohydrazide N-phosphonate” [58], “8-hydroxyquinoline” [59-61], “N,N'-((ethane-1,2-diylbis(azanediyl))bis(ethane-2,1-diyl))bis(quinoline-2-carboxamide)” [62], “5-{[(4-dimethylamino-benzylidene)-amino]-methyl}-quinolin-8-ol” [63] derivatives on mild steel in acidic medium has been extensively studied. In a recent work, the inhibition capability of quinoline derivative “5-benzyl-8-propoxyquinoline” [64] on Q235 steel in sulfuric acid medium has been determined as 97.7%, and it was shown that the inhibition efficiency has quite high even at high concentrations and temperatures. Furthermore, in another comprehensive study on mild steel in 15% HCl solution, the authors have found that the inhibition potential of quinoline derivative “dibenzylamine-quinoline” [65] has reached its highest value at 363 K with 95.4%, which is quite sufficient to corrosion inhibitor of oil and gas acidification. Mohammadloo and coworkers have investigated the possible usage of “8- hydroxyquinoline” [66] in smart coating applications and have suggested that intelligent corrosion detection can be achieved using 8-HQ as corrosion indicator and inhibitor, based on results obtained from fluorescence microscope.

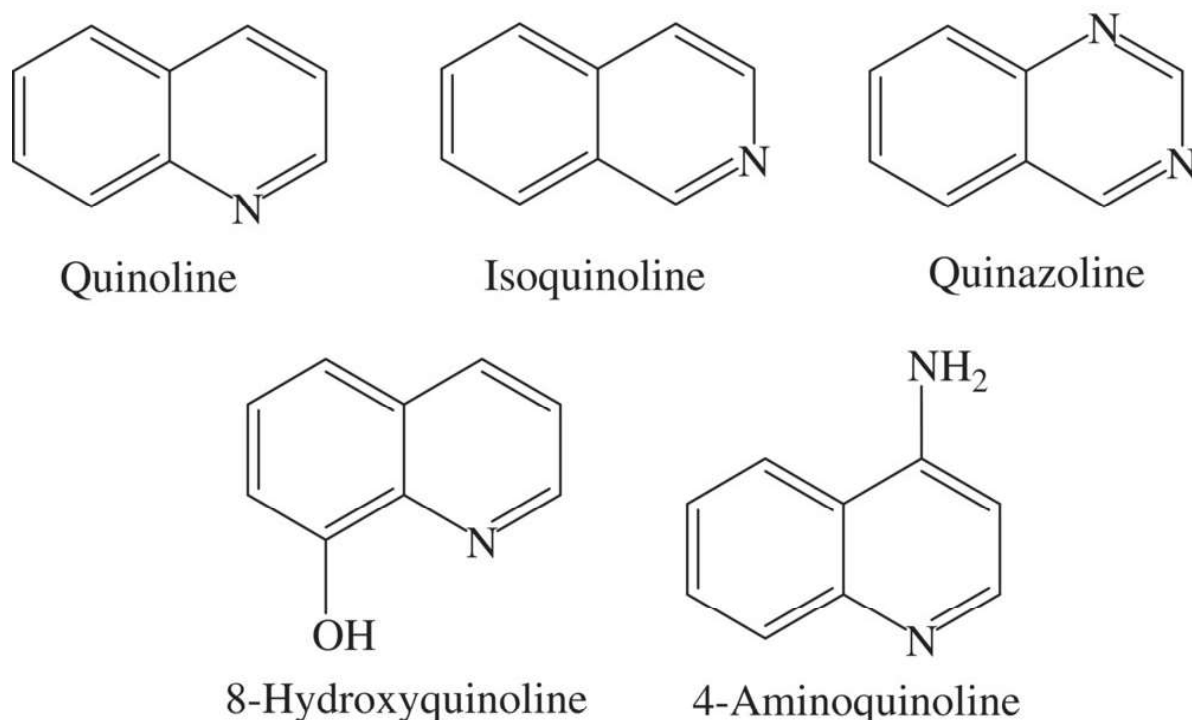


Figure 4.6 The chemical structures of the main quinoline compounds.

4.2.1.6 Carboxylic Acid and Biopolymers

As known well, composites and conductive polymers are adsorbed by metal surfaces, suppress the dissolution process, and provide the formation of a protective film for corrosion. In this context, the advantage of polymers and composites over other protective coatings such as paint is that they are environmentally friendly since they do not contain toxic substances [67–70]. In this context, Ateş and Özyılmaz [68] obtained polycarbazole, polycarbazole/nanoclay, and polycarbazole/Zn-nanoparticles film layers by chemical and electrochemical techniques and investigated the corrosion inhibition properties of them on SS304 in saltwater. In their study, they determined that the protection efficiency of the film layers polymerized chemically (PE: 99.81% for PCz, 99.46% for PCz/nanoclay and 99.35%, PCz/Zn-nanoparticle) has

been greater than the values obtained by electrochemical methods (PE: 70.68% for PCz, 65.97% for PCz/nanoclay, 66.28% for PCz/Zn-nanoparticle) [68]. Recently, carbohydrates and derivatives, containing the polar groups such as OH, -NH₂, and -COCH₃, which facilitate their solubility in electrolyte, have been greatly investigated to use as environmentally friendly corrosion inhibitors for metal and alloys [71]. Vazquez et al. have investigated eight carbohydrates (three commercially obtained and five synthesized) for corrosion inhibition of API 5L X70 steel in acidic medium: the corrosion process is mixed type according to the thermodynamic analysis results and the best inhibition potential has been determined as 87% (for synthesized Methyl-4,6-*O*-Benzylidene- α -D-glucopyranose) at 50 ppm [72]. Also, chitosan as a linear aminopolysaccharide is a copolymer of D-glucosamine and *N*-acetyl-D-glucosamine and is obtained by deacetylation of chitin. Due to the abundance of -OH and -NH₂ polar groups in the chitosan structure, they are easy to be adsorbed by the metal surface and are reported to be a good corrosion inhibitor [72-75]. In addition, among green corrosion inhibition research, plant extracts “multi-phytoconstituents from *dioscorea septemloba*” [76] on carbon steel in acidic solution and seven natural polymers for AZ31 Mg-alloy [77] in the saline media have been also explored. Polymeric corrosion inhibitors with recently reported environmentally friendly inhibition properties are given in [Figure 4.7](#).

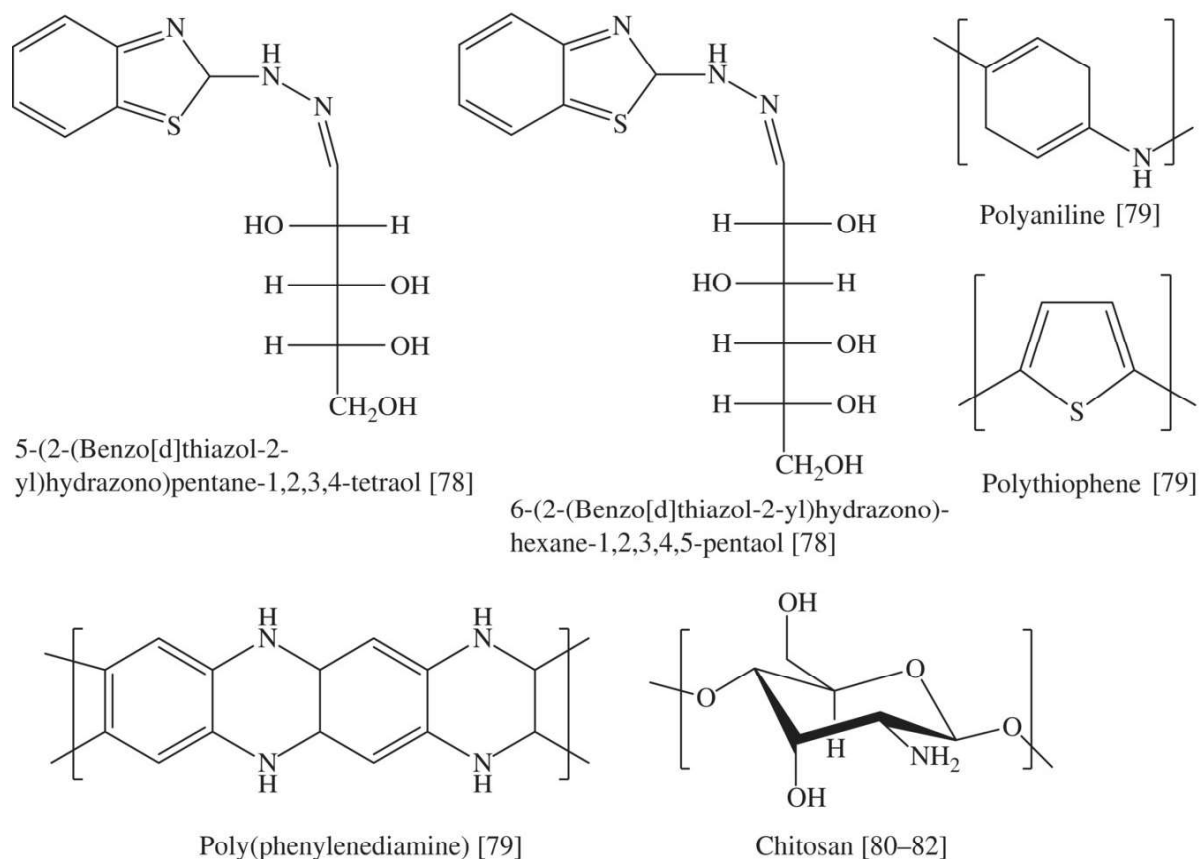


Figure 4.7 The chemical structures of the green corrosion inhibitors.

From Refs. [78–82].

4.2.1.7 Inorganic Corrosion Inhibitors

In addition to organic corrosion inhibitors, inorganic molecules and inorganic salts are also considered as inhibitor in the prevention of corrosion of metal surfaces. Many inorganic complexes, ions, and salts were successfully used against the corrosion of metal surfaces in different corrosive environments [83–85]. Inorganic corrosion inhibitors prevent the corrosion via reaction of anodic and cathodic parts of the system. On the other hand, organic corrosion inhibitors prevent the corrosion process adsorbing on metal surfaces. The most widely used inorganic corrosion inhibitors are the salts of zinc, copper,

nickel, arsenic, and additional metals. It should be noted that arsenic compounds are widely considered compared to others. The mentioned arsenic compounds scrape at the cathode cell of metal surfaces when they are mixed in the corrosive medium. It is important to note that the plating decreases the percentage of hydrogen ion interchange. The reason of this situation is the formation of iron sulfide. The reaction between iron sulfide and acid is known as a dynamical process. In the literature, some advantages and disadvantages regarding the using of inorganic corrosion inhibitors are reported. The advantages of them are that they can be used for a long time at high temperatures. Additionally, compared to organic corrosion inhibitors, they are cheaper. As disadvantage, it can be noted that they lose speedily their abilities to connect in the acid solutions that are stronger than 17% hydrochloric acid [86]. Inorganic corrosion inhibitors are classified as anodic and cathodic inhibitors.

4.2.1.8 Anodic Inhibitors

Anodic inhibitors are also known as passivation inhibitors. They cause a reducing anodic reaction. Namely, they support the metal surfaces blocking the anode reaction. In addition, they form a film adsorbed on metal surface. Usually, these inhibitors form the mentioned cohesive and insoluble film reacting with corrosion product initially formed. The corrosion inhibitors and the corrosion potentials of the metals studied affect the anodic reaction [87]. As a result of the reaction with the metal ions (M^{n+}) on anode of corrosion inhibitors, insoluble and impermeable metallic ions hydroxide films occur. If concentrations of inhibitor molecules reach to sufficient height, the cathodic current density becomes higher than the critical anodic current density. Consequently, the metal is passivated. In anodic inhibitors, it is quite important that

concentrations of inhibitor molecules should be high in the solution considered. If concentration of inhibitor is low, the film formed cannot cover the entire metal surface. This situation causes a localized corrosion [2]. Nitrates, molybdates, sodium chromates, phosphates, hydroxides, and silicates are the examples of anodic corrosion inhibitors.

4.2.1.9 Cathodic Inhibitors

In the course of corrosion process, the cathodic corrosion inhibitors prevent the occurrence of the cathodic reaction of the metal surfaces. These mentioned inhibitors having some metal ions form insoluble compounds that precipitate in cathodic sites. Here, a compact and adherent film restricting the diffusion of reducible species in these areas settles down on metal surface. The oxygen diffusion and electrons conductive in these areas provide that these inhibitors have a high cathodic inhibition. Magnesium, zinc, and nickel ions can be given as example for cathodic inhibitors because they form the insoluble hydroxides as $(\text{Mg}(\text{OH})_2, \text{Zn}(\text{OH})_2, \text{Ni}(\text{OH})_2)$ reacting with the hydroxide ions of water. The formed insoluble hydroxides are deposited on the cathodic sites of the metal surfaces to protect them. As other examples of cathodic inhibitors, the oxides and salts of antimony, arsenic, and bismuth, which are deposited on the cathode region in acid solutions, can be presented. It is well-known that these inhibitors minimize the release of hydrogen ions [88]. In the current literature, many studies regarding the performances against the corrosion of metal surfaces of inorganic corrosion inhibitors are available [84,89-91].

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