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Heterocyclic Compounds Containing N atoms as Corrosion Inhibitors: A review

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Abstract

Corrosion is the most important and costly cause of severe operational problems encountered in oil and gas production systems. It may occur anywhere in the production system, from the oil/gas well bottom to the final transfer of produced gas or oil to the refinery. Internal corrosion of carbon steel pipelines is a common and serious problem in oil and gas production, which is designed for long-term operation. It involves an interaction between the metal wall and the flowing fluids. This problem has caused the consideration of many corrosion control programs and research in various oilfields around the world. There is global use of carbon steels as materials of construction for pipes in the oil and gas industries. Compounds triazoles and their derivatives have attracted huge attention due to their simple preparation and superior corrosion inhibition activity towards mild steel in acidic environments. Therefore, the present review aims to highlight the carried out using Compounds triazoles and their derivatives for steel protection in different corrosive environments.

Keywords: Corrosion, Heterocyclic compounds, Triazoles, Acidic environment

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1-Introduction

Corrosion is one of the most important industrial problems facing the developed world. Corrosion causes annually close of many of factories and stop installations, loss of many industrial products, leading to damage them and then pollution of the environment, increased maintenance and repair costs, loss of efficiency of performance and ending with disasters and accidents such as the collapse of bridges, dams and buildings and the risks resulting from accidents due to corrosion [1-3]. Under an acidic environment, the mild steel would corrode faster and form oxides as products. Hydrochloric acid and sulphuric acid (HCl and H2SO4) are the widely used chemicals in the petroleum industry. Alike, in oil gas industries carbon dioxide (CO2) creates corrosion, and the action was noticed in 1940 by Ikeda et al.[4] CO2 gas present in natural gases dissolves in brine solution and forms a weak carbonic acid which results in steel pipeline corrosion. To control steel corrosion, As well Copper and its alloys are used in modern industries. Copper is a relatively noble metal; however, it is susceptible to corrosion by acids and strong alkaline solutions, especially in the presence of oxygen or oxidants. In the pH range

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between 2 and 5, the dissolution of Cu is relatively rapid and the formation of a stable surface oxide layer, which can passivate metal surfaces, is impossible. Copper can only be passivated by forming an oxide surface layer in weak acid or alkaline solutions [5]. The inhibitors are one of the most practical and cost-effective methods currently employed. The inhibitors may be organic molecules containing polar groups including N, S, and/or O atoms and heterocyclic compounds with polar functional groups and conjugated double bonds.[6-8] The inhibitor molecules can adsorbed on the metal surfaces and block the active sites on the surface and thereby reduce the corrosion rate.[9-12] In addition, the N, S, and/or O atoms can form a chemical bond with the Fe and would protect the steel surface in aggressive environments. Corrosion inhibitors have been considered to be the first line of defense against corrosion in oil gas industries.[13-15].

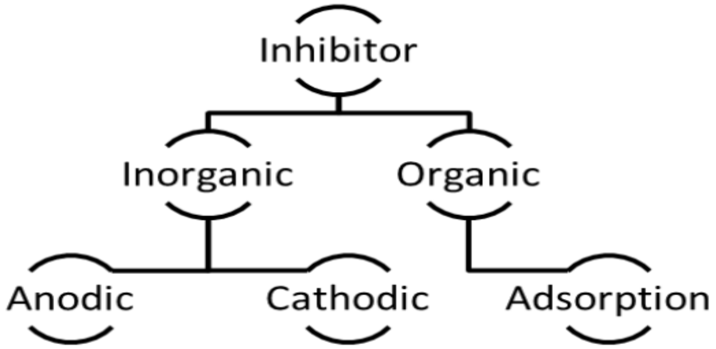


Figure 1: Classification of inhibitors [16]

2-Heterocyclic Compounds Containing S, N, and O atoms

Recently, N-containing organic compounds are proposed as a potential inhibitor for steel corrosion in a saturated CO2 environment. Particularly, imidazoline and its derivatives are getting more attention, due to its excellent adsorption character and chemical film formation properties.[17]. Roque et al.,[18] documented the use of benzimidazole derivatives as efficient corrosion inhibitors for carbon steel surface by density functional theory (DFT) and electrochemical measurements. DFT and electrochemical results clearly show that the benzimidazole derivatives adsorbed over the metal surface by giving the π-electron density from the delocalized region (N=C=N) through its highest occupied molecular orbital (HOMO) to the metal lowest unoccupied molecular orbital (LUMO).

Villamizar et al.,[19] studied the imidazolines inhibition efficiency on carbon steel corrosion in a CO2 environment. Nitrogen-based organic compounds such as benzimidazole, imidazoline amides, imidazoline amino amides, and their derivatives have been successfully used as an inhibitor in oil and gas industries.[20-24]

Sehmi et al.,[25]studied the pyrazole carboxamide inhibition of mild steel in hydrochloride acid medium the corrosion inhibition mechanism of mild steel by pyrazole molecule may be due to the presence of reactive sites in the structure of pyrazoles; these sites give rise to interactions with the metal through to the heteroatoms, the conjugated systems, and the (π) bonds, which can share their electrons with metal to establish a strong coordinate type bond. Indeed, the presence of lone electron pairs on heteroatoms (nitrogen atoms) with the protons H+ of HCl medium leads to the formation of cationic pyrazole molecules, which are easily adsorbed at the electrode interface trough to the synergistic action of anion chloride as given in Fig. 2(physisorption).

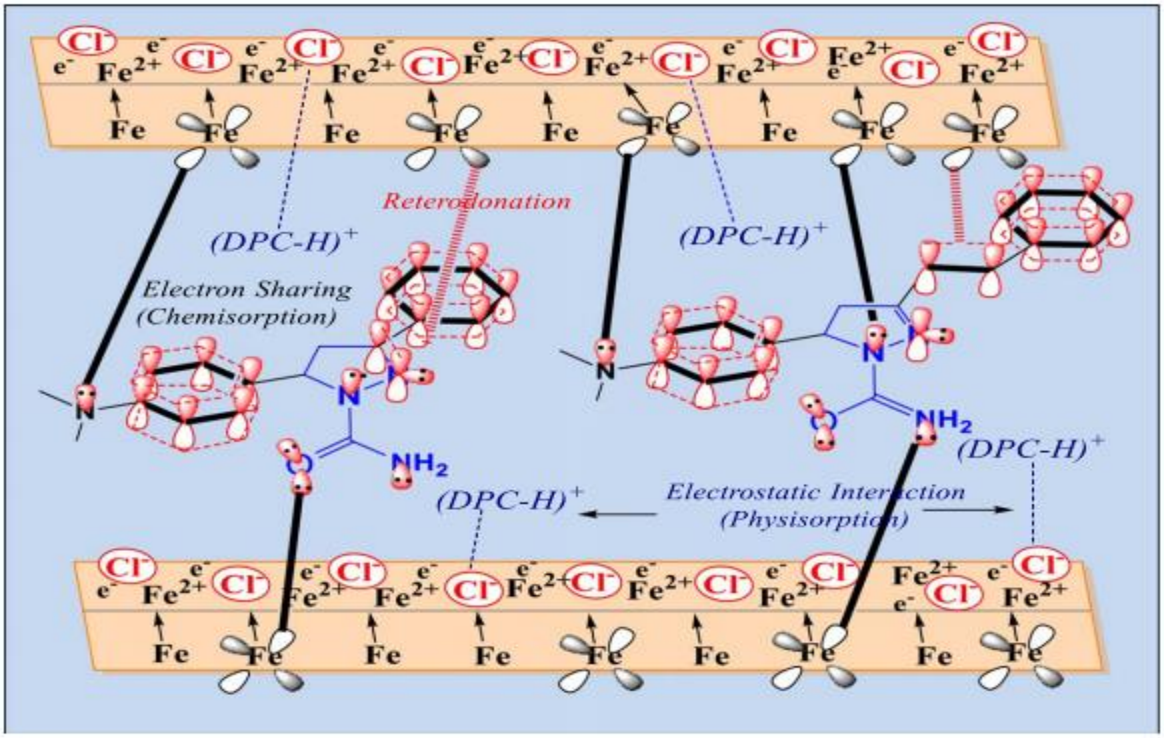
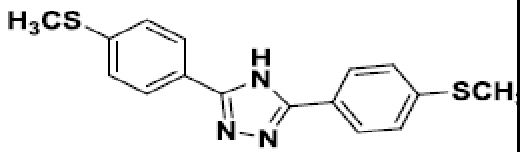
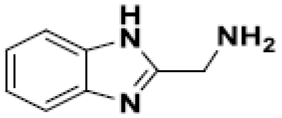
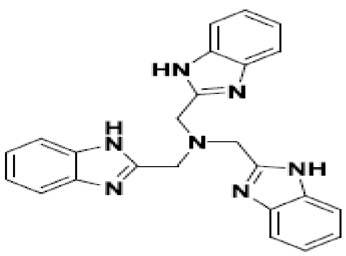
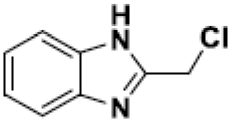
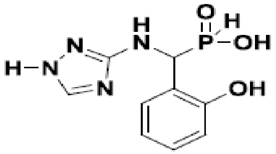
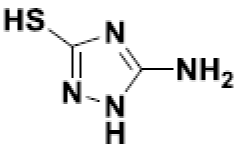
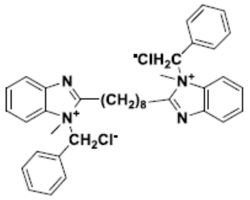
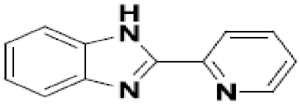
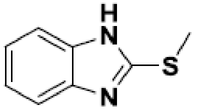
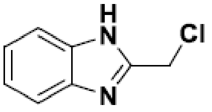


Figure 2: Suggested mechanism of corrosion inhibition of mild steel with DPC molecules.

Table 1: Heterocyclic derivatives.

S. No	Molecular Structure and name of Inhibitor	Medium	Type of INH	Conc. of INH	Maximum IE (%)	Ref.
1	 3,5-Bis(4-methyltiophenyl)-4H-1,2,4-triazole	1 M HCl	Mixed	5 × 10−4 m	96.8 %	26
2	 2-aminomethylbenzimidazole	Mild steel	1. 0 mM	0.5 M HCl	82.0	27

3	 tri(2-benzimidazolylmethyl) amine	Mild steel	2.0 mM	1.0 M HCl	91.8	28
4	 2-(chloromethyl)benzimidazole	Iron	0.2 mM	1.0 MHNO 3	87.1	29
5	 3-Salicylialidene amino-1,2,4-triazole phosphonate	Neutral aqueous solution	Mixed	5 ppm	57 %	30
6	 5-Amino-3-mercapto-1,2,4-triazole	0.1 M HCl	Mixed	2 × 10−3 m	90 %	31
7	 1,8-bis(1-chlorobenzyl-benzimidazolyl)-octane	Mild steel	130 µM	1.0 M HCl	99.1	32
8	 2-(2-pyridyl)benzimidazole	Iron	50 mM	1.0 M HCl	72.4	33
9	 2-(methylthio)benzimidazole	Iron	0.2 mM	1.0 MHNO 3	93.8	34
10	 2-(chloromethyl)benzimidazole	Iron	0.2 mM	1.0 MHNO 3	87.1	34

3- Triazole Heterocyclic Compounds

The heterocyclic compounds that contain all the three N, O, and S atoms are used as corrosion inhibitors for mild steel in acidic environmentsTriazole and triazole-type compounds containing nitrogen, sulfur, and heterocycle on the corrosion inhibition of metal in acidic media have attracted more attention because of their excellent corrosion inhibition performance [35–38]. The corrosion inhibition study includes three major methods such as weight loss, potentiodynamic polarization, and electrochemical impedance spectroscopy. Some new triazole derivatives have been still continuously synthesized and investigated as inhibitors for corrosion of metals in acidic solutions [39–41]. For example, Zhang et al. studied the corrosion inhibition of a newly synthesized oxadiazol-triazole derivative for mild steel in sulphuric solution, their results indicated that the compound was an effective corrosion inhibitor for mild steel in acid solution and its efficiency attained more than 97.6% at 298 K [42]. Sardar et al.,[43] prepared various heterocyclic compounds for inhibition of mild steel corrosion in acidic media. Fig. 3. Molecular structures of heterocyclic compounds reported as efficient inhibitors for the corrosion of mild steel in 1 M HCl and 1 M H2SO4. The reported compounds (aromatic oxadiazoles, aromatic triazoles, mercapto triazole, aromatic aldehydes, substituted dithiazolidines, and substituted thiadiazolines) are highly efficient towards the protection of mild steel which forms aggressive environments. Among various compounds tested, aromatic triazole derivatives are proved to show better inhibition efficiency than aromatic oxadiazole derivatives. It is due to the presence of an additional benzene ring and 3 nitrogen atoms in triazoles when compare to the superior performance of triazoles as compared to two heteroatoms (O and N) contained oxadiazoles. The inhibitions efficiency of aromatic oxadiazoles followed the order CMO > PMO > HMO and the inhibition efficiency of aromatic triazoles followed the order CPMT > DPMT > APMT > HPMT. The high performance of CPMT is due to the presence of an additional n-bond between carbon atoms (- C = C -) in conjugation with an aromatic ring. The thermodynamic parameters of all the compounds (Fig. 4), the free energy of adsorption (Gads), value showed low and negative value at different temperatures (30°C-50°C), indicate the spontaneous adsorption of inhibitors on the surface of mild steel in 1M HCl and 1M H2SO4. The adsorption behavior of all the organic molecules on the mild steel surface obeys Langmuir's adsorption isotherm. The potentiodynamic polarization studies confirm that all compounds in both 1 M HCl and 1 M H2SO4 are mixed-type inhibitors except oxadiazoles showed predominantly cathodic behavior.

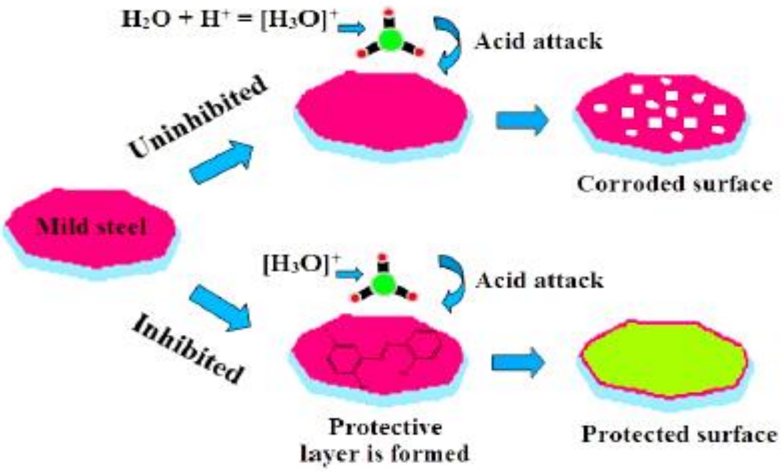
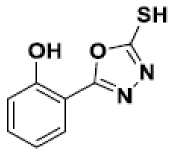
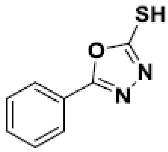


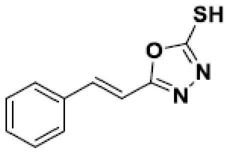
Figure3: mechanism of corrosion inhibition of mild steel in acidic medium. [44]



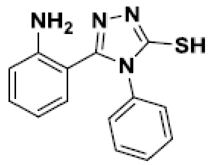
2-hydroxyphenyl-5-mercapto-l-oxa-3, 4-diazole (HMO)



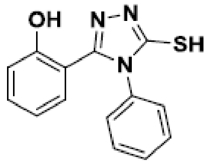
2-phenyl-5-mercapto-1-oxa-3, 4-diazole (PMO)



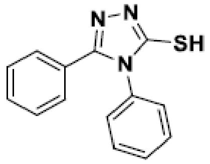
2-cinnamyl-5-mercapto-1 oxa-3,4-diazole (CMO)



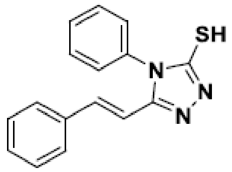
3-aminophenyl-4-phenyl-5-mercapto-1, 2, 4-triazole (APMT)



3 -hydroxyphenyl-4-phenyl-5-mercapto-l, 2, 4-triazole (HPMT)



3, 4-diphenyl-5-mercapto-l, 2, 4-triazole (DPMI)



3 -cinnamyl-4-phenyl- 5-mercapto-1,2,4-triazole (CPMT)

The researches by Fouda and Ellithy showed that some 4-phenylthiazole derivatives could inhibit the corrosion of 304L stainless steel in hydrochloric acid solution, but the inhibition effect was not very excellent [45]. However, the Synergistic effect occurred on the addition of KSCN to acid containing 4-phenylthiazole derivatives which inhibited 304L stainless steel corrosion, and the phenomenon of synergism took place at very low concentrations for the systems studied. Wang et al. also investigated the effect of some mercapto-triazole derivatives synthesized containing different heteroatoms and substituents in the organic structures on the corrosion and hydrogen permeation of mild steel in hydrochloric acid solution and their results revealed that all the mercapto-triazole derivatives performed excellently as corrosion inhibitors [46]. Especially, some N- and S-containing triazole derivatives are environmentally-friendly corrosion inhibitors compared with some commercial acid corrosion inhibitors which are highly toxic, such as chromate and nitrite [47]. El Mehdi and co-workers[48] studied corrosion inhibition ability of newly prepared triazole-derivatives such as 5-di(m-tolyl)-4-amino-1,2,4-triazole (m-DTAT) and 3,5-di(m-tolyl)-4H-1,2,4-triazole (m-THT)in 1 M HCl solution. Although both the m-DTHT and m-DTAT demonstrate the excellent inhibition efficiency towards corrosion of mild steel in 1 M HCl solution, the m-DTAT reached better inhibition efficiency of 95% whereas the m-DTHT showed 91 %. Surprisingly the electrochemical measurements showed that the m-DTHT is a mixed type in nature whereas m-DTAT is cathodic. On contrary, the adsorption of m-DTHT and m-DTAT inhibitor molecules on the mild steel surface is found to obey the Langmuir adsorption isotherm. The slight change in the IE can be explained by the availability of the pair of free electrons of the nitrogen atom, to form a donor–acceptor-type link, necessary for the formation of a protective chemisorbed film.

Numerous studies revealed that many 1,2,4-triazole derivatives can act as effective corrosion inhibitors for copper.1,2,4-Triazole derivatives, namely 3-vanilidene amino 1,2,4-triazole phosphonate, 3-anisalidene amino 1,2,4-triazole phosphonate [49], and 3-amino-5-mercapto-1,2,4-triazole [50,51], have been studied for their corrosion inhibition of copper in neutral aqueous environment by various techniques. A combination of electrochemical methods and surface examination techniques has been used to investigate the protective film and explain the mechanistic aspects of corrosion inhibition. Many of these studies revealed that inhibition of copper corrosion was due to the adsorption of triazole derivative on the surface of copper and adsorbed triazole derivative formed a complex with Cu^+ which prevented the formation of copper chloride complexes.

4-Mechanisms of actions of inhibitors

Inhibitors are substances or mixtures that in low concentration and aggressive environments inhibit, prevent or minimize corrosion. [52]Generally, the mechanism of the inhibitor is one or more of cited below:(a) electrostatic interaction between the charged surface of the metal and charge inhibitor molecule, (b) interaction of unshared electron pairs in the inhibitor molecule with the metal, (c) interaction of p-electron with metal and (d) a combination of (a) and (c) types.[53-56] Adsorption of inhibitor molecules on metal surfaces also depends on the physico-chemical properties such as functional groups, steric factor, aromaticity, electron density at the donor atoms, and π orbital character of donating electrons and electronic structure of the molecules. Adsorption can be described by two main types of interactions such as physisorption and chemisorptions. Physisorption involves electrostatic interactions between ionic charges or dipoles on the adsorbed species and the electric charge at the metal or solution interface. The heat of adsorption is low and therefore this type of adsorption is stable at low temperature. Chemisorption involves charge sharing or charge transfer from the inhibitor molecules to the metal surface and it forms a coordinate type bond. It is much stronger adsorption energy than physical adsorption and also more stable at higher temperatures.[57].

According to Musa et al.,[58], there are two possible routes for the adsorption process of heterocyclic compounds on mild steel surface; (1) forces of interactions, of the donor-acceptor, between the empty d-orbitals that are present on the surface of mild steel and pi-electrons of the heterocyclic compound, and (2) inhibitor adsorption process through the forces of interactions with the Cl^- ions that are already adsorbed. In general, inhibition activity of heteroatom follows the reverse order of electronegativity; the inhibition performance followed in the order of $O < N < S < P$. In addition, the addition of heterocyclic compounds in the acidic solutions commonly alters the electrochemical behavior or decreases the aggressiveness of the acidic environments. Interestingly, the heterocyclic compounds that have a sulfur (S), phosphorus (P), nitrogen (N), or oxygen (O) heteroatom effectively take part in the adsorption process to protect the metals.

Saha et al.,[44] Proposed an inhibition mechanism for mild steel corrosion under the acidic environment, the mild steel can be highly corroded to metal dissolution in an aggressive acid medium. Therefore, the corroded metal surface has a large number of cracks and pits. However, the inhibited mild steel surface shows very smooth morphology due to the formation of a protective layer over the mild steel surface. The adsorption of heteroatoms such as N, S, O, and Cl of inhibitor molecules on the metal surface is also the main reason.

5-Conclusions

Heterocyclic compounds are effectively used as corrosion inhibitors for steel under various aggressive environments. heterocyclic are proven to be the efficient inhibitors for corrosion of metals. The heterocyclic compounds inhibit the metal surface by forming a protective film on the surface. The inhibition of heterocyclic compounds may be anodic or cathodic or mixed type. Most of the inhibitors showed an increasing trend in the inhibition efficiency with a concentration of inhibitor. Overall, this review has summarized the Compound triazoles that are used for the corrosion inhibition of steels in various aggressive environments.

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