OPEN ACCESS
Saudi Journal of Engineering and Technology
Abbreviated Key Title: Saudi J Eng Technol
ISSN 2415-6272 (Print) | ISSN 2415-6264 (Online)
Scholars Middle East Publishers, Dubai, United Arab Emirates
Journal homepage: https://saudijournals.com

Review Article

Assessment of Inhibitive Drugs for Corrosion Inhibition Applications in Petrochemical Plants – A Review

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DOI: 10.36348/sjet.2022.v07i05.001 | **Received:** 05.11.2021 | **Accepted:** 08.12.2021 | **Published:** 12.06.2022

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Abstract

The world is paying more attention and trying to eliminate the use of substance that are toxic to human and the environment. The use of some organic and inorganic corrosion inhibitors exhibit toxicity and research has been ongoing on the use of eco-friendly substitutes. Plant extract, dyes and drugs have shown good corrosion inhibitive capabilities and this work gives a summary of several works that have been done on the use of drugs as efficient corrosion inhibitors for mild steel which is mostly used in the petroleum and petrochemical industry in acidic media such as HCl and H_2SO_4.

Keywords: Corrosion, Inhibitive Drugs, Kinetic and Thermodynamic Assessment, Petrochemical Plants.

1. INTRODUCTION

The major feedstock for a petrochemical plant is obtained from crude oil or natural gas. These petrochemical plants have a primary function of converting low end value products gotten from the refining process into more useful high value materials [1-3]. Carbon steel (mild steel) is the most popularly used material for construction in a petrochemical as a result of its mechanical and structural strength as well as its low cost, although it is very much subject to corrosion [4-6]. Corrosion is the deterioration of a metal due to chemical or electrochemical reactions with its environment [7-8]. Several factors contribute to corrosion which include the reactivity of metal, presence of impurities, presence of air, moisture, gases like sulphur dioxide and carbon dioxide, and presence of electrolytes [5]. Over the years, corrosion engineers have come up with different ways of controlling and monitoring corrosion which involves the use of coatings, cathodic protection and also the use of corrosion inhibitors, which has become a cost effective and acceptable means of corrosion control especially in acidic environment found in the petrochemical industry [1, 4].

The goal of the review is to summarize works on corrosion inhibition of mild steel in different corrosive environment with the use of drugs as an eco-friendly and efficient inhibitor

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2. CORROSION

Fontana and Greene [8] defined corrosion as an unwanted deterioration of a material as a result of interaction with its environment and this reaction significantly affects the quality of the material. Corrosion in a petrochemical plant can be found in different forms and has been classified into uniform corrosion, galvanic corrosion, crevice corrosion, pitting, selective leaching, erosion corrosion, stress corrosion cracking, intergranular corrosion, corrosion fatigue and fretting corrosion [3, 9]. However, in the petroleum industry, the most common form of corrosion occurrence is uniform, erosion and pitting corrosion [2, 3, 6, 7].

Corrosion is generally known as an electrochemical reaction and its control has been hinged on the disruption of the reaction [10]. Walsh *et al.* [11] broadly classified the approaches to corrosion control into four major categories which are material and design, modification of the electrolyte, change in the electrode potential and surface coating.

2.1. Material and Design

This method of corrosion control is fundamental and has to do with the appropriate selection of materials considering the environment it would be subjected to within its operating parameters. There is no ideal material that can serve all conditions based on resistance to corrosion, so selection of materials is based on information and experience which are included in the standards, procedures, reports, and articles [1]. Hence the environment in which they are to be used, cost and availability are critical factors in the selection of the appropriate material.

2.2. Surface Coating

The purpose of coating is usually to isolate the surface of the material from its operating atmosphere. Organic coatings are paints and other media based on alkyl and epoxy resins. Inorganic coatings are like ceramics, enamels and glasses while metallic coating is the application of an inexpensive resistant metal to the material of interest and this is achieved by electrodeposition or electroplating [11-13].

2.3. Change in Electrode Potential

This involves raising or lowering the electrode potential to a point away from their corrosion potential, where they become stable. The methods to change the electrode potential are called anodic and cathodic protection and they work on only the external surface of the material. For anodic protection, the electrode potential of the metal (anode) is shifted to the region of passivity by applying a low current which is equal to the corrosion rate of the protection system forming a protective film on the metal surface [9, 11-15]. In cathodic protection, the metal to be protected is connected to a less conductive metal, such as magnesium, aluminum, zinc or their alloys, as sacrificial anode or connecting the metal to the negative terminal of a power supply, which is known as impressed current [1]. Cathodic protection is widely used in industries for the protection of metallic structures such as vessels, tanks, buried pipes.

2.4. Modification of Electrolyte

It involves altering the chemical composition of the corrosive environment by removing the corrosive constituents or by the addition of substances that can reduce the corrosion rate within that environment [11]. Removal of the corrosive agents can be achieved by means such as neutralizing acidity by the addition of lime or reduction of humidity by means of desiccants such as silica gel. Substance added to reduce the rate of corrosion of the metal is known as corrosion inhibitors.

3.0. CORROSION INHIBITORS

Corrosion inhibitors are substances which are added in small amounts to the metal environment in order to prevent corrosion from progressing or reducing the corrosion rate of the metal [16-19]. The selection of inhibitors that are added to a process mainly depends on the solubility and the dispersibility of the chemical within the environment/electrolyte [17]. The selection and the amount of inhibitor used depend on the acid type and its strength, the steel type, the desired protection time, and the expected temperature [20-23]. The use of corrosion inhibitors is regarded as one of the most cost-effective and practical methods for corrosion protection especially in industrial application [21]. This facilitates the use of lower-grade carbon steel, which significantly reduces the capital costs of construction project when compared with the use of higher-grade materials for the same project [25].

Corrosion inhibitors are classified based on the chemical structure or origin as organic and inorganic inhibitors. The organic inhibitors can be synthetic or natural. Organic inhibitors are the most widely used inhibitors in the petroleum and petrochemical industry and they react mainly by adsorption onto the surface of the metal [30]. They are mainly heterocyclic atoms containing sulphur, nitrogen, oxygen and phosphorous but the synthetic organic inhibitors are however expensive and toxic to the environment and human thereby posing problem in handling and waste management [15, 26-29]. Inorganic inhibitors are substances that contains salts of zinc, copper, nickel, arsenic, and other metals which operates by suppressing the electrochemical reaction at the metal's anode-cathode region. They are less expensive than the organic inhibitors and they work efficiently in higher temperatures but are also toxic to the environment [26]. The natural organic inhibitors are gotten from natural sources and are not toxic in application and to the environment. This property has gained scientists interest in the use of these compound as efficient and eco-friendly substitutes to inhibitors which are toxic. Recent studies have been based on the use of dyes, plant extract and inhibitive drugs.

The use of dyes as corrosion has shown promising characteristics and can be scientifically attributed to the presence of heteroatoms and π-electrons in their compound [[31-36]. These inhibitors are non-toxic and have shown remarkable corrosion inhibition efficiencies as reported by several authors [27, 39-52].

Recent studies have also shown that some plant product or extract are good corrosion inhibitors. They offer better advantages over the synthetic organic and inorganic inhibitors as they are inexpensive, readily available, renewable and most of all, environmentally friendly. Several researches have been done with several plants and their products (leaves, stems, barks, roots, fruits) found to be good corrosion inhibitors in acidic or alkaline media but the challenge with the use of this type of inhibitor is the likelihood of contamination by microorganisms [40-41]. Several scientists around the world have reported many plant extracts which are good corrosion inhibitors [36, 53-74].

4.0. DRUGS USED AS CORROSION INHIBITORS

Drugs have similar chemical structures as many organic compounds used as corrosion inhibitors. This facilitated research into the use of several drug as corrosion inhibitors and several have shown positive characteristics as seen from literatures. Many of the drugs are hydrophilic (water loving) and biodegradable, hence, it is environmentally friendly [75]. The corrosion inhibition ability of the drugs can be attributed to the presence of polar atoms such as S, N, O, P in their compound. These atoms possess lone electrons which are readily transferred to the surface of the metal which is being adsorbed to the surface, forming a protective barrier [37]. Several works have been carried out on the use and effectiveness of certain drugs for corrosion inhibition application.

Kumar and Karthikeyan [34] carried out a study on the use of cloxacillin as a corrosion inhibitor for mild steel in hydrochloric acid solution. They found that cloxacillin had an efficiency of about 81% at a concentration of $15x10^{-4}$M of the drug and it acted by the adsorption of the drug into the metal surface following the Temkin's adsorption isotherm [75]. Akpan and Offiong [76] also used ciproflaxin in their corrosion inhibition studies on mild steel in HCl at room temperature. The inhibition efficiency was seen to increase with an increase in the drug concentration and decreases with an increase in temperature. It was also seen that its inhibition kinetics followed a first order and the Langmuir's adsorption isother.

Ebenso *et al.* [77] carried out a study using quinoline and its derivatives like quinaldine and quinaldic acid. They found these drugs to be efficient corrosion inhibition for mild steel in hydrochloric acid. The highest efficiency value that was achieved from their study was 94.21%. Other authors also investigated quinoline derivates for its corrosion inhibition effect [97-98, 117-118]. Ahamad *et al.* [78] also used primaquine as corrosion inhibitor for mild steel in hydrochloric acid. The drug was found to be efficient with value of 98% due to adsorption of the protonated molecule on the metal surface thereby blocking the active sites for corrosion to take place. Obot and Egbedi [79] also carried out studies using an antifungal drug known as ketoconazole with the brand name Nizorol. It was found that efficiency of the drug as a corrosion inhibitor increased with concentration but also decrease with an increase in temperature thereby suggesting a physical adsorption process taking place and it obeyed the Langmuir isotherm.

Three antibacterial drugs, penicillin G, ampicillin and amoxicillin, were analysed for its inhibition efficiency on carbon steel in hydrochloric acid environment using electrochemical techniques [80, 129-134]. The drugs were observed to obey the Langmuir isotherm by their adsorption on the metal surface and polymerization studies revealed that they are mixed type inhibitors. Atenolol drug was also investigated by Karthik and Sundaravadivelu [81] employing using weight loss and electrochemical experiments. The drug showed a maximum inhibition efficiency of 93.8% at 300 ppm concentration. Adsorption of the drug on mild steel surface obeyed the Langmuir isotherm. Tafel polarization study suggested that atenolol acts as mixed type inhibitor. The inhibition performance of atenolol was also studied by Fourier-transform infrared (FT-IR) spectroscopy and scanning electron microscopy (SEM) methods. The experimental results were supported by quantum chemical calculations. The use of gramine or donaxine drug as corrosion inhibitor was also investigated by Quatorone *et al*. [82] on mild steel in 1 M hydrochloric acid solution using weight loss and electrochemical techniques within a temperature range of 20-50oC. Gramine exhibited maximum inhibition efficiency of 98% at 7.5 mM concentration and the adsorption of the drug on metallic surface obeyed the Langmuir isotherm. Polarization study revealed that gramine acted as mixed type inhibitor. Obot *et al*. [83] also investigated the use of Metronidazoleas drug as an environmentally friendly inhibitor for mild steel corrosion in 0.5M hydrochloric acid solution using gravimetric and potentiodynamic polarization techniques. The metronidazole acted as anodic type inhibitor and showed a maximum efficiency of 80.01% at 10μM concentration. Adsorption of metronidazole on mild steel surface followed the Temkin isotherm. Dubey and Potdar [84] carried out corrosion inhibition studies using ofloxacin, amifloxacin, enofloxacin, pefloxacin, ciprofloxacin and norfloxacin drugs on mild steel in sodium chloride solution. It was found that these drugs were efficient corrosion inhibitors with increasing efficiency as temperature increases reaching values of up 98.9%.

Tinidazole also showed a maximum efficiency of 90% at 400 ppm concentration for its corrosion inhibition effect on mild steel in 1M hydrochloric acid solution using weight loss and electrochemical techniques, when analysed by Reza *et al*. [85]. Shukla and Quraishi [86] studied the use of cefalexin drug for the corrosion inhibition of mild steel in 1 N HCl using electrochemical and weight loss measurements. They suggested that the drug operated by a physical adsorption process unto the metal surface and a decrease in inhibition efficiency with an increase in temperature between 35–65oC was reported. Ayoola *et al*. [87] and Eddy *et al*. [112] also investigated the use of chloramphenicol drug, at varying concentrations, as corrosion inhibitors on mild steel in 0.1M solution of HCl using weight loss technique, linear polarization method and open circuit potential method. Their result showed that the corrosion rate decreased with an increase in the concentration of the inhibitor drug molecule and it obeyed the Langmuir adsorption model.

The use of Cephapirin drug as corrosion inhibitor substance on carbon steel in 1M HCl solution was also investigated by Mahmoud *et al*. [88]. It was found from this study that the inhibition efficiency increased with increasing concentration of the drug and a protective film was formed on the metal which was observed by UV-visible reflectance Spectroscopy.

Perlman *et al*. [89] also studied the use of Streptomycin on mild steel in 1M of HCl using weight loss measurement, Tafel polarization, and electrochemical impedance spectroscopy (EIS). The drug showed a maximum inhibition efficiency of 88.5% at 500 ppm concentration and Polarization analysis showed it was a mixed type inhibitor. The corrosion inhibition efficiency of Pheniramine drug on mild steel corrosion in 1 M HCl solution was examined using gravimetric analysis, linear polarization resistance, and electrochemical impedance spectroscopy and potentiodynamic polarization measurements by Ahamad and Quaraishi [90]. It was suggested that Pheniramine drug is a mixed type inhibitor and it followed the Langmuir adsorption isotherm. Al-Shafey *et al*. [91] carried out a study on the effect of expired drugs, Phenytoin sodium drug. It showed good efficiency up to 79% and obeyed the Langmuir adsorption isotherm. The use of cephalothin on carbon steel in 1M hydrochloric acid solution for its corrosion effect employing electrochemical impedance spectroscopy and scanning electron microscope (SEM) analysis, was studied by Aldana-Gonzalez [92]. The drug exhibited an efficiency of 92% at 600 ppm concentration and also obeyed the Langmuir adsorption isotherm. El-Naggar [93] and Arslan *et al*. [139] also studied the use of some sulfa drug compound for corrosion inhibition capabilities on mild steel in acidic medium of HCl and H2SO4. They suggested that the compounds showed good efficiency and its inhibition mechanism was mainly through adsorption. It was also found that the compounds were more efficient in their inhibition activities in HCl than in H2SO4 solutions.

Drug name	**Corrosive Medium**	**Inhibitive effect**	**Ref.**
Telmisartan	1M HCl	Was found to be a mixed type inhibitor and followed Temkin adsorption isotherm. Max. efficiency of 97.39% at 125 mgL^{-1} conc. of inhibitor	94
Amodiaquine	1M HCl	Max. efficiency of 44.33% at 0.006M and obeyed Langmuir adsorption isotherm.	95
Sparfloxacin	2.5M HCl	Investigated using gravimetric, gasometric and thermometric analysis. An efficiency of 97.47% at 12×10^{-4} M was observed and obeyed Langmuir isotherm.	96
Ceftriaxone	HCl	The factor determining ceftriaxone inhibition efficiency is the p-electron of aromatic ring and a lone pair of electrons of nitrogen atom	97
Cefotaxime	HCl	Cefotaxime was reported to be very effective as corrosion inhibitor (95.8%)	98
Ofloxacin	HCl	Chemical adsorption was proposed due to the value of ΔGads being negative and above 40. A maximum inhibition efficiency of 94.74% was recorded for this inhibitor at a concentration of 3x10-3 M at 303 K.	99-101
Sulfadiazine	HCl	Sulfadiazine was found to show the best inhibition efficiency due to due to the presence of electron withdrawing groups (oxadiazole moiety) in its molecule.	93, 102
Streptomycin	HCl	the underlying mechanism that determines inhibitor performance in acid solution is the adsorption of streptomycin molecule onto mild steel surface in its protonated form.	103
Dapsone	HCl	Reported efficiency of 95% at an inhibition concentration of 0.219 mM and observed to be a mixed type.	104
Sparfloxacin	H_2SO_4	A maximum inhibition efficiency of 97.47% at a concentration of $12x10^{-4}$M sparfloxacin was reported based on the results of weight loss measurements.	105
Meclizine	1M HCl	They suggested physisorption of this drug owing to decrease in inhibition efficiency at elevated temperatures and reported ΔGads value of -38 kJ/mol at 303 K	106
Famotidine	0.1 N HCl and H_2SO_4	famotidine acts as an inhibitor, bringing down the corrosion considerably even at low concentration.	107
Tramadol	0.5 M HCl and 0.25 M H_2SO_4.	They reported that tramadol reduced the rate of corrosion of mild steel in both acids but was more efficient at a concentration of 21.6x10-4 M in HCl (82.6% IE) than in H2SO4 (76% IE).	108
Methocarbamol	H_2SO_4	They asserted that methocarbamol significantly inhibits the corrosion of mild steel at 303 K, with efficiency reaching up to 67.12% at a concentration of 2.0 x 10^{-3} M. Due to the fact that increasing temperature up to 333 K eventuates in a decrease in inhibition efficiency the authors concluded that the underlying mechanism that causes inhibition performance of methocarbamol is likely to be attributable to physical adsorption.	109, 139
Irbesartan	1M HCl and 0.5M H_2SO_4	Corrosion inhibition of 94% and 83% was achieved at 300 mg/L in HCl and H_2SO_4 respectively.	110
Melatonin	0.5M H_2SO_4	Mixed type inhibitor with efficiency of 94.76% at a concentration of 500 ppm and surface coverage is 0.948.	111
Fluconazole	2.5M HCl	Anodic type inhibitor with efficiency of 96% at 0.30mM and obeyed Langmuir adsorption isotherm.	112
Metformin	1M HCl	Mixed type and efficiency 96% at 400ppm and follows Langmuir adsorption isotherm.	113

Other authors also explored the use of imidazole and its derivatives [90, 120-125]. Other drugs such as Pyrazolone [114-115], Oxadiazole [116, 126-128], Cefazolin [119], Rhodanine [100, 140-142], Ceftazidime [135-137], Famotidine [143-144], risperidone [145-146], pyridine and its derivatives or salts [147-150], were also analysed and found to be efficient corrosion inhibitors, though at varying capacities.

5.0 CONCLUSION

This review summarizes the work done by different author on the use of corrosion inhibitive drugs for mild steel in different acidic media. The use of drugs has gained so much attention by researchers as a result of its cheap cost and biodegradability. Several drugs compounds were reviewed and they all exhibited corrosion inhibition capabilities and most of them showed high efficiency at low temperature and at increased concentration. However, the efficiency reduced with increase in temperature reveals that most of the drugs were absorbed onto the metal surface by physical adsorption.

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