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REVIEW

Corrosion protection with eco-friendly inhibitors

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Online at stacks.iop.org/ANSN/2/043001**Abstract**

Corrosion occurs as a result of the interaction of a metal with its environment. The extent of corrosion depends on the type of metal, the existing conditions in the environment and the type of aggressive ions present in the medium. For example, CO_3^{-2} and NO^{-3} produce an insoluble deposit on the surface of iron, resulting in the isolation of metal and consequent decrease of corrosion. On the other hand, halide ions are adsorbed selectively on the metal surface and prevent formation of the oxide phase on the metal surface, resulting in continuous corrosion. Iron, aluminum and their alloys are widely used, both domestically and industrially. Linear alkylbenzene and linear alkylbenzene sulfonate are commonly used as detergents. They have also been found together in waste water. It is claimed that these chemicals act as inhibitors for stainless steel and aluminum. Release of toxic gases as a result of corrosion in pipelines may lead in certain cases to air pollution and possible health hazards. Therefore, there are two ways to look at the relationship between corrosion and pollution: (i) corrosion of metals and alloys due to environmental pollution and (ii) environmental pollution as a result of corrosion protection. This paper encompasses the two scenarios and possible remedies for various cases, using 'green' inhibitors obtained either from plant extracts or from pharmaceutical compounds. In the present study, the effect of piperacillin sodium as a corrosion inhibitor for mild steel was investigated using a weight-loss method as well as a three-electrode dc electrochemical technique. It was found that the corrosion rate decreased as the concentration of the inhibitor increased up to 9×10^{-4} M; 93% efficiency was exhibited at this concentration.

Keywords: corrosion, environment, aggressive ions, eco-friendly inhibitors, piperacillin sodium

Classification numbers: 1.00, 4.02

1. Introduction

Corrosion is the destructive phenomenon which affects almost all metals. Iron is the most widely used metal and one of the first for which corrosion was encountered [1]. Numerous scientific and engineering discoveries have helped in the understanding of various corrosion mechanisms. The understanding of iron and steel corrosion was reasonably developed by the start of the 20th century: one of the first modern textbooks on corrosion prevention and control was published by McGraw-Hill in 1910 [2].

Three main reasons for the importance of corrosion are economics, safety and conservation. The economic impact can be reduced by reducing material losses, as well as subsequent economic losses, e.g. the corrosion of piping, tanks, metal components of machines, ships, bridges, marine structures, etc. Corrosion of turbine blades, bridges, airplane components, automotive steering mechanisms, pressure vessels, boilers and metallic containers for toxic chemicals are a few examples of catastrophic consequences, hence safety is a critical consideration in equipment design. The loss of metal by corrosion and the cost of rebuilding requires further



investment of all these resources—metal, energy, water and human. The replacement of several million domestic hot-water tanks and millions of corroded automobile mufflers each year because of failure by corrosion emphasizes the impact of direct losses. The losses include the extra cost of using corrosion-resistant metals and alloys instead of carbon steel [3].

Getting rid of unnecessary waste and its associated pollution is a major concern. Corrosion is a form of waste that, like all other waste, pollutes the environment. The environmental consequences of corrosion are both severe and complex. They often extend far beyond the immediate issue of resource depletion. Greater awareness of harmful consequences of corrosion will help us to preserve the environment.

Pakistan may not yet be as developed as the US or Europe, but corrosion is a multibillion-dollar problem for the country. A recent survey by Pakistani scientists regarding the cost of corrosion for Pakistani companies, municipalities and households gives an estimate of 250–300 billion Pakistani rupee (approximation 3 billion US dollars) per year [4]. The estimated price tag does not include the cost of environmental remediation after the occurrence of chemical spills. Nor does it consider the decrease in production that occurs as a result of corrosion. It may be challenging to implement a broad corrosion-control initiative in Pakistan, which has been damaged in recent years by unrest. The country's efforts at development have also been further undermined by last year's flooding.

2. Corrosion issues in the oil and gas industry

Several billion dollars has been lost in many catastrophes. Pipeline ruptures and oil spillages have been experienced, causing environmental pollution. In addition, resources are lost in cleaning up the environmental mess. Large-scale ecological damage also results from corrosion. Petroleum, chemical and mechanical engineers and chemists are greatly concerned about the possible occurrence of corrosion in industrial plants. It is understood that corrosion can also affect the chemistry of processes, and the product of corrosion can affect the purity of the reaction products [5].

The study of corrosion is multi-disciplinary in nature. Its calculation involves knowledge of the viscosity, specific heat capacity, thermal conductivity and density of the fluid concerned. In some cases a thorough study of the property of the material from which the plant is fabricated is required. In the oil refinery, distillations of crude oil into fractions are performed in perforated trays. Corrosion may attack the pipelines of crude and refined oil. Most petroleum products are stored in varieties of containers and oil reservoirs whose materials may be corrosive because of variations in the temperature, heat exchange and pressure of the petroleum products which are affected by environmental factors. Some crude oils and fractions with a high content of sulfur compounds called mazcaptan or thiols [5] are corrosive in nature.

Corrosion rates can vary from zero to severely high amounts in oil-field production environments. Corrosion control strategies work when crude oil is efficiently separated

from other species. CO₂ and H₂S gases in combination with water are the main culprits [6]. H₂S can be highly corrosive but can, in some cases, form a protective sulfide scale that prevents corrosion. Corrosion can also be caused by micro-organisms that attach to pipe walls. Internal erosion of pipelines may be due to solids, such as sand, which can also cause crevice corrosion under stagnant conditions. Oxygen, if not controlled, can greatly aggravate corrosion problems. External corrosion problems in the oil and gas production industry are normally similar to those encountered in the pipelines. The use of protective coatings for offshore environments can effectively reduce the corrosion of platforms and tanks [7].

Naphthenic acid is a generic name used for all the organic acids present in crude oil. Most of these acids are believed to have the chemical formula R(CH₂)_nCOOH where R is a cyclopentane ring and *n* is typically greater than 12. Many crude oils from various sources have a high naphthenic acid content [8]. High temperature crude corrosivity of distillation units is a big problem in the refining industry. Naphthenic acid and sulfur compounds considerably increase corrosion in high temperature. Unexpected high corrosion rates are found on alloys that would normally be expected to resist sulfide corrosion. Alloys like AISI (the steel system of the American Iron and Steel Institute) 316 and 317 have been found to be sensitive to corrosion under these conditions. Various parameters causing corrosion due to naphthenic acid and sulfur compounds include the chemical, content, boiling point distribution, and decomposition temperature. Alloy composition, fluid velocity, degree of vaporization and temperature are also important factors affecting the corrosion process [9–11].

3. Sewer structure corrosion

Fan *et al* [12] presented an overview of causes of sewer deterioration and its control methods. They emphasized minimizing hydrogen sulfide production to avoid subsequent sewer line corrosion. Sulfide generation is a bacterial process occurring in the submerged parts of the sewer. Fresh domestic water is usually sulfide-free; however, sulfide can dissolve in fresh water at low velocity or stagnant conditions and low dissolved oxygen content, in elevated waste water temperature and high strength waste water, etc. The sulfide is released from the waste water as H₂S gas generating odor and initiating corrosion problems. Sulfuric acid may also be formed by another type of bacteria, which causes damage to waste water piping and facilities. Consequences may be in the form of pipe failure, disruption of service and uncontrolled release of waste water.

Sulfide (S⁻²) and bisulfide (HS⁻) ions are colorless and odorless. H₂S is also odorless in aqueous form; however, once it leaves the dissolved phase and enters the gas phase, it starts to create odor. In high concentrations this gas is hazardous to humans: 10 ppm can cause nausea, headache and eye irritation, above 100 ppm causes serious breathing problems, and above 300 ppm causes death in a few minutes [13]. Temperature and settleable solids are factors which affect sulfide concentration in sewer water. Higher waste water temperatures increase the metabolic activity of

the sulfate-reducing organisms, causing faster conversion of sulfate to sulfide and increasing sulfide concentrations. Sewer sediments and dissolved H_2S can be eliminated by in-sewer chemical addition or in-line sewer flushing.

Thiobacillus aerobic bacteria colonize at the pipe crown, walls and other surfaces in waste water pipes and structures, consuming H_2S and oxidizing it to H_2SO_4 . Favorable conditions exist in waste water collection systems for some portion of the year. The effect of H_2SO_4 on concrete surfaces exposed to the sewer environment can be devastating. The pH of the solution is decreased from 11 or 12 to 9–9.5 due to the acidic effect of CO_2 and H_2S gases, which support the bacteria [14]. Sulfuric acid attacks the concrete and gets access to rebars through the cracks developed in the concrete. Formation of a mineral called 'ettringite' which is calcium sulfaluminate hydrate, is also a by-product of sulfuric acid corrosion of concrete. Ettringite expands during formation and lifts the corroded concrete away from the sound concrete [15].

4. Corrosion of pollution control equipment

Pollution control equipment like baghouse filters and electrostatic precipitators, and other associated components such as conditioning towers, ducts, etc, frequently fail due to corrosion. These areas are exposed to frequent condensation, especially when there are cold air leaks, low external temperatures, and frequent shutdowns. Corrosion is more severe when there is a higher concentration of acidic compounds in the combustion gases. The sources are usually sulfur content in the feed of the fuel, chloride content in the feed or in air near the coast, and CO_2 , SO_x and NO_x compounds from the combustion process. The acidic condensate typically has a pH below 1.0 and contains sulfuric acid and sulfurous acid, mostly from the sulfur in the fuel. This high temperature acid condensate rapidly corrodes the metal. Corrosion in pollution control equipment in the cement industry has been highlighted by Mazeika [16]. Additional costs are required to combat these issues or the consequences may be more severe. By recognizing the short- and long-term economic impact of corrosion damage, one can optimize the capital investment when selecting a cost-effective corrosion control solution. With a good understanding of the expected operating conditions, it is possible to implement a suitable corrosion prevention scheme.

5. Detergent pollution and corrosion

Ions such as CO_3^{2-} , NO_3^- , Cl^- and NO_3^- present in the medium have different effects on metals. For example, CO_3^{2-} is adsorbed on the metal surface and produces insoluble carbonates with iron. As a result, the metal is isolated from the medium and corrosion is prevented [17]. NO_3^- and carbonates increase the dissolution of the metal and decrease the corrosion [18]. On the other hand, halide ions are adsorbed on the metal surface, preventing the formation of the oxide phase on passivating metals, allowing corrosion [19–24]. The chloride ion increases corrosion on many metals. Nitro compounds or nitrite ions decrease the free energy of the system and induce passivity [19]. Organic molecules with

groups like $-OH$, $-CHO$, $-COOH$, $-CN$, $-CO$, $-NH_2$ and $-SO_3$ also interact with the metal, providing effective protection [18].

Zor *et al* [25] studied the effect of detergent pollution on corrosion of metals. They investigated the effects of linear alkylbenzene (which is a raw material of detergent and has surface active properties) and linear alkylbenzene sulfonate (which is formed by the sulfonation of linear alkylbenzene on metals) on iron, using a three-electrode dc corrosion technique [1, 3] in 3.5% NaCl solution with varying ion concentrations. These organic molecules are found together in waste water [26–29]. It was found that the corrosion rate decreased as the concentration of linear alkylbenzene increased. This was due to the passivating behavior of the detergent which generated a barrier between the metal and environment.

6. Corrosion remedies to decrease pollution

Human health, animals and vegetal life or various materials, for example, metals and nonmetals, are at risk due to the hazardous nature of many organic and inorganic pollutants [30–32]. Hazards may occur naturally or as a result of human activities (such as industrial pollution, transportation accidents and damage, radioactive pollution, water pollution, petroleum pollution, etc). Corrosion is considered one of these causes [33]. The use of some environmentally unacceptable materials such as chromium salts has been restricted because chromium (Cr^{+6}) is highly toxic and carcinogenic, and been replaced with environmentally friendly compounds for alloy coatings [34, 35]. Strontium chromate, zinc chromate, and chrome phosphate, etc are heavy-metal-based and largely carcinogenic. Lead-based coatings cause health complications in children. Small amounts of chromic acid or potassium dichromate can cause kidney failure, liver damage and blood disorders. Chromate mists entering the lungs may eventually cause lung cancer [36]. Chemical pickling of materials such as stainless steel (SS) by using $HF-HNO_3$ mixture [37–40] results in the disadvantage of generation of nitrous gas emissions and nitrate effluents, which pollute the environment. HCl has proved to be more economical and efficient in the pickling of metals, acidization of oil wells and in cleaning scales, compared to other mineral acids [41]. The great advantage of HCl over other acids in cleaning and pickling operations lies in its ability to form metal chlorides, which are extremely soluble in aqueous phase, compared to sulfate, nitrate and phosphate.

Carbon steel and stainless steel and other corrosion resistant alloys are widely used for various engineering applications. The effect of chloride ions is not very harmful for carbon steel but the same is dangerous for passivating alloys due to destructive attack on the passivating film [1, 2]. Inhibitors play an important role in controlling corrosion; however, selection of the inhibitors should be carried out carefully considering the unwanted polluting effect of some chemicals.

The anodic inhibitor shifts potential in a more positive (anodic) direction [42]. These inhibitors function to reduce the dissolution rate of the passivating oxide [43], re-passivating the

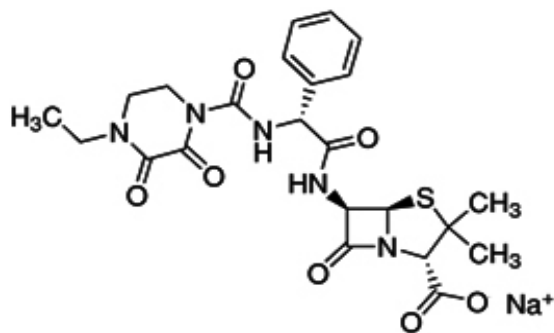


Figure 1. Molecular structure of piperacillin sodium.

surface by repair or reformation of the oxide film, repairing the oxide film by plugging pores with insoluble compounds and preventing the adsorption of aggressive anions. An example of this type of inhibitor is chromate ions, which should be replaced by more environmental inhibitors, such as rare earth elements [44, 45], also, molybdate, nitrite and phosphate [46]. In the case of cathodic inhibitors the potential shifts towards more negative values (cathodic direction). Examples of cathodic inhibitors are polyphosphates, e.g. sodium polyphosphate ($\text{Na}_2\text{P}_2\text{O}_7$), sodium tripolyphosphate ($\text{Na}_5\text{P}_3\text{O}_{10}$), etc. These inhibitors are considered safe. Mixed inhibitors work to inhibit both anode and cathode reactions. Benzotriazole is widely used for copper and copper alloy protection [47]. Quinolines and thiourea are used to inhibit the dissolution of mild steel in H_2SO_4 . Amines, amides and acridines are used to inhibit steel corrosion in HCl.

7. Green inhibitors

Recent research revolves around the use of rare earth metals as a green alternative for chromium species, as the majority have null toxicity. For example, Ce-salts used as inhibitors for tinplate [48, 49]. Hybrid organic-inorganic sol-gel matrices, with up to 20 wt% incorporated ceria nanoparticles, have been employed as coatings for an AA2024-T3 aluminum alloy. Although the performance of the coated alloy depends on the nanoparticle content, electrochemical studies revealed beneficial effects of incorporation of ceria nanoparticles [50]. Natural products are extracted and used as environmentally-friendly corrosion inhibitors. The effect of Henna extract on the corrosion of mild steel in 1 M HCl solution was investigated through electrochemical techniques and surface analysis [51]. The examined compounds act as a mixed inhibitor and inhibition efficiency increases with inhibitor concentration. Handling the toxic inhibitors must be considered in processes where compounds may be inhaled or contacted [52]. Obot and Obi-Egbedi [53] examined the effect of ginseng root as an inhibitor for aluminum alloy in HCl solution. The inhibition efficiency was found to increase with the concentration of the inhibitor but was observed to decrease with an increase of temperature. Ginseng refers to species within *Panax*, a genus of slow-growing perennial plants with fleshy roots. The phenomenon of physical adsorption is proposed from the kinetic and thermodynamic parameters evaluated. The use of environmentally friendly sodium benzoate (SB) and sodium dodecylbenzenesulfonate

Table 1. Corrosion parameters for mild steel in 1 M HCl solution without and with inhibitor from weight-loss measurement at 30 °C for 3 h.

Inhibitor concentration ($\text{M} \times 10^{-4}$)	Weight loss (mg cm^{-2})	Corrosion rate (mm year^{-1})	Inhibition efficiency (%)
0	13.08	48.3	–
1.8	4.17	15.5	68
3.6	2.62	9.74	82
5.4	1.31	4.83	90
7.2	0.91	3.21	93
9.0	0.91	3.21	93

Table 2. Polarization and EIS parameters for the corrosion of mild steel in 1 M HCl without and with piperacillin sodium at 30 °C.

Inhibitor concentration ($\text{M} \times 10^{-4}$)	Polarization resistance (Ωcm^2)	Inhibition efficiency (%)
0	15.6	–
1.8	72.1	78.3
3.6	104	85.0
5.4	142	89.0
7.2	253	93.8

(SDBS) as inhibitors for magnesium alloys has proved a substantial decrease of corrosion rate in seawater [54, 55].

8. Corrosion control by piperacillin sodium

A recent study performed by our group on the protection of mild steel in an acidic medium using a pharmaceutical inhibitor named piperacillin sodium revealed successful protection with an efficiency of 93%. Piperacillin sodium is the commercial name of (2S,5R,6R)-6-[[[(2R)-2-[(4-ethyl-2,3-dioxopiperazine-1-carbonyl)amino]-2-phenylacetyl]amino]-3,3-dimethyl-7-oxo-4-thia-1-azabicyclo [3.2.0]heptane-2-carboxylic acid, monosodium salt. Its molecular weight is 539.5 g mol^{-1} . Clinically, piperacillin sodium is used to treat intra-abdominal infections, respiratory and urinary tract infections, skin and soft-tissue infections, and other infections caused by susceptible organisms. The LD_{50} value of piperacillin sodium is $> 7000 \text{ mg kg}^{-1}$ in rats, which makes it attractive because of less toxicity. Figure 1 shows the molecular structure of piperacillin sodium.

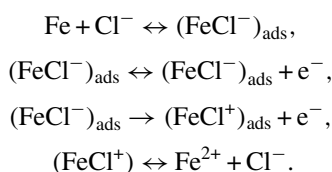
The effect of piperacillin sodium as corrosion inhibitor for mild steel was investigated using a weight-loss method as well as a three-electrode dc electrochemical technique [1, 3]. Results are shown in table 1. It is evident from the results that the corrosion rate decreased as the concentration of the inhibitor was increased up to $9 \times 10^{-4} \text{ M}$; an efficiency of 93% was exhibited at this concentration.

Linear polarization [3] exhibited a substantial rise in polarization resistance with an increase in inhibitor concentration, and showed similar efficiency, as shown in table 2.

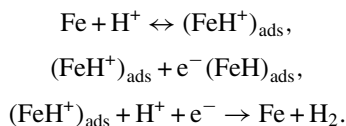
9. Mechanism of corrosion and inhibition

In hydrochloric acid solution the following mechanism is proposed for the corrosion of mild steel [55]. The anodic

dissolution mechanism of mild steel is



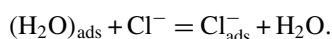
The cathodic hydrogen evolution mechanism is



Generally, the corrosion inhibition mechanism in an acid medium is adsorption of the inhibitor on the metal surface. The process of adsorption is influenced by different factors like the nature and charge of the metal, the chemical structure of the organic inhibitor and the type of aggressive electrolyte. The charge of the metal surface can be determined from the potential of zero charge (pzc) on the correlative scale (ϕ_c) [56] by the equation

$$\phi_c = E_{\text{corr}} - E_{q=0},$$

where $E_{q=0}$ is the potential of zero charge. However, in the present study the value of E_{corr} obtained in HCl is 472 mV versus SCE. In hydrochloric acid solution the sodium salt of the inhibitor is changed into free acid. The latter compound remains soluble due to the protonation of the NH group. Benerijee and Malhotra [57] reported the pzc of iron in hydrochloric acid solution is -530 mV versus SCE. Therefore, the value of ϕ_c is $+58$ mV versus SCE, so the metal surface acquires positive charge. The adsorption of cationic piperacillin species does not take place and the adsorption of Cl^- ion occurs and the surface becomes negatively charged. Now, due to the electrostatic attraction, the protonated Piperacillin molecules adsorb physically, i.e. physisorption, thereby giving high inhibition by Piperacillin molecules. In the presence of Cl^- ions, the following rapid reaction occurs on the metal surface:



The substitution of water molecules with Cl^- ions leads to the probability of the formation of adsorbed ion pairs and neutral molecules formed by the adsorbed Cl^- ions and piperacillin cations [58].

10. Conclusion

Inhibitors play a vital role in providing protection against corrosion. The selection of the inhibitor is important for environmental protection. Not all inhibitors are eco-friendly. There is a growing trend to use plant extracts and pharmaceutical compounds as corrosion inhibitors. Most of these compounds are environmentally-friendly. Piperacillin sodium proved to be an eco-friendly inhibitor for mild steel in 1 M HCl solution with an efficiency of 93% at 7×10^{-4} to 9×10^{-4} M concentration.

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