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Organic Compounds as Corrosion Inhibitors from Natural Products

Asmaa.J.AL-Lamei*, Najlaa.K.Muftin, F.M.Hussein, Amel S.Mahdi

Department of Chemistry ,College of Science/AL-Mustansiriyah University, Iraq.

Corresponding Author:** ***Asmaa.J.AL-Lamei, *Department of Chemistry, College of Science/AL-Mustansiriyah University, Iraq.*

Abstract: *Corrosion is an unavoidable but controllable process, as controlling corrosion of metals is of technical, economic, environmental and great importance. Intensive efforts are underway to develop new corrosion inhibitors of plant origin for minerals subject to different environmental conditions. Plants represent a class of exciting compound sources that are currently being examined for use in corrosion protection from minerals in most systems, as a potential alternative to toxic synthesis inhibitors. There are a great number of studies display the ability of organic compounds to inhibit corrosion . All these studies show that organic compounds, especially those that contain N, S and O, have demonstrated strong inhibition efficacy. However, many corrosion inhibitors are prohibited due to the environmental and toxic risks they create. It takes advantage of the natural extracts of plant materials. They are rich sources of ingredients that have a very high inhibition effect. This revised article briefly provides a detailed description of natural products as corrosion inhibitors in various corrosive mediums that have been used in the past few decades.*

Keywords: *Natural products, organic compounds, corrosion inhibition.*

1. INTRODUCTION

The term of "Green Chemistry" provides a suitable design of any related research in non-polluting way with minimal waste and toxic levels[1]. Over decades in corrosion-related researches field the main practical approach against corrosion is the use of synthetic organic inhibitors. Numerous organic compounds have been used as anti-corrosion inhibitors for various metals and alloys. The synthetic organic inhibitors obtain heteroatoms (O,N,S,and P) showed noticeable inhibition efficiency, but most of organic inhibitors are highly toxic and greatly affected both human and environment. The growing concern of the harmful of organic inhibitors led to discover alternative approaches instead. Harmful impact of the organic inhibitors forced the chemical suppliers and operators who had to follow with the increasing demands and more stringent rules that could contribute to minimize impact of the organic inhibitors to nature, humans and aquatic life. . So the benefit of natural originating inhibitors have been increase recently. Which provides better chances of being eco-friendly and harmless [2] . Green corrosion inhibitors are eco-friendly, non-toxic, biodegradable, cheap and do not contain heavy metals. In recent years many researchers have been successfully used naturally products in order to inhibit the corrosion of metals. They developed a green inhibitors such as plant extracts, fruits peel extracts, dried stems, leaves, seeds. And other natural products. The natural-occurring substances involve of many natural organic compounds such as pigments, flavonoids, ascorbic acid etc. the extracts of these substances constitute of nitrogen, oxygen and sulfur-containing compounds which make them good and effective anti-corrosion inhibitors [3]. S.S. Mahmoud [4] studied the corrosion of Muntz brass (63%Cu,37%Zn) in HCl solution using naturally occurring extracts which they are A-brown skin onion, B-onion bulb, C-the cloves of garlic blub, D-orange peel and E-henna leaves. J. C. da Rocha1et.al[5] investigated the Grape Pomace Extracts for carbon steel in1molL^{-1}HCl solution.. The inhibition action of the extract was examined using weight loss, potentiodynamic polarization, surface analysis spectroscopy. The grape extract was form to be active against steel corrosion. The inhibition ability was increased with increasing extract concentration and decreased with increasing temperature(T). The adsorption of the grape extract was result Langmuir adsorption isotherm. K. Agarwal [6] studied the inhibitive action of Lemon peel(LP) and Fenugreek leaves(FL) extracts on mild steel corrosion in1MHCl. The investigation was carried out by using electrochemical and non-electrochemical methods which confirmed that both of extracts are good inhibitors against corrosion. The electrochemical measurements recorded that both of inhibitors begin as mixed-type inhibitors. The adsorption of these inhibitors on the steel alloy followed Langmuir adsorption isotherm via

physisorption. A.Ismail and M. A.MohdTajuddin[7] studied the inhibition action of banana peel extract on stainless steel 304corrosion in sea water 3.5%NaCl. The electrochemical measurements revealed that the banana extract had an effective action against the breakdown of passive film on the steel alloy. The addition of the extract inhibited the localized attack of the corrosive medium. A.M.Al-Fakih et.al[8] studied the effect of turmeric and ginger extracts on mild steel corrosion in1M HCl. Their inhibition performance was investigated using weight loss method and potentiodynamic polarization. Weight loss measurements were carried out at various temperatures for 1himmersion time. The polarization measurements revealed that both of inhibitors acted as mixed-type inhibitors. The present work has been undertaken which enlightens, studies on potential green corrosion inhibitors for metals in different media.

2. General Concepts of Corrosion

Corrosion terminology comes from a Latin word "corrodere" which means (tognaw),corrosion can be defined in different ways but the most interpretation definition of the term is a destructive attack on the metallic materials(metals) with in the surrounded environments. It's an electrochemical reaction and ordinarily begins at a metal surface which can be determined by either the change in weight of a given metal or by change in the chemical or physical properties with time[9] . Corrosion causes consequences series that become globally as significant problem; besides our everyday experiencing with this kind of degradation it also leads to shut down of plants, destruction of metallic infra structures and waste valuable resources which required a costly maintenance. In order to minimize its effect, major corporations, establishments and governments have been recruited groups and committees to investigate corrosion-issues related. Corrosion control is done mechanisms and by using corrosion-resistant materials as possible, protective systems or devices[10] . Corrosion is undesired phenomenon naturally occurred due to the fact that metals and their temporary unstable form of a pure metallic state have great tendency to convert to a high thermodynamically stable form which represent as ores or natural minerals that’s the main reason metals are existed in many forms such as oxides or hydroxides, etc., according to their existing environment[11] .Corrosion prevention is the most important topic in industrial engineering as a result its necessary to have a full awareness insight in to electrochemistry, physical chemistry, chemical metallurgy and composition such of these related areas of knowledge is what the corrosion fundamentals is[12] based on.

Corrosion Forms

Corrosion can be classified in to several categories; these types can occur to all metallic materials and could operate and propagate by different mechanisms. With all types of corrosion there are many mutual factors that could influence the rate of corrosion. The most important factor that governing corrosion are the aggressivity of medium, degree of acidity(pH), temperature, concentration, the aeration and degree of oxidizing power. Corrosion types can be classified as follows:

Uniform Corrosion

A class of corrosion that occurred when a metal surface corroded at the same rate. When a metal placed in a corrosive medium the oxidizing sites(anode areas) shift to different positions until the entire metal surface has been anodic at the same time[13].

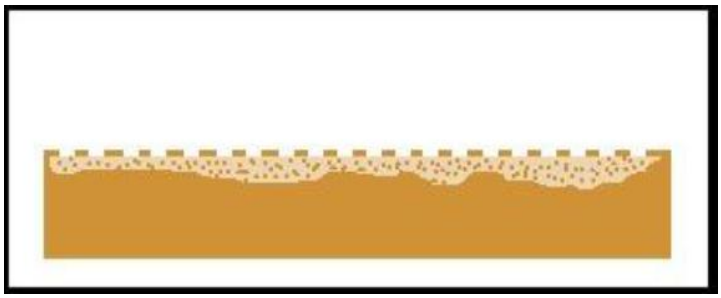

Figure1. *Uniform Corrosion*

Erosion Corrosion

Erosion corrosion("Flow-Assisted") refers to corrosion brought via highly-relative velocity between a corrosive environment and the metal surface. Corrosive medium can sweep away the metal ions before they can deposit on the surface which depends on its reactivity[14].

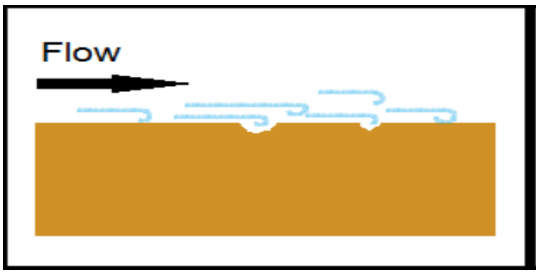

Figure2. *Erosion Corrosion*

Pitting Corrosion

Pitting corrosion represents a highly-localized attack on the surface resulting holes(pits) that could expand to dig in within the inside of a metal such of this type can be observed on an alloy surface that presented in certain environment [15].

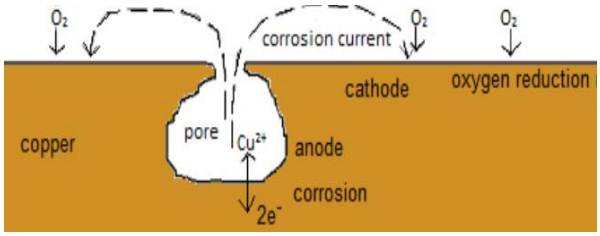

Figure3. *pitting corrosion*

Inter-granular Corrosion

The infra-microstructure of a metal makes up of grains ;these grains are separated by boundaries .Inter-granular corrosion can be defined as a localized attack occurred among these boundaries or adjacent to it. There are many reasons for this type of corrosion such as enrichment of a certain element in manufacture alloying operation, presence of the impurities and to the decrease of one alloying element in the boundaries [16].

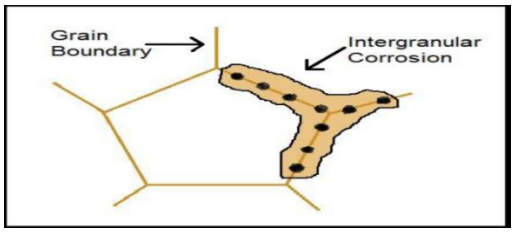

Figure4. *Inter-granular corrosion*

Galvanic Corrosion

A contactor dissimilar corrosion, such type of corrosion occurred when dissimilar metals are existed in a certain corrosion electrolyte one of them is noble relative to another. Galvanic corrosion depends on the standard potentials of the existing metals that arranged in the galvanic [17] series.

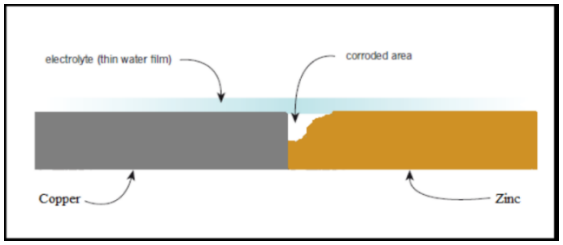

Figure5. *Illustration diagram for galvanic corrosion*

Stress Corrosion Cracking

Stress corrosion cracking promotes a crack fatigue within the metallic structure. This type of corrosion occurred when a metal experience a tensile and mechanical stress at the surface which leads to form a crack that expands within the metallic structure[18].

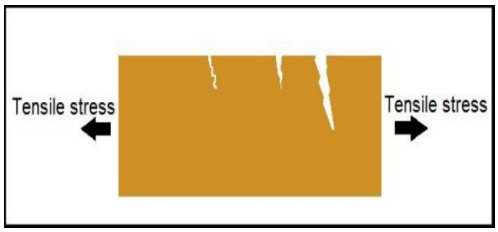

Figure6. *Stress cracking corrosion*

Crevice Corrosion

Crevice corrosion is a localized attack that occurred at the narrow spaces between two metals . The stagnant environment within the crevice can differentiate from the bulk environment which promotes the metal dissolution[19].

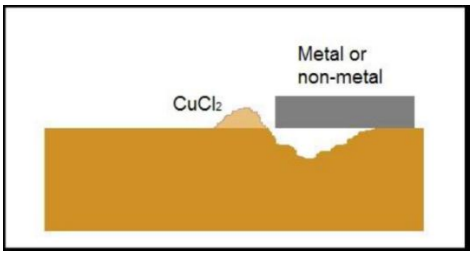

Figure7. *Illustration diagram for crevice corrosion*

Factors Influencing Corrosion:

The nature and degree of corrosion reliant on the environment and the metal. The significant factors which may impact the corrosion process are :

are : (a) Nature of the metal (b) Concentration of electrolyte (c) Environment (d) Temperature (e) Electrode potential and (f) Hydrogen over voltage .

Corrosion Inhibitors

Corrosion inhibitors can be describe in different ways but most applicable one is, chemical compounds when added in basic amounts to the corrosive environment they activate lowering the corrosion rate of a metal or an alloy without a honest change in the concentration or any other properties of this environment .Inhibitors can help the metal or alloy to enhance their resistance against corrosion by different corrosion inhibition mechanisms[20].

Over recent years, many researchers have dedicated considerable efforts to find suitable approaches for minimizing corrosion rate as much as possible. It's widely accepted that the most practical way to reduce the corrosion rate is by using inhibitors. The mechanism of the inhibition activity is not perfectly under stood however, there are many suggested mechanisms related the possibility of inhibitors to reduce the corrosion rate. Which they are:

-An adsorption process in which ions or molecules of the inhibitor adsorbed onto metal surface.

-increase/decrease in the catholic or anodic rate reactions.

-Decrease in the rate of diffusion of reactants on to the metal surface. In acid media, nitrogen-containing compounds, sulfur-base compounds, alkaloids, aldehydes and etc. are used as inhibitors.

The significant role of the inhibitors is to form a barrier of one or multi layers against the aggressive attack of acid solution. The formation of the protective film is greatly associated with chemical and/or physical adsorption mechanisms which involving a variation in the charge of the adsorbed molecules and transfer of charge from one site to the other .Those adsorbed substances(ions/molecules) are adsorbed at metal/solution interface and retard catholic or anodic reaction subsequently[21] reduce the corrosion rate .

Green Inhibitors

The term of "Green Chemistry" provides a suitable design of any related research in non-polluting way with minimal waste and toxic levels[1]**.** Green corrosion inhibitors are eco-friendly, non-toxic,

biodegradable, cheap and do not contain heavy metals. The inhibitors like plant extracts apparently possess biocompatibility due to their biological origin. Identical to the general classification of "inhibitors", "green inhibitors" can also be organize into two categories, namely organic green inhibitors and inorganic green inhibitors. Over decades in corrosion- related researches field the main practical approach against corrosion is the use of synthetic organic inhibitors. Numerous organic compounds have been used as anti -corrosion inhibitors for various metals and alloys. The synthetic organic inhibitors having hetero toms O, N, S, and P) showed remarkable inhibition efficiency, but most of organic inhibitors are highly toxic and greatly affected both human and environment. Several reports are available on the divers natural products used as green inhibitors as shown in Tables 1.

Table1. *Green inhibitors used for corrosion inhibition of Metal.*

Metal	Inhibitor source	Active ingredient	References
steel	Tamarind		**22**
steel	Tea leaves		**23**
C-Steel Ni,Zn	Lawsonia extract (Henna	Lawsone (2-hydroxy-1, 4-napthoquinone resin and tannin, coumarine, Gallic, acid, and sterols)	**24**
Mild steel	Musa sapientum peels (Banana peels)		**25**
Steel	Mango/orange peels		**26**
Al, steel	Aqueous extract of tobacco plant and its parts	Nicotine	**27**
Zn	Onion juice	S-containing acids (glutamyl peptides) S-(1-propenyl) L-cysteine sulfoxide, and S-2-carboxypropyl glutathione	**28**
Sn	Natural honey (acacia Chestnut)		**29**
Carbon steel	Natural amino acids—alanine, glycine, and leucine		**25**
Fe, Al	Benzoic acid		**30**

Organic green inhibitors

The natural-occurring substances contain of many natural organic compounds such as pigments, flavonoids, ascorbic acid etc. the exact of these substances constitute of nitrogen, oxygen and sulfur-have compounds which make them good and effective anti-corrosion inhibitors. O, N, and S are the active centers for the process of adsorption on the metal surface. The authors widely applicable of plant extracts [31,32], as well as other organic inhibitors [33,34] against corrosion of steel in acidic fluids. A.M. Al-Fakih et.al [8] studied the effects of turmeric and ginger extracts on mild steel corrosion in 1 M hydrogen chloride. The weightiness analysis and electrochemical impedance spectroscopic studies were used to determine the inhibition efficiency of the inhibitors. Even though that the extract of leaves and bark display important effect on the corrosion rate separately, the combination of these two extracts appeared rather higher efficiency. the inhibitive action of Lemon peel and Fenugreek leaves extracts was reported for corrosion of mild steel in 1 M HCl The research was carried out by using electrochemical and non- electrochemical methods which confirmed that both of extracts are good inhibitors against corrosion. The surface analysis using (XPS) revealed the layer formation of the plant extract on the surface of mild steel, the results indicated the able to be done formation of iron phosphate that was catalyzed by the formation of the iron-plant extract organo-metallic complex. A. Ismail and M. A. Mohd Tajuddin [7] studied the inhibition action of banana peel extract on stainless steel 304 corrosion in seawater 3.5% NaCl. . Leaves extract of Millingtonia hortensis for mild steel corrosion in 1N hydrochloric and 1N sulfonic acid solutions using mass loss measurements was studied by S. Kulandai Therese and V.G. Vasudha [35]. Common oils are one of the green inhibitors taken away plant origin.

M.Znini et al. declare the corrosion inhibition effect of essential oil obtained from Salvia aucheri mesatlantica for mild steel corrosion in 0.5 M H2SO4 solution using weight loss and electrochemical polarization measurements [36]. The component part of Pennyroyal oil was R-(+)-pulegone . The inhibition efficiency of the inhibitor increased with T, which clearly stated the chemisorption of the inhibitor on the surface of the steel. And the inhibitor was classified as cathodic form inhibitor. According to Pengju [37] , similarly, inhibition effect of Jojoba oil and Artemisia oil were studied for steel corrosion in HCl medium under different temperatures[38,39]. The inhibitory effect of natural clove oil for corrosion inhibition of Ni and its alloys in various concentrations of HCl solutions was reported by Abdullah et al [40]. Dahmani et al and Sethuraman et al described the corrosion inhibition of mild steel in acidic medium using various plant extracts for example, black pepper [41] . Sethuraman [42] and Bothi [43] has analysis the corrosion of mild steel in HCl acid as well in H_2SO_4. The inhibition of corrosion of SX 316 stainless steel in acid media using khilah seeds (Ammi visnaga) extract was designed by El-Etre [44] . The mechanism of adsorption of the inhibitor molecules were chiefly physisorption. Despite the fact that the decrease in the inhibition efficiency was attended at increase temperatures, the seed extract exhibited mostly 71% of inhibition efficiency for 120 ppm at 80 ^{o}C. The anti-corrosion behavior of lupine (Lupinous albus L., White lupine)) extract on the corrosion of steel in aqueous solution of H2SO4 and HCl acid was prepared by Abdel-Gaber et al [45] . The inhibition of corrosion of aluminium in NaOH solution using leaves and seed extract of Gossipium hirsutum L. (Upland Cotton)was studied [46]. The leaf extract showed higher inhibition efficiency (97%) than the seed extract (94%). Similary, the inhibition of corrosion of mild steel in H2SO4 acid solution using extracts of Carica papaya leaves, seeds, heartwood, and bark was studied [47]. The leaf extract appear higher inhibition efficiency compared to the extracts from seed, heart wood and bark. The corrosion inhibition of aluminium in both acidic and basic conditions from Gongronema latifolium extract was studied[48]. The inhibitor exhibit higher inhibition efficiency in acid solution than in NaOH. The suggested mechanism for the corrosion inhibition in acid solution was chemisorption, whereas physisorption was apply for the inhibitor's effectiveness against sodium hydroxcied solution. Corrosion inhibition of zinc in acid media was examined using Aloe vera gel extract [49]. in this case, the gel was pressed out from fresh leaves. The juice was accordingly diluted and then the solution was directly studied acid corrosion of zinc. El-Etre consistent the inhibition probable of the wet extract of zallouh seed for corrosion of carbon steel in hydrochloric acid solution [50] .The rather low value of activation energy in the presence of zallouh extract marked that the mechanism of inhibition was provided by the physical adsorption of inhibitor molecules on the steel surface.

Inorganic green inhibitors:

Inorganic green inhibitors Corrosion is a simple process; the relation rate of corrosion is relevant to the change in standard Gibbs free energy (ΔG°). The reaction's automation is relevant to the negative value of Gibbs free energy , i.e. higher corrosion rate [51]. Metals and alloys when exposed the environment corrode to form stable corrosion products [52]. Inorganic elements or metals have a crucial role in living organisms, when they are at trace amounts. The higher concentrations of many metals cause toxicity to all forms of lives. agreements applicable It for the derivatives of metals. The appearing step in forming a corrosion protective film or coat in the presence of different media that occurs on metallic surfaces on the active sites is adsorption. many factors affect the adsorption of inhibitor on the metallic surface and isolate it including Temp., adsorption mode, type of electrolyte employed, and the nature and surface charge of metals [53]. The action of green inhibitors depends on the structure of the active ingredient; many researchers have supposed numerous theories to explain the mechanism of their effect. The operating ingredient derived from natural inhibitors changes from one plant species to another but their structures are strictly related to their organic coordinate. As an example, garlic contains allyl propyl disulfide, Anodic inhibitors operate by a significant shifting in corrosion potential towards anodic direction (noble direction). Inorganic anions such as molybdate, chromate and phosphate are usually used. The metal is display a special sort of layer called " nonviolent layer" which is formed as a result of the adsorption of these anions on the metal surface. The passivation experience that develop for some metals in certain environments is not really understand however, the formation of passive layer depends on the amount of oxygen, the nature of the environment and the concentration of aggressive ions[54]. Seeds of mustard to have an alkaloid berberine that has a long chain of aromatic rings and an N atom in the ring. The high inhibition_efficiency, chromates exhibit high toxicity and as a result prohibited to

use for industrial applications [55] . lanthanide salts are found to appear excellent inhibition properties [56, 57] . lanthanide salts can be constituent as green inhibitor or eco-friendly inhibitor. Lanthanide salts alike lanthanide chlorides were described to possess toxicity that is analogous to sodium chloride . A few research application were reported on the corrosion inhibition properties of lanthanides all along the last decade. Thoughts the nitrate ion is considered as an anodic inhibitor, Interestingly, for rare earth chlorides decrease in inhibition efficiency was observed on increasing the inhibitor concentration. This negative effect was interpreted as due to the increase in the concentration of the chloride ions.

Effect of Functional Groups on Inhibitors

Inhibitors can a tie to metal surfaces not only by electrostatic interaction but also by electron transfer to the metal to from a coordinate type of connection. This form of interaction is preferred by the presence in the metal of vacant electronic orbitals of low energy as they appear in transition metals. Electron transmission from the adsorbed group is favored by the presence of proportion relatively bound electrons, such as may be found in anions and neutral organic molecules containing lone pair of electrons or π-electron systems associated with multiple bonds or aromatic rings. In organic compounds adept lone pair of electrons for coordinate bonding occur in functional groups containing elements of group V and VI of the periodic table. The affection to stronger coordination bond formation and hence active adsorption by these elements increases with decreasing electro-negativities in the order O < N < S < Se[58,59] and also depends on the nature of the functional groups containing these elements.ng these elements.

Corrosion Inhibitors Classification

Corrosion inhibitors being selected according to various critica such as metal and environment. There is no particular classification for inhibitors however, the most general and comprehensive one is presented in figure (8)[59]

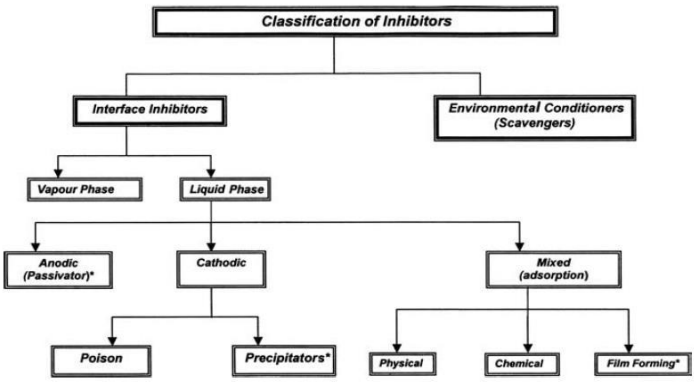

Figure8. *General Classification of Inhibitors*

Environmental Conditioners (Scavengers) One of most effective approach against corrosion is the removal of active corrosive species from an environment. The scavengers or cordial have ability to scavenging the active substances and reducing the corrosion rate. In oxygenate acidic media the reduction of oxygen can be the main cathodic reaction besides the evolution of hydrogen gas. Like of these situations the oxygen scavengers show an important role in decrease the oxygen content and resulted to decrease in the corrosion rate. Sodium sulfite (Na_2SO_3) is most common oxygen scavenger it's reacted with oxygen to produce sodium sulfate as shown in the equation[59]:

$2Na_2SO_3 + O_2 \rightarrow 2Na_2SO_4$

Interface Inhibitors

The interface inhibitors suppress the corrosion of a metal by create a film at metal/ electrolyte interface. The interface inhibitors can be classify in to vapor-and liquid-phase inhibitors[59].

Vapor-Phase Inhibitors:

Vapor-phase inhibitors abbreviated(VPIs)are the inhibitors that can be used in closed environment to provide a temporary protection against corrosion. They operate via either in a molecular form or in a

dissociate form. The pair of these assemble can be adsorbed on a metal surface and decrease the corrosion rate [59]

Liquid-Phase Inhibitors

Liquid-phase inhibitors can be classified according to their inhibitive action mechanism anodic, cathodic or mix-type inhibitors.

Anodic Inhibitors(Inorganic Inhibitors)

Anodic inhibitors operate by a significant shifting in corrosion potential towards anodic direction(noble direction).They block the anodic active sites by forming a protective layer; hence inhibit the dissolution of a metal. In organic anions such as molybdate, chromate and phosphate are usually used. The metal is display a special type of coating called" apathetic layer" which is formed by reason of the adsorption of these anions on the metal surface. The passivation experience that occurred for some metals in convinced environments is not actually understand however, the formation of passive layer depends on the amount of oxygen, the nature of the environment and the concentration of aggressive ions[59].

Cathodic Inhibitors

Cathodic inhibitors can reduce the corrosion rate by either reduce the reduction rate (cathodic poisons)or by a selective precipitating on cathodic activesties. instances the cathodic inhibitors decrease cathodic reaction rate by reduce the diffusion of reducible (oxygen/hydrogen) active species on metal[59] surface hence reduce the corrosion rate .

Mixed-type Inhibitors (Organic Inhibitors)

Mixed-type inhibitors also called(Adsorption inhibitors) are inhibitors that can be adsorbed on both cathodic and anodic active sites as a result they can retard both cathodic and anodic reactions and subsequently reduce the corrosion rate. About80% of mixed-type inhibitors are organic inhibitors. The greater important basic property of organic inhibitors effectiveness comes from its adsorption nature which is depend on the chemical structure of the inhibitors, type of environment and surface charge of the metal. The adsorption case of organic inhibitors occurred by two possible mechanisms: physisorption and chemisorption. The physical adsorption represents an electrostatic interaction between the metal surface and inhibitor's charged species despite occurring rapidly, physisorption breaks down with increasing temperature. Chemisorption contain charge transfer share between the adsorbed inhibitor molecules and the metal surface. Since chemisorption involves formation of a coordinated bond between adsorbed inhibitor molecules and the metal surface is required longer time to formulate than physisorption. In contrast of physisorption, the chemisorption is increased with increasing temperature. Chemisorption is greatly depends on the inhibitor molecular structure. Since chemisorption involves electron donation, the inhibitors must be contain heteroatoms. Heterocyclic compounds containing double or triple bonds, hydrocarbon chain, sulfur containing compounds and nitrogen containing compounds are considered to be effective inhibitors. The ability of a compound to adsorb chemically depends on amount of heteroatoms and their chemical component such as electronegativity and basicity. The greater electronegativity of an atom the lower its tendency to coordinate as the follow[60]order : O < N < S < P Increase the electronegativity Increase the basicity(Inhibition performance).

Plant extracts as corrosion inhibitors

Plant extracts to compose various organic compounds which have corrosion inhibiting stability. The explain of natural products origin as corrosion inhibitors is becoming the subject of amount investigation due principally to the low cost and eco-friendliness of these products, and is fast replacing the synthetic and expensive hazardous organic inhibitors. Plant extracts established different organic compounds which have corrosion inhibiting abilities. Produced of these compounds as well as the corrosion inhibition abilities modify widely depending on the part of the plant [61] and its location. The extracts from the leaves, seeds, heartwood, roots and fruits of plants have been reported to inhibit metallic corrosion in acidic media [61,62,63]. A summary of plants extracts used as corrosion inhibitors have recently been given in Lekan Taofeek Popoola [60] and Klodian and Matjaž Finšgar [63] .

3. CONCLUSION

Generally green inhibitors are very good inhibitors under a range of corrosive environments for max of the metals. The non-toxicity and biodegradability are the extensive favor for these inhibitors. As to; they do have achievement boundaries. Despite the number of publications are witnessing the green inhibitors as a potential candidate against corrosion at different environments, further research efforts are needed to apply the green inhibitors widely at an industrial level. This article has shown that the usage of such eco-friendly corrosion inhibitors is the only way onward in the search for safer and environmentally secure protection against metal corrosion**.**

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