Plant extracts as corrosion and scale inhibitors: A review

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Abstract

The definition, classification, properties, chemical composition and uses of plant extracts were reported in this review article. Corrosion of metals and alloys influences infrastructures, buildings, industrial economy, modern and old ancient works. Protection of metals and alloys against corrosion can be achieved by using different methods such as cathodic protection, environmental control, anticorrosion coatings and adding inhibitors. Plant extracts were studied as low cost eco-friendly corrosion inhibitors. They not contain heavy metals or other toxic compounds. In addition they are biodegradable and renewable source of materials. Plant extracts were classified into inhibitors for corrosion of the following metals and alloys in different aggressive corrosion media: carbon steel, aluminium and its alloys, copper and its alloys, zinc and its alloys and nickel and its alloys. Mineral scale deposits in industrial water represent a major problem leading to unexpected shutdowns and costly chemical or cleaning actions. Control of scale formation including acidification, ion exchange softening and scale inhibitors addition. The environment concerns lead the research to develop new environmentally friendly chemicals, currently named "Green" scale inhibitors. Plant extracts are recently used as green antiscalents and classified into inhibition of crystallization of each of carbonates and sulphates.

Keywords: *corrosion inhibitors, antiscalants, plant extracts, steel, aluminium, copper, nickel.*

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I. Plant extracts

It is estimated that there are 250,000 to 500,000 species of plants on Earth [1, 2], and botanists are still discovering around 2000 more every year [3]. The majority of the plants used by humans are dedicated to medicine, phytotherapy in particular. A passing fad for some, medicine for others, this traditional practice is based on the use of the natural therapeutic properties of plants in treating pathologies. Although it is the oldest care technique in existence, it has been supported by various types of research (clinical and pharmacological). In particular, its success arises from a deeper understanding of plants and from a qualitative selection of varieties.

A plant extract is a substance or an active with desirable properties that is removed from the tissue of a plant, usually by treating it with a solvent, to be used for a particular purpose. Plant extraction is solid/liquid extraction, eventually followed by purification stages. It is thus defined as an operation of the separation of one or several constituents (solid or liquid) contained in a solid object by solubilisation in a fluid. This fluid, generally known as a solvent, may be a liquid or a gas (water vapour or supercritical fluids).

Plants contain many types of chemical constituents such as: alkaloids, glycosides, organic acids, resins (including resin acids, resin alcohols and hydrocarbon resins), volatile oils, sugars (including starches, inulin, gums and phlegmatic, *etc*.) , amino acids, proteins and enzymes, tannins, plant pigments (including chlorophyll, carotenoids, flavonoids, beet red bases and quinones, *etc*.), oils and waxes, and inorganic ingredients (trace elements) [4, 5].

According to the formulations of plant extracts, it can be divided into: water-soluble plant extracts (including water and propylene glycol, propylene glycol, butylene glycol, and glycerol extract), oil-soluble plant extracts (including various vegetable oils, such as the exact of sunflower oil, coconut oil and olive extract oil, sometimes use isopropyl myristate extract), essential oils, spray-dried powder, enzyme-hydrolyzed vegetable protein powder, the pure active ingredients, peeled fruit core powder, liposome encapsulated microcapsules, polysaccharides or other porous polymer-encapsulated microcapsules and microspheres absorbed extract. It has been also used of freshly prepared fruit or vegetable juice in the professional beauty salon or family. If the plant parts used for the plant extract are different, the active ingredients are also different, including: roots, stems, leaves, bark, flowers, garland, fruits, seeds, shoots and the like. This classification method is more meaningful in the preparation, application, storage and transportation, and is used frequently by the manufacturer of some plant extracts.

According to markets and markets analysis, the plant extracts market is estimated to be valued at USD 23.7 billion in 2019 and is projected to reach USD 59.4 billion by 2025, at a CAGR of 16.5% from 2019 to 2025 [6].

Besides nutrition and pharmaceutical purposes, plant extracts were used also in many applications. For example, the biosynthesis of silver nanoparticles (Ag NPs) using extract of *Salvia spinosa* [7]. Antimicrobial assay verified bactericidal activity of biosynthesized Ag NPs against Gram-positive and Gram-negative bacteria. According to the results, by growing the plants under controlled conditions, it is feasible to synthesize nanoparticles with desired properties. Moreover, Cassiolato *et al*. [8] estimated the efficiency of plant extract to neutralize soil acidity. Three laboratory methods were evaluated: $\Sigma(Ca+Mg+K)$ of the plant extracts; electrical conductivity of the plant extracts and titration of plant extracts with NaOH solution between pH 3 to 7. These methods were compared with the effect of the plant extracts on acid soil chemistry. All laboratory methods were related with soil reaction. Increasing $\Sigma(Ca+Mg+K)$, electrical conductivity and the volume of NaOH solution spent to neutralize H^+ ion of the plant extracts were correlated with the effect of plant extract on increasing soil pH and exchangeable Ca and decreasing exchangeable Al. During the last years, plant extracts were employed as scale inhibitors in many industrial applications in order to develop new cleaning chemicals for green environment.

II. Plant extracts as corrosion inhibitors

Corrosion of metals and alloys influences industrial economy, infrastructures, buildings, modern and ancient artworks. Due to atmospheric interaction and exposure to aggressive environments, such as NaCl and acid solutions, metals and alloys undergo corrosion. Corresponding to the spontaneous return of metals to their ores. Protection of metals against corrosion can be achieved by using different methods such as; cathodic protection, anticorrosion coating and commercial inhibitors. Various organic compounds have been used as corrosion inhibitors through adsorption on to the surface of the metals and alloys. The adsorption of corrosion inhibitors on the metal surface retards the anodic and/or cathodic reaction rates through forming passive film and/or changing the mechanism of corrosion. Unfortunately, most of the organic inhibitors are expensive and non-degradable which results in environmental pollution upon discharging. Recently, the increasing interest of the environmental preservations oriented many research efforts to replace the organic inhibitors with environmentally friendly materials. Plant extracts were studied as low cost and eco-friendly corrosion inhibitors; they also not contain heavy metals or other toxic compounds [9–11]. In addition, they are biodegradable and renewable source of materials. Plant extracts contain organic compounds such as tannin, organic and amino acids, alkaloids and pigments; they exhibit inhibitory action [12–14].

II.1. Plant extracts as inhibitors for corrosion of carbon steel

Carbon steel has excellent mechanical properties, good ductility, toughness, weldability and low cost, hence it can be used in different applications [15]. Typical applications include; pipelines, structural shapes, automobile body components, ships, bridges, tin cans *etc*. [16]. However, in many applications the corrosion results in the destruction and degradation of steel, minimizing its lifetime. One of the most effective and economic methods to protect metals against corrosion is using inhibitors.

II.1.a. Plant extracts as inhibitors for corrosion of carbon steel in acid media

G. Gunasekaran *et al*. (2004) [17] studied he inhibitive effect of *Zanthoxylum alatum* plant extract on the corrosion of mild steel in 20, 50 and 88% aqueous phosphoric acid using weight loss and electrochemical impedance spectroscopy technique. The extract was able to reduce the corrosion of steel more effectively in 88% than in 20% phosphoric acid. Results indicated that this extract is effective up to 70°C. Surface analysis (XPS) and (FT-IR) were carried out to analyses the mechanism of corrosion inhibition of mild steel in phosphoric acid media.

K.O. Orubite *et al*. (2004) [18] studied the inhibitive action of the extract of the leaves of *Nypa fruticans* Wurmb on the corrosion of mild steel in hydrochloric acid solution using weight loss and hydrogen gas evolution techniques. The highest of 75.18% was observed.

The effect of extracts of Chamomile (*Chamaemelum mixtum* L.), Halfabar (*Cymbopogon proximus*), Black cumin (*Nigella sativa* L.), and Kidney bean (*Phaseolus vulgaris* L.) plants on the corrosion of steel in 1 M H_2SO_4 solution were studied by A.M. Abd-El-Gaber *et al*. (2006) [19] using potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The potentiodynamic results indicated that all extracts act as efficient mixed-type inhibitors. The inhibitive action of plant extracts was discussed on the basis of adsorption of stable complex at the steel surface. Theoretical fitting of different isotherms: Langmuir, Flory-Huggins and Kinetic-thermodynamic model were tested to clarify the nature of adsorption.

The inhibitive action of aqueous extract of garlic peel on the corrosion of carbon steel in 1 M HCl was studied by S.S.A.A. Pereira *et al*. (2012) [20]. The mechanism of the inhibition was discussed on the basis of the adsorption of the sulphur compound in the extract on the steel surface. The experimental results of adsorption fits Langmuir isotherm.

K. Krishnveni *et al*. (2013) [21] studied the inhibitive properties of the *Morinda tinctoria* plant leaves extract in the corrosion of steel in acid medium using colorimetric, weight loss, AC impedance and Tafel polarization techniques. The extract act as mixed-type inhibitor and the process of inhibition is through charge transfer.

The inhibitive action of chitosan extracted from *Archachatina marginata* shells on the corrosion of plain carbon steel in acid media was studied by A.E. Okoronkwo *et al*. (2014) [22] using weight loss and thermometric methods. Characterization of the obtained chitosan was obtained with Fourier transform infrared spectroscopy analysis. The results indicated that chitosan has good inhibition efficiency of 93.2%.

The inhibitive effect of the aqueous extract of *Musa paradisica* (Banana) peels on mild steel corrosion in 1 M HCl was studied by G. Ji (2015) [23] using weight loss, electrochemical impedance spectroscopy, Tafel polarization and atomic force microscopy techniques. The adsorption behavior of the extract was studied and the results indicated that, the adsorption process of the extract is controlled by Langmuir adsorption isotherm.

Pomegranate (*Punica granatum*) peel extract as green corrosion inhibitor for mild steel in hydrochloric acid solution was studied by H. Ashassi-Sorkhabi *et al*. (2015) [24] using weight loss, polarization and electrochemical impedance techniques. The results indicated that this extract is an efficient green inhibitor for corrosion of mild steel in HCl solution.

The inhibitory effect of *Gentiana olivieri* extract on the corrosion of mild steel in 0.5 M HCl was investigated by E. Baran *et al*. (2016) [25] using potentiodynamic polarization and electrochemical impedance spectroscopy techniques. Impedance results showed maximum inhibition efficiency of 93.7% at 800 ppm of the extract. Potentiodynamic polarization results showed that the extract act as a mixed-type inhibitor.

Phyllanthus amarus leaf extract has been investigated as a corrosion inhibitor for mild steel in 1 M HCl solution by K.K. Anupama *et al*. (2016) [26] using weight loss, potentiodynamic polarization and electrochemical impedance spectroscopy techniques. As concentration of the extract increased, inhibition efficiency also increased and finally reached 95% at 303 K. The quantum mechanical parameters well explained the effect of standard features on the electron donating ability of *Phyllanthus*.

The inhibitive effect of *Leptadenia pyrotechnica* extract on the corrosion of mild steel in H_2SO_4 solution was studied by G. Singh *et al.* (2016) [27] using weight loss and thermometric techniques. The results indicated that the extract gave inhibition efficiency of 87.04 at 0.8% extract in 1 N H_2SO_4.

The inhibitive effect of the extract of the leaves of *Alhagi maurorum*, *Morus nigra* and Apricot leaves extracts on the corrosion of steel in 0.5 M H_2SO_4 solution were studied by B.A. Abd-El-Nabey *et al.* (2016) [28] using weight loss, thermometric, potentiodynamic polarization and electrochemical impedance spectroscopy techniques. Effect of solvent of extraction on the inhibition efficiencies of the three extracts were discussed. Theoretical kinetic-thermodynamic model of adsorption of the extract on the steel surface was tested to fit the experimental data. The activation parameters of the corrosion reaction of steel with H_2SO_4 in the absence and presence of the three extracts were determined. The correlation between the thermodynamic and kinetic data and molecular structure of the chemical constituents of the three extracts were discussed.

Inhibition of the corrosion of mild steel in 1 M HCl solution by Lemon Balm extract was studied by N. Asadi *et al*. (2018) [29] using electrochemical and theoretical approaches. According to the electrochemical impedance spectroscopy results the maximum inhibition efficiency of 95% was obtained in a solution containing 800 ppm extract. The excellent corrosion inhibition effect of the extract on mild steel in HCl solution was related to the adsorption of active inhibitive compounds such as caryophyllene, germacrene, citral, luteolin, chlorogenic acid and rosmarinic acid on the mild steel surface.

Synergistic corrosion inhibition effect of rice husk extract and KI for mild steel in H_2SO_4 was studied by M. Pramudita *et al.* (2019) [30] using weight loss method. The adsorption of KI increased the efficiency of the extract and the highest efficiency was 95.89% at 1250 ppm. The experimental results indicated that, the adsorption process of the extract obeys Langmuir adsorption isotherm.

Peganum harmala seed extract was examined as corrosion inhibitor for mild steel in HCl solution by G. Bahlakeh *et al.* (2019) [31] using potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The presence of many electron donor atoms based on nitrogen and oxygen in the molecules of *Peganum harmala* seed extract is responsible for its effective adsorption behavior on the metal surface through chelation with iron atoms. Increase in the concentration of the extract and time of immersion leads to an increase of the inhibition efficiency up to 95%.

The inhibitory behavior of Mustard seed extract for mild steel in 1 M HCl solution was investigated by G. Bahlakeh *et al.* (2019) [32] using weight loss, potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The impedance results proved that the maximum inhibition efficiency reached 94% for 200 ppm of the extract. The study of adsorption isotherm showed that, the adsorption of Mustard seed extract molecules obeyed Langmuir isotherm.

The inhibition effect of Parsley (*Petroselinum sativum*) leaves extract on mild steel in 1 M HCl solution was investigated by M. Benarioua *et al.* (2019) [33] using weight loss, potentiodynamic polarization, electrochemical impedance spectroscopy and scanning electron microscopy. The inhibition efficiency increased with increase of the extract concentration up to 92.39% at 500 ppm. Polarization results showed that the extract acts as a mixed-type inhibitor and obeys Langmuir adsorption isotherm.

Borage flower aqueous extract as an environmentally sustainable corrosion inhibitor for acid corrosion of mild steel was studied by A. Dehghani *et al.* (2019) [34] using potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The impedance results showed that, with the increase in Borage flower extract content in HCl solution and immersion time the inhibition efficiency achieved 91% at 500 ppm after 5 hours.

Tamarindus indica aqueous extract as a new green inhibitor for mild steel corrosion in acidic media was studied by A. Dehghani *et al.* (2019) [35] using potentiodynamic polarization, electrochemical impedance spectroscopy, scanning electron microscopy and contact angle measurements. Impedance measurements showed maximum inhibition efficiency of 93% at 800 ppm extract after 2.5 hours steel immersion in aggressive media. The adsorption of the extract molecules on the steel surface was controlled by Langmuir isotherm.

V.C. Anadebe *et al.* (2019) [36] studied the application of pigeon pea leaf extract as anti-corrosion agent for mild steel in HCl solution. The work involved investigation of the corrosion inhibition process using combination of experimental, theoretical modeling and optimization studies. The inhibition efficiency of 87.13%, 91%, 92.1% and 90.7% were obtained from the experimental studies of thermometric, gravimetric, potentiodynamic polarization and electrochemical impedance spectroscopy techniques respectively.

Ziziphora leaves extract was examined as green inhibitor for acidic corrosion of mild steel by A. Dehghani *et al*. (2020) [37]. The structure of extract photochemical was characterized by Fourier transform infrared spectroscopy (FT-IR) and ultraviolet-visible (UV-Vis) spectroscopy. Electrochemical impedance spectroscopy analysis indicated that, this extract up to 800 ppm gave inhibition efficiency of 93% (after 2.5 hours). The polarization results showed that, the extract act as mixed-type inhibitor and its adsorption on the steel surface obeys Langmuir adsorption isotherm.

Dardagan Fruit extract was examined as ecofriendly corrosion inhibitor for mild steel in 1 M HCl by A. Sedik *et al*. (2020) [38] using electrochemical techniques. The surface techniques; scanning electron microscope, atomic force microscopy, energy dispersive x-ray spectroscopy, contact angle measurements was also performed to get information about the surface properties of mild steel. The results indicated that this extract has good protection efficiency for corrosion of mild steel in acid medium, it gave 92% inhibition efficiency after 1 hour and 97% after 6 hours immersion in 3000 ppm extract.

The extract of *Terminalia chebula* was examined as eco-friendly inhibitor for corrosion of carbon steel in acid media by A. Saxena *et al.* (2020) [39]. This extract showed great corrosion inhibition at specific concentration. The surface morphology of steel was studied using scanning electron microscopy and atomic force spectroscopy. The experimental results fits the Langmuir adsorption isotherm.

A. Saxena *et al.* (2020) [40] studied the inhibition characteristics of *Musa acuminata* for corrosion of low carbon steel in 0.5 M H_2SO_4 using weight loss, potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The results indicated that, this extract showed an effective corrosion inhibition for corrosion of low carbon steel in acid media.

II.1.b. Plant extracts as inhibitors for corrosion of carbon steel in neutral and alkaline media

Silene marmarica was studied as an environmentally friendly inhibitor for steel corrosion in 0.5 M H_2SO_4 by O.A. Abdullatef *et al*. (2015) [41] using potentiodynamic polarization techniques. The results showed that *Silene marmarica* inhibited the corrosion of steel in 0.5 M H_2SO_4 and the presence of iodide ions produced a synergistic effect. The adsorption behavior of *Silene marmarica* in absence and presence of iodide ions was formed to fit both Langmuir isotherm and the Kinetic–thermodynamic model.

The inhibition efficiencies of Kraft lignin (KL) and Soda lignin (SL) of the corrosion of mild steel in 3.5% NaCl solution at pHs 6 and 8 were studied by E. Akbarzadeh *et al*. (2011) [42] using weight loss, electrochemical techniques and surface analysis for 50–800 ppm inhibitor concentration at 25°C. Both KL and SL extracts acted as good inhibitors, the KL gave maximum inhibition efficiency of 95% and 92% for pH 6 and pH 8 respectively, whereas SL gave 97 and 95% inhibition efficiency for pH 6 and 8, respectively at 800 ppm of inhibitor concentration.

M. Sangeetha *et al*. (2011) [43] studied the inhibition characteristics of *Phyllanthus Amarus* extract (PAE)-Zn^{2+} system for controlling corrosion of carbon steel in aqueous solution containing 60 ppm Cl^{-} ion using weight loss method, the results indicated that the formulation containing of 2 ml of PAE and 25 ppm Zn^{2+} has 98% inhibition efficiency. Polarization study indicated that this system functions as mixed-type inhibitor, AC impedance spectra reveal that a protective film is formed on the steel surface, and FTIR spectra indicated that the protective film consists of Fe^{2+}-*Phyllanthus* complex.

D.E. Abd-El-khalek *et al.* (2012) [44] investigated the aqueous extract of *Nicotiana* leaves as corrosion inhibitor of steel in acidic and neutral chloride solutions using potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The extract was found to be more effective in controlling corrosion of steel in acidic solution than in neutral one.

D. Asra Awizar *et al.* (2013) [45] studied the corrosion inhibition efficiency of nano-silicate extracted from rice husk ash (RHA), particle size in the range 10–20 nm for carbon steel in distilled water using potentiodynamic polarization and weight loss measurements, the inhibition efficiency of nano-silicate attained 99% after 6 hours exposure.

V. Johnsirani *et al.* (2013) [46] studied the efficiency of the aqueous extract of *Eclipta alba* Leaves in controlling the corrosion of carbon steel in sea water using the weight loss method. The results indicated that the formulation consisting of 6 ml extract and 25 ml of Zn^{2+} has 92% inhibition efficiency in controlling corrosion of carbon steel in sea water. Polarization study showed that above system functions as a mixed type inhibitor. AC impedance spectra indicated that a protective film is formed on the metal surface which was analyzed by FTIR spectra and AFM analysis.

H. Wang *et al.* (2014) [47] studied scale and corrosion performance of tobacco rob as extract in artificial sea water using potentiodynamic polarization and static tests. The results showed that the extract had good scale and corrosion inhibition properties for carbon steel in artificial sea water.

M.A. Deyab (2016) [48] investigated the inhibition effect of Seaweed extract on the corrosion of mild steel in saline formation water by weight loss, polarization and EIS techniques. Polarization results indicated that Seaweed extract retards the anodic reaction. The inhibition efficiency increases with the increase in Seaweed extract concentration but decreases with the rise in temperature.

P. Parthipan *et al.* (2017) [49] studied the inhibition of the corrosion of carbon steel API 5lX by neem extract in presence of different bacterial strains (*Bacillus subtilis* A1, *Streptomyces parvus* B7, *Pseudomonas stutzeri* NA3 and *Acinetobacter baumannii* MN3) using weight loss method, potentiodynamic polarization and electrochemical impedance spectroscopy techniques and surface techniques; Fourier transform infrared spectroscopy (FT-IR). The results indicated that neem extract consists of azadirachtin and other phytochemical compound which was play a key role in control of bacterial biofilm on metal surface and inhibit the corrosion process.

M.A. Deyab *et al*. (2017) [50] investigated the anti-corrosion properties of lemongrass (*Cymbopogon citratus*) extract (LGE) for carbon steel in produced oilfield water using weight loss method and electrochemical impedance spectroscopy technique. They found that the extract serve as an effective corrosion inhibitor for carbon steel *via* adsorption of its constituents on the steel surface.

K. Nasr *et al*. (2018) [51] investigated the inhibition performance of the aqueous extract of *Matricaria recutita* chamomile on the corrosion of S235JR steel in 0.5 M NaCl using electrochemical impedance spectroscopy (EIS) and polarization measurements. They found that its inhibition efficiency increases with increasing the concentration of the extract and an increase in the immersion time. The optimum inhibition efficiency of chamomile extract, 98.9%, was achieved for 5235JR steel when immersed in 15% v/v of the extract for 2 hours.

V. Grudić *et al.* (2018) [52] studied the inhibition of the corrosion of X52 5L carbon steel in 0.5 M NaCl solution by propolis extract using the potentiodynamic polarization method. The results indicated that, the propolis extract acts as anodic type inhibitor whose efficiency increase with increasing temperature and with stirring of the solution.

F.E.T. Heakal *et al*. (2018) [53] studied the corrosion inhibition of the carbon steel by cornflower (*Centaurea cyanus*) extract (CFE) in harsh saline formation water using weight loss method, potentiodynamic polarization and EIS techniques. The polarization results indicated that, this extract acts as mixed-type inhibitor with protection efficiency of 69% at extract addition of 10 ppm.

Z. Mohammadi *et al*. (2018) [54] studied the inhibition efficiency of *Mazuj gall* extract in simulated cooling water. Potentiodynamic polarization, electrochemical impedance spectroscopy, gravimetric measurements and ultraviolet-visible spectroscopy were used to investigate the corrosion inhibition efficiency and mechanism. The scale inhibition efficiency of the extract was evaluated using static beaker testing and scanning electron microscopy. The results indicated that, 1000 rpm *Mazuj gall* extract provided corrosion inhibition efficiency of 94.3% and scale inhibition efficiency of 97.2% .

S. Devikala *et al*. (2019) [55] studied the inhibition properties of the extract of *Asafoetida* for the corrosion of mild steel in 3.5% NaCl solution using the potentiodynamic polarization and EIS techniques. The formation of protective layer on the surface of the metal was confirmed by FT-IR, XRD and SEM.

The role of *Juglans regia* green fruit shell (JRS) extract as corrosion inhibitor for mild steel in 3.5% NaCl solution was studied by S.A. Haddadi *et al*. (2019) [56] using potentiodynamic polarization, EIS and electrochemical current noise analysis. The inhibition efficiency of 1000 ppm JRS extract was 94% for steel plate in 3.5% NaCl solution. The EIS results indicated that, the inhibition efficiency increases with the immersion time progress up to 48 hour.

Z. Sanaei *et al*. (2019) [57] studied the synergistic inhibition capacity of Chicory leaves extract (CLE) combined with Zn^{2+} ion against mild steel corrosion in $Cl^{–}$ ion solutions. The results from electrochemical techniques indicated that the effective synergic corrosion inhibition impact of the CLE (100 ppm) in presence of Zn^{2+} (100 ppm) with inhibition power of 96%.

M.T. majd *et al*. (2019) [58] investigated the synergistic impact between the *Eucalyptus* leaves extract (ELE)and Zn^{2+} cation for inhibition of the corrosion of the mild steel in NaCl solution. Surface study was examined by ultra-violate visible analysis (UV-Vis), scanning electron microscope (SEM), energy dispersive spectroscopy (EDS), atomic force microscope (AFM), grazing incidence X-ray diffraction (GIXRD), Fourier transmission infrared (FTIR) spectroscopy and contact angle (CA) measurements. The corrosion rate measurements of the mild steel samples were carried out by electrochemical techniques in NaCl solution containing different ratios of EIE and Zn^{2+} ions. The results indicated that 200 ppm ELE + 600 ppm Zn^{2+} had the highest inhibition efficiency (90%) and synergistic impact.

M.T. majd *et al*. (2019) [59] studied inhibition system of Persian *Echium amoenum* (PEA) and zinc metal for mild steel protection in 3.5% NaCl solution. The functional groups and complexes is in the structure of PEA and PEA: Zn containing solution were investigated by UV-Vis analysis, however, The composition and morphology tests of surface films were carried out by Fourier transform infrared spectroscopy (FT-IR). Scanning electron microscope (SEM), atomic force microscope (AFM), nuclear magnetic resonance spectroscopy (NMR), grazing incident x-ray diffraction (GIXRD) analysis and static contact angle (CA) measurements. The electrochemical impedance spectroscopy (EIS) and potentiodynamic spectroscopy (PDs) were used to measure the corrosion rate of mild steel samples in NaCl solution containing different ratios of PEA: Zn system, the results indicated that, the 200 ppm PEA + 600 ppm Zn sample has the highest efficiency (95%).

M.T. majd *et al.* (2020) [60] investigated the inhibition characterization of the extract of Esfand seed (ESE) for the corrosion of low carbon steel in a saline solution. Surface and electrochemical analysis were carried out by ultraviolet-visible spectroscopy (UV-Vis), Fourier transform infrared spectroscopy (FT-IR), grazing incident x-ray diffraction (GIXRD) analysis, field emission scanning electron microscope (FE-SEM), energy dispersive spectroscopy (EDS), atomic force microscope (AFM), electrochemical impedance spectroscopy (EIS) and potentiodynamic methods. The electrochemical studies established the efficiencies of zinc cation addition to the extract containing solute on its inhibition efficiency. The inhibition efficiency of the low carbon steel samples was 98.8% after 265 hour immersion inhibited by 300 ppm ESE + 700 ppm Zn.

II.2. Plant extracts as inhibitors for corrosion of aluminium and its alloys

Due to the combination of the following properties, light weight (density = 2.7 $g\cdot cm^{-3}$), mechanical strength, good corrosion resistance and nontoxic quality, aluminium and its alloys have a great number of engineering application [61, 62]. Aluminium and its alloys are the popular choice for many critical applications in the field of aerospace, food industry, construction, heat exchange and electrical transmission. In many environments a naturally generated, protective oxide film provide aluminium with corrosion immunity. This film is amphoteric and can be dissolve when it is exposed to either acid (pH less than 5) or alkali (pH greater than 9 media. Aluminium and its alloys, due to their wide applications come in contact with acids or bases during pickling, descaling and electrochemical etching [63]. The corrosion resistance of aluminium and its alloys using inhibitors has been investigated in great variety of environments.

II.2.a. Plant extracts as inhibitors for corrosion of aluminium and its alloys in acid media

The inhibitive effect of *Hibiscus subdariffa* (karkade) extract as green corrosion inhibitor for aluminium and zinc in 2 N HCl solution were studied by El-Hosary *et al*. (1972) [64]. They studied also (1993) [65] the inhibition of corrosion of aluminium, copper, and steel by Molasses.

G.O. Avwiri *et al.* (2003) [66] studied the action of *Vernonia amygdalina* (biter leaf) on the acidic corrosion of 25 and 3RS aluminium alloys using weight loss method. The results showed he highest inhibition efficiency of 49.5% for 0.1 M HCl and 72.5% for 0.1 M HNO_3 respectively.

The inhibitive action of the mucilage extracted from modified stems of prickly pears toward acid corrosion of aluminium was investigated by A.Y. El-Etre (2003) [67] using weight loss, thermometry, hydrogen gas evolution and polarization techniques. It was found that the extract act as a good inhibitor for corrosion of aluminium in 2 M HCl solution and its inhibition action was discussed in view of Langmuir adsorption isotherm. It was found also that the *Opuntia* extract provides a good protection to aluminium against pitting corrosion in chloride ion containing solution.

The leaf extracts of *Chlomolaena odorata* L. were studied for inhibition of the corrosion of aluminium in 2 M HCl by I.B. Obot *et al*. (2010) [68] using gasometric and thermometric techniques. Results indicated that this extract functioned as an excellent inhibitor for corrosion of aluminium in the acidic solutions. The adsorption of the components of the extract on the aluminium surface obeys Langmuir isotherm.

O.K. Abiola *et al*. (2010) [69] studied the inhibitive effect of *Cocos nucifera* L. for the corrosion of aluminium in 0.5 M HCl solution using chemical technique. The results showed that this extract showed significant inhibition for corrosion of aluminium with 93% efficiency at highest concentration. The inhibition action was attributed to the adsorption of the extract components molecules on the aluminium surface following Langmuir adsorption isotherm.

The corrosion inhibition effect of *Aningeria robusta* extract for aluminium in 2 M HCl solution was investigated by I.B. Obot *et al.* (2011) [70] using hydrogen evolution method at 30 and 60°C. Inhibition efficiency of the extract increased with an increase of the *Aningeria robusta* extract and the inhibition efficiency synergistically increased on addition of potassium iodide.

The inhibitive effect of leaf extract of *Euphorbia hirta* on aluminium corrosion in HCl and NaOH solutions were studied by L.A. Nnanna *et al*. (2011) [71] using weight loss method at 30 and 60°C. This extract is more effective for corrosion of aluminium in the acidic medium than in the alkaline medium. The adsorption characteristics of the leaf extract were described by Langmuir isotherm.

M.M. Fares *et al*. (2012) [72] studied the inhibition behavior of pectin for corrosion of aluminium in hydrochloric acid solution. The inhibition efficiency first increased linearly up to 2.0 g/L – and then it continues steady exponential increase until it reached plateau. The maximum inhibition efficiency obtained at 10°C using pectin concentration = 8.0 g/L was = 91%, whereas at 40°C it severally declined to 31%. Adsorption of pectin macromolecule on aluminium surface demonstrated proper Langmuir isotherm fit.

X. Li *et al*. (2012) [73] studied the inhibitive effect of *Dendrocalamus brandisii* extract on the corrosion of aluminium in HCl and H_3PO_4 solution using weight loss, potentiodynamic polarization and electrochemical impedance spectroscopy method. The results showed that it is a good inhibitor in 1 M HCl solution while a moderate inhibitor in H_3PO_4. It acts as cathodic inhibitor in HCl solution, while it acts as mixed-type inhibitor in H_3PO_4.

S. Deng *et al*. (2012) [74] studied the inhibitive effect of *Jasminum nudiflorum* Lindl. leaves extract on the corrosion of aluminium in HCl solution using weight loss, potentiodynamic polarization curves, electrochemical impedance spectroscopy and scanning electron microscope methods. The results indicated that this extract acts as a good inhibitor for aluminium corrosion in 1 M HCl, its adsorption obeys Langmuir adsorption isotherm and acts as cathodic type inhibitor.

A.I. Ali *et al*. (2012) [75] studied the inhibition of aluminium corrosion in 2 M HCl by black mulberry (*Morus nigra* L.) using weight loss, electrochemical polarization technique and hydrogen evolution measurements. Black mulberry extract acts as good inhibitor for aluminium corrosion in the acid solution. The inhibitive action of this extract was discussed in view of the adsorption of its components on the aluminium surface.

The effect of *Commiphora pedunculata* gum on the corrosion of aluminium alloy AA 3001 in HCl solution was studied by P.O. Ameh *et al*. (2014) [76] using gravimetric and thermometric methods. The results indicated that this gum is a good adsorption inhibitor for the corrosion of aluminium in HCl solution.

The effect of beetroot (betanin) extract on the corrosion inhibition of aluminium in aqueous solution at pH 3 has been investigated by A. Nithya *et al.* (2015) [77] using mass loss method, potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The results indicated that betanin extract is a good green inhibitor for aluminium in an aqueous solution at pH 3. It acts as a mixed-type inhibitor and maximum inhibition efficiency 98% was obtained at 2 ml extract.

M. Akin *et al.* (2015) [78] studied the inhibitive action of *Juglans regia* L. extract as green inhibitor for stainless steel and aluminium in 1 M HCl using weight loss method, potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The inhibition efficiency was found 88.8% for aluminium. The effect of immersion time (2–8 hours) was discussed and adsorption mechanism was fitted with Langmuir isotherm.

The inhibition behavior of *Thymus algeriensis* extract on the corrosion of 2024 Al alloy was investigated by A. Khadraoui *et al.* (2016) [79] using weight loss, gasometry and electrochemical impedance spectroscopy techniques. The inhibition efficiency of the *Thymus algeriensis* extract is 78.7% at 0.75 g/L. The inhibitive effect of the extract was discussed in view of adsorption of its components on the aluminium surface following Langmuir isotherm.

Y.C. Sharma *et al*. (2016) [80] studied the corrosion inhibition of ethanol extract of *Psidium guajava* seeds for aluminium in 0.5 M HCl using weight loss, FTIR spectroscopy and SEM analysis techniques. The results showed that, this extract performed well as inhibitor for the corrosion of aluminium in hydrochloric acid solution and the inhibition mechanism was by adsorption process through Langmuir isotherm.

O.U. Abakedi *et al*. (2016) [81] studied the inhibition effect of the extract of *Maesobatrya barteri* root extract on the corrosion of aluminium in hydrochloric acid solution using gasometric and thermometric methods. The adsorption of the components of the extract on the aluminium surface obeyed Langmuir adsorption isotherm and the adsorption process was physical in nature.

The inhibitive effect of the as-synthesized baicalin derivatives was investigated by Y.T. Du *et al*. (2017) [82] as ecofriendly green inhibitor on the corrosion of aluminium in 1 M HCl solution using weight loss and electrochemical techniques. The results indicated that the bacalin derivatives exhibited excellent performance as inhibitor for aluminium corrosion and acted as mixed-type inhibitors. The maximum inhibition efficiency at 25°C is 95% at 0.9 g/L. The experimental results of adsorption obeys Langmuir adsorption isotherm.

Polygonatum odoratum extract was studied as corrosion inhibitor for aluminium in 1 M HCl by M. Prabakaran *et al.* (2018) [83] using weight loss, potentiodynamic polarization, electrochemical impedance spectroscopy and scanning electron microscope techniques. This extract acts as mixed-type inhibitor.

The inhibition of the corrosion of aluminium in 0.5 M HCl solution by Areca palm leaves extract was studied by N. Raghavendra *et al.* (2018) [84] using weight loss, AC impedance spectroscopy, potentiodynamic polarization and scanning electron microscopy methods. The adsorption of molecules of the components of the extract on the aluminium surface fitted Langmuir adsorption isotherm.

S. Bashir *et al.* (2019) [85] studied the corrosion inhibition behavior of Shatavari (*Asparagus racemosus*) for aluminium in acid medium using weight loss, quantum chemical analysis and scanning electron microscopy. The results indicated that 4000 ppm concentration of the extract give inhibition efficiency of 72.28%. The experimental results obeys Langmuir adsorption isotherm.

II.2.b. Plant extracts as inhibitors for corrosion of aluminium and its alloys in neutral and alkaline media

The inhibitive action of the extract of the leaves of *Sansevieria trifasciata* on aluminium corrosion in 2 M HCl and 2 M KOH solutions were studied by E.E. Oguzie (2007) [86] using gasometric technique. The results indicated that this extract functioned as a good inhibitor in both environments. Synergistic effects increased the inhibition efficiency in the presence of halide ions. A mechanism of physical adsorption was proposed of the inhibition behavior and the adsorption process obeyed Freundlich isotherm.

A.M. Abdel-Gaber *et al.* (2008) [87] studied the inhibition of the aluminium corrosion in 2 M NaOH by damsisa (*Ambrosia maritime* L.) extract using chemical and electrochemical techniques. The extract behaved as mixed-type inhibitor. The results indicated that the damsisa extract could serve as an effective inhibitor for corrosion of aluminium in alkaline solutions. The impedance results verified the remarkable stability of extracts during storage up to 35 days.

O.K. Abiola *et al.* (2009) [88] studied the corrosion inhibition behavior of *Gossypium hirsutum* L. leave and seed extracts for aluminium in 2 M NaOH solution using chemical technique. The results indicated that the leave extract was more efficient than seed extract. The leave extract gave 97% inhibition efficiency while the seed extract gave 94% at the highest concentration.

R. Rosliza *et al.* (2010) [89] examined the use of Tapioca starch for improvement of corrosion resistance of AA 6061 alloy in sea water using gasometry, potentiodynamic polarization, linear polarization resistance and electrochemical impedance spectroscopy measurements. The results indicated that the presence of tapioca starch in the sea water decrease the corrosion rate, corrosion current density, double layer capacitance and increase the value of polarization resistance. The Langmuir adsorption isotherm fits well the experimental data.

J. Halambek *et al*. (2010) [90] studied the effect of natural oil extracted from *Lavandula angustifolia* L. oil as corrosion inhibitor for Al-3Mg in 3.5% NaCl solution using weight loss, polarization measurements and scanning electron microscope. It was found that this extract protect aluminium alloy against pitting corrosion in NaCl solution.

The corrosion behavior of aluminium in 0.5 M NaOH in presence of Azwain (*Trachyspermum copticum*) seed extract by S. Ambrish *et al*. (2012) [91] using weight loss and electrochemical techniques was studied. The experimental results indicated that the inhibition efficiency increases with increasing the extract concentration and reached 94% at 5000 ppm. Tafel polarization results indicated that the extract act as a mixed-type inhibitor. The adsorption of the molecules of the extract on the surface of aluminium followed Langmuir adsorption isotherm.

The inhibition characteristics of the aqueous extract of fruits of *Terminalia chebula* (TCE) was studied by D. Prabhu *et al*. (2013) [92] for the corrosion of 6063 aluminium alloy in 0.5 M NaOH solution using weight loss and potentiodynamic methods. Inhibition efficiency increased with concentration of the extract. The adsorption of the TCE extract on the metal surface obeyed the Langmuir adsorption isotherm, acted as mixed-type inhibitor and followed physical adsorption on the surface of the metal.

E.M. Nawafleh *et al*. (2013) [93] studied the inhibition efficiency of the extract of *Salvia judaica* on corrosion of aluminium in 1 M NaOH solution using weight loss method. Results indicated that the extract act as corrosion inhibitor for the corrosion of aluminium in 1 M NaOH. The adsorption of the extract on the aluminium surface obeyed Langmuir adsorption isotherm.

S. Geetha *et al*. (2013) [94] investigated the influence of the extract of *Vitex negundo* leaves on the corrosion of aluminium in 1 M NaOH using chemical and electrochemical methods. It was found that this extract act as corrosion inhibitor for alkaline corrosion of aluminium through a Langmuir adsorption isotherm.

J. Halambek *et al*. (2013) [95] investigated the effect of 30% v/v ethanolic solution of *Laurus nobilis* L. oil on the corrosion of aluminium and AA5754 aluminium alloy in 3% NaCl solution using weight loss method, potentiodynamic polarization and linear polarization resistance techniques. The results indicated that addition of 10 to 50 ppm oil concentration leads to the inhibition of the corrosion of aluminium and its alloy in the tested media.

M.K. Irshedat *et al.* (2013) [96] studied the effect of the extract of *Lupinus varius* L. on the corrosion of aluminium using weight loss method. This extract inhibited the alkaline corrosion of in aluminium 1 M NaOH solution and the inhibition efficiency of the extract increased with increasing the concentration of the extract and decreased with increasing temperature. The adsorption of the inhibition molecules on aluminium surface obeyed Langmuir adsorption isotherm.

The inhibition effect of *Sinapis alba* (SA) extract was examined for corrosion of aluminium in 1.0 M NaOH solution by T.T. Bataineh (2014) [97] using weight loss method, electrochemical polarization method and the surface morphology was analyzed by scanning electron microscopy. The inhibition efficiency increases with the extract concentration and reaches 79.98% with highest extract concentration at 50°C. This adsorption of the extract on aluminium surface obeyed Langmuir adsorption isotherm. Electrochemical polarization studies showed that the *Sinapis alba* extract acts as mixed-type inhibitor.

H.A. Fetouh *et al*. (2014) [98] studied the effect of aqueous extract of Damsisa, Lupine and Halfa-bar on the corrosion of 7075-T6 aluminium alloy in 0.5 M NaCl solution using electrochemical impedance spectroscopy and potentiodynamic polarization techniques. The results indicated that the three extracts act as cathodic-type inhibitors for the corrosion of aluminium alloy in the NaCl solution. Theoretical fitting of Langmuir, Flory Huggins adsorption isotherms and the kinetic-thermodynamic model were tested to clarify the adsorption mechanism.

H. Gerengi *et al.* (2015) [99] investigated the *Mimosa* extract as green corrosion inhibitor for AA6060 aluminium alloy in acid solution using electrochemical impedance spectroscopy (EIS) and dynamic electrochemical impedance spectroscopy (DEIS). The results indicated that this extract inhibited the corrosion of the aluminium alloy with 45% efficiency at 2750 ppm.

The inhibition effect of the aqueous extract of *Alstonia Scholaris* ark was studied by N. Chaubey *et al*. (2016) [100] for the corrosion of aluminium alloy in 1 M NaOH solution using gravimetric, potentiodynamic polarization, linear polarization resistance and electrochemical impedance spectroscopy techniques. The results showed that, this extract act as good inhibitor for corrosion of aluminium in 1 M NaOH solution with maximum efficiency of 92.6 at 20 g/L. The experimental results was found to fit the Langmuir adsorption isotherm.

The aqueous extract of *Neolamarckia cadamba* was investigated by N. Chaubey *et al.* (2015) [101] as corrosion inhibitor for aluminium alloy in 1 M NaOH using weight loss and polarization resistance, Tafel polarization and electrochemical impedance spectroscopy techniques. The results indicated that, this extract inhibited the corrosion of aluminium in NaOH solution with efficiency of 87% at 0.6 g/L and it acts as mixed-type inhibitor.

N. Chaubey (2015) [102] studied the inhibition of performance of the peels extracts of each of *Pisum sativum* (PS), *Solanum tuberosum* (ST) and *Citrus reticulata* (CR) on the corrosion of aluminium alloy in 1 M NaOH solution using weight loss, electrochemical impedance spectroscopy, linear polarization and potentiodynamic polarization techniques. The three peel extracts showed good performance, PS exhibited maximum inhibition efficiency of 94.5% at 1.5 g/L and behaved as mixed-type inhibitor.

The inhibitive effect of *Cleome droserifolia* leaves extract on the corrosion of aluminium metal in 1 M NaOH solution was studied by M.M.A. Qudah (2015) [103] using the weight loss method at different temperatures. The surface morphology of aluminium metal was analyzed using scanning electron microscope. The extract of 14 g/L at 35°C gave the highest inhibition efficiency of 78.6% and its adsorption on the aluminium surface obeyed Temkin adsorption isotherm.

The inhibition characteristics of the plants leaves extract of each of *Cannabis sativa* (CS), *Rauvolfia serpentina* (RS), *Cymbopogon citratus* (CC), *Annona squamosa* (AS) and *Adhatoda vasica* (AV) was studied by N. Chaubey *et al*. (2017) [104] as inhibitors for corrosion of aluminium alloy in alkaline media using gravimetric, electrochemical impedance spectroscopy, potentiodynamic polarization and linear polarization resistance techniques. The extracts showed maximum inhibition efficiency 97% at 0.2 g/L and act as mixed-type inhibitors.

The effect of Kalmegh leaf extract on the corrosion of aluminium in 1 M NaOH solution was studied by N. Chaubey (2017) [105] using electrochemical impedance spectroscopy and potentiodynamic polarization techniques. The results indicated that the inhibition efficiency of this extract increased with increasing of its concentration and maximum inhibition efficiency obtained was 82.45% at higher concentration. The adsorption of the inhibitor on aluminium surface obeyed Langmuir adsorption isotherm and it acts as mixed-type inhibitor.

The effect of stem bark extracts of three trees namely *Moringa oleifera* (MO), *Terminalia arjuna* (TA) and *Mangifera indica* (MI) on the corrosion of aluminium alloy in 1 M NaOH solution were studied by N. Chaubey *et al.* (2017) [106] using gravimetric, potentiodynamic polarization and electrochemical impedance spectroscopy measurements. The results indicated that the three extracts act as mixed-type inhibitors and obeyed Langmuir adsorption isotherm. MO extract exhibited the maximum inhibition efficiency of 85.3% at 0.6 g/L.

H. Elgahawi *et al*. (2017) [107] studied the inhibitive effect of *Limum usitaissimum* (ELUS) seeds extract on the corrosion of aluminium alloy AA 2024 in 3.5% NaCl solution using potentiodynamic polarization and electrochemical impedance spectroscopy and electrochemical noise measurement techniques. Results indicated that the extract of ELUS effectively inhibited the corrosion of aluminium alloy with efficiently range from 65 to 82% for the corresponding extract concentration from 80 to 1200 ppm. Potentiodynamic polarization results showed that ELUS act as cathodic-type inhibitor, however, the electrochemical impedance spectroscopy results indicated that the corrosion process was governed by charge transfer.

N. Raghavendra *et al.* (2018) [108] studied the effect of green color Arecanut husk extract as inhibitor for corrosion of aluminium in both 0.5 M HCl and 0.1 M NaOH solutions using weight loss method and potentiodynamic polarization technique. The influence of temperature and time on adsorption of the extract constituent on the surface of aluminium has been studied. The results indicated that the introduction of the extract appreciably decreased the rate of aluminium corrosion.

Konjac glucomanan was extracted from commercial product and studied by K. Zhang *et al*. (2018) [109] as a green corrosion inhibitor for AA5052 aluminium alloy in 3% NaCl solution using high performance gel permeation chromatography (GPC), thermo-gravimetric analysis (TGA), Fourier transform infrared (FT-IR) spectra, electrochemical measurements and surface characterization techniques. Potentiodynamic polarization results indicated that, konjac glucomanan behaves as a mixed-type inhibitor, moreover, electrochemical noise (EN) indicated that the growth and propagation stages of the pitting corrosion germinating on the metal surface are packed by polysaccharide additives.

The corrosion inhibition behavior of bee products for aluminium alloy in alkaline media was studied by J. Ryl (2019) [110] using potentiodynamic polarization and impedance techniques. Various bee products were found to be efficient corrosion inhibitors of aluminium in different environments, in particular, bee pollen was found to be highly effective in alkaline media. The highest inhibition efficiency exceeding 90% at 10 g/L was recorded for the water-ethanol extract.

Different plant parts extracts of *Tribulus terrestris* were employed against aluminium corrosion in basic media by P. Rathi *et al*. (2019) [111] using weight loss, thermometric method, potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The surface analysis was carried out by Fourier transform infrared spectroscopy and scanning electron microscopy. The results showed an inhibition efficiency greater than 70% with leaves extract in 0.5 M NaOH solution.

II.3. Plant extracts as inhibitors for corrosion of copper and its alloys

Copper is one of the most important metals in industry because of its high electrical and thermal conductivities, so it is used in many industrial applications such as conductors in electrical power lines, pipelines, heat conductors of heat exchangers, sea water desalination and shipbuilding [112–115]. Copper metal suffer different forms of corrosion depending on the environment, so one of the major challenges in copper applications is controlling the copper corrosion.

The efficiency of acid extract of *Azadirachta indica* seed as corrosion inhibitor for mild copper metal in 1, 2, 3 N HNO_3 solution was studied by T.V. Sangeetha *et al.* (2011) [116] using weight loss method. The results indicated that *Azadirachta indica* seed act as good corrosion inhibitor for copper in nitric acid having efficiency of 95% at 1% inhibitor concentration in different time duration.

The inhibitive effect of Mangure tannin as green inhibitor for the corrosion of copper in 0.5 M HCl solution was studied by A.M. Shah (2013) [117] using weight loss, potentiodynamic polarization, electrochemical impedance spectroscopy, scanning electron microscope, with energy dispersive x-ray (EDX), atomic absorption spectroscopy (AAS) and ion chromatography (IC). The highest inhibition efficiency achieved 82% at 3 g/L. The results indicated that Mangrove tannin act as cathodic inhibitor and its adsorption process obeys Langmuir isotherm.

Cannabis plant extract was tested as green corrosion inhibitor in 0.5 M H_2SO_4 solution by B.A. Abd-El-Nabey *et al.* (2013) [118] using weight loss, potentiodynamic polarization, electrochemical impedance spectroscopy and optical micrograph techniques. The results indicated that the dissolution process of copper is controlled by diffusion and cannabis extract act as cathodic inhibitor. Langmuir, Flory-Huggins and Kinetic-thermodynamic model were tested to clarify the nature of adsorption.

The inhibitive action of caffeine isolated from black tea on corrosion of copper in 0.5 M NaCl solution was studied by S. Gudić *et al*. (2014) [119] using potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The results showed that caffeine acted as an effective corrosion inhibitor for copper in NaCl solution with an inhibition efficiency up to 92%. Polarization results indicated that caffeine act as cathodic type inhibitor by adsorption on the copper surface according to Langmuir isotherm.

Inhibition of the Cu 65/Zn 35 brass corrosion in 0.1 N Na_2SO_4 solution with pH 7 and 4 by natural extract of *Camellia sinensis* was investigated by T. Ramde (2014) [120] using potentiodynamic polarization, electrochemical impedance spectroscopy and scanning electron microscope techniques. The results indicated that the extract is a very effective corrosion inhibitor for brass corrosion in both acidic and neutral media.

The corrosion inhibition effect of *Calligonum comosum* extract on copper in 2 M HCl solution has been investigated by M. Shabani-Nooshabadi *et al*. (2015) [121] using potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The polarization studies showed that this extract act as mixed-type inhibitor and a maximum inhibition efficiency of 80.06% was obtained in 0.8 g/L extract. The inhibitory action of the extract was discussed on the basis of Langmuir adsorption isotherm.

The inhibitive effect of alcoholic extract of *Mimusops elengi* leaves on copper corrosion in natural sea water was investigated by P. Deivanayagam (2015) [122] using weight loss measurements. The maximum inhibition efficiency was 86.84% after 120 hours immersion. The adsorption of inhibitor on the metal surface followed the chemical adsorption mechanism.

The inhibitive action of *Alhagi Maurorum* plant extract on the corrosion of copper in 0.5 M H_2SO_4 solution was studied by B.A. Abd-El-Nabey (2015) [123] using weight loss, potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The results indicated that this extract act as a good cathodic-type inhibitor and its adsorption on the copper surface obey the isotherms of Langmuir, Flory-Huggins and Kinetic-thermodynamic model.

Olive leaf extract as a natural corrosion inhibitor for pure copper in 0.5 M NaCl solution was studied by C. Rahal (2016) [124] using potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The highest inhibition efficiency was 90% after 24 hours immersion. The electrochemical results indicated that this extract acted as cathodic-type inhibitor and hindering the reduction of dissolved oxygen.

Myrrh extract as a corrosion inhibitor for a copper-zinc alloy in 3.5 Wt.% NaCl solution polluted by 16 ppm sulphide was studied by H.S. Gadow *et al*. (2017) [125] using weight loss method, electrochemical measurements, AFM, UV spectroscopy and FT-IR. The results indicted hat the inhibition efficiency of the extract increase with its concentration and reach 67% at 300 ppm and 25°C. Potentiodynamic results indicated that the extract act as mixed-type inhibitor and its adsorption at the alloy surface obey Langmuir isotherm.

Capparis spinosa L. extract was used as green inhibitor for copper corrosion in strong acid media by F. Wedian *et al.* (2017) [126] using weight loss and polarization measurements. The results indicated that this extract act as an efficient inhibitor for corrosion of copper in strong acid medium with maximum inhibition efficiency of 82.7% at 440 ppm of the extract. The weight loss, potentiodynamic and chemical calculations were in a good agreement.

Ziziphus lotus (wild jujube) were tested as corrosion inhibitor for copper in sea water by R. Oukhrib *et al.* (2017) [127] using weight loss and polarization methods. The morphology of the copper surface was analyzed after immersion in inhibited and uninhibited electrolytes using the scanning electron microscope. The results revealed that the extract act as efficient inhibitor for the copper corrosion in sea water, the highest efficiency is 93% at 5 g/L.

The inhibitive action of natural Propolis on bronze corrosion in a weakly acidic solution containing Na_2SO_4 and $NaHCO_3$ at pH 5 was studied by S. Varvara *et al*. (2017) [128] using potentiodynamic polarization, electrochemical impedance spectroscopy and scanning vibrating electrode technique measurements. The results showed that propolis act as effective inhibitor for corrosion of bronze with highest efficiency 98.9% at 100 ppm after 12 hours immersion. The adsorption of the extract on bronze was found to follow Langmuir adsorption isotherm.

Berry leaves extract was examined as a green inhibitor for corrosion of copper in nitric acid solution by A.S. Fouda *et al*. (2018) [129] using weight loss, potentiodynamic polarization, electrochemical impedance spectroscopy and electrochemical frequency modulation techniques. The results indicated that this extract act as effective inhibitor for copper corrosion in 2 M HNO_3 with inhibition efficiency of 90.1% at 300 ppm and 45°C. The adsorption of the extract on the copper surface was found to obey Temkin isotherm.

Aqueous and hydrolysis extracts of Olive leaf was studied as a green inhibitor for corrosion of copper in 0.5 M NaCl by P. Refat *et al.* (2020) [130] using electrochemical impedance spectroscopy and voltammetry techniques. The acid hydrolysis extract obtained at high temperature mainly contained hydroxyl tyrosol and clenolic acid and gave the highest inhibition efficiency of 95%.

II.4. Plant extracts as inhibitors for corrosion of zinc and its alloys

Zinc is one of the most important nonferrous metals with numerous industrial applications and finds extensive use in metallic coating. Zinc corrodes in a solution with a pH lower than 6 and higher than 12.5, but within the range, the corrosion is very slow [131]. In aggressive environments, zinc metal undergoes corrosion, giving white colored rust [132, 133]. The formation of the white rust on zinc surface is prevented by the application of chromate treatment [134]. Recent environmental regulation restrict the use of chromate solution and recommended its replacement with ecofriendly non-toxic and non-polluting substances.

The inhibitive action of *Aloe vera* leaves extract on the corrosion of zinc in HCl solution was studied by O.K. Abiola *et al*. (2010) [135] using weight loss technique. The extract inhibited the corrosion of zinc in 2 M HCl. The adsorption of the inhibitor molecules on the zinc surface obeyed Langmuir isotherm. A first order kinetics relationship with respect to zinc was obtained with and without the extract.

The inhibitive action of Lubine, Hafabar and Damssisa on the corrosion of zinc in 0.5 M NaCl solution were studied by B.A. Abd-El-Naby *et al*. (2012) [136] using potentiodynamic polarization and electrochemical impedance spectroscopy techniques. Potentiodynamic polarization results indicated that the three extracts act as mixed-type inhibitors and give good inhibition efficiency. Theoretical fitting of the isotherms of Langmuir, Flory-Huggins and kinetic thermodynamic model were tested to clarify the nature of adsorption.

Ethanolic extract of *Mansoa alliacea* was investigated as green inhibitor for corrosion of zinc in 3% NaCl solution by F. Suedile *et al*. (2014) [137] using potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The results indicated that the extract could serve as an effective inhibitor for the corrosion of zinc in 3% NaCl solution. The extract gave 90% inhibition efficiency. Potentiodynamic polarization results indicated that the extract acts as mixed type inhibitor.

The inhibitive action of *Allium cepa* extract on the corrosion of zinc and carbon steel in 2 M H_2SO_4 was studied by A.J. Chinweuba (2014) [138] using weight loss method. The results indicated that this extract act as good inhibitor for corrosion of carbon steel and zinc in 2 M H_2SO_4 solution. The inhibition efficiency increases up to 97.7% for mild steel and 89.1% for zinc in 2 M H_2SO_4.

The inhibitive action and synergistic properties of extract of *Moringa oleifera* leaves on the corrosion of zinc in HCl solution was investigated by F.A. Ugebe *et al*. (2015) [139]. They studied the corrosion behavior of zinc immersed in 0.5 M, 1.0 M, 1.5 M and 2.0 M HCl solutions each containing varied concentrations of the extract (0.1, 0.3 and 0.5 g/L) and halides (0.1, 0.3 and 0.5 M in each of KCl and KI) for synergism using thermometric method. The data revealed that this extract is an efficient inhibitor for corrosion of zinc in acid solution due to its phytochemical, saponins, tannins, flavonoids, glycosides, carbohydrates, reducing sugars, terpenoids, steroids and alkaloids.

The alkaloid extract of *Calendula officinalis* (Pot Marigold) leaves was examined as corrosion inhibitor for corrosion of carbon steel, aluminium and zinc metals in 5 M HNO_3 solution by B.U. Ugi *et al*. (2016) [140] using weight loss, potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The weight loss results showed that the plant extract is an excellent corrosion inhibitor to all studied metals, the polarization results indicated that it acts as a mixed-type inhibitor.

Fenugreek seeds extract was investigated as a green corrosion inhibitor for zinc in 2 M H_2SO_4 and 2 M HCl solutions by H.A. Al Lehaibi (2016) [141] using weight loss, electrochemical techniques, scanning electron microscope and x-ray photoelectron spectroscopy (XPS) analysis. The maximum inhibition efficiency values are 90.7% after 1 hour and 66.6% after 0.5 hour by 200 ppm of the extract in H_2SO_4 and HCl solutions, respectively.

The inhibitive action of the ethanolic extract of oil from *Picralima nitida* leaves for corrosion of zinc in 1.0 M HCl solution was studied by J.N.O. Ezeugo *et al*. (2017) [142] using weight loss and thermometry method. Results indicated that the inhibition efficiency of the extract reached 86.78% at 1.2g/L after 8 hours. The inhibitive action of the extract was discussed in view of Langmuir adsorption isotherm.

Solanum nigrum extract was investigated by A.S. Fouda *et al*. (2017) [143] as green inhibitor for corrosion of zinc in 3.5% NaCl and 16 ppm Na_2S solution using weight loss, Tafel polarization and electrochemical impedance spectroscopy techniques. The results illustrated that this extract act as effective inhibitor for corrosion of zinc in 3.5% NaCl and 16 ppm Na_2S solution. The inhibition efficiency reached 81.5% at 500 ppm after 180 minutes immersion. The adsorption of the extract molecules on the zinc metal surface obeys Freundlich and Langmuir adsorption isotherm.

The inhibitive action of Fucoidan as green inhibitor for corrosion of zinc in sea water was investigated by C. Wang (2017) [144] using potentiodynamic polarization and electrochemical impedance spectroscopy techniques. Potentiodynamic polarization results showed that this extract acts as anodic type inhibitor. The corrosion inhibition of Fucoidan was confirmed by the scanning electron microscope and atomic force microscope. Langmuir adsorption isotherm was found the appropriate adsorption model.

Corrosion inhibition of zinc in 0.5 M HCl solution by *Ailanthus altissima* extract has been studied by A.S. Fouda (2018) [145] using weight loss, hydrogen evolution, potentiodynamic polarization, electrochemical impedance spectroscopy and electrochemical frequency modulation techniques. Results indicated that this extract exhibited high inhibition efficiency for corrosion of zinc in HCl solution and its inhibitive effect was discussed on the basis of the adsorption of its components on the zinc surface following Temkin isotherm.

Picralima nitida seed extract was investigated as a green inhibitor for corrosion of zinc in 0.5 M H_2SO_4 by J.N.O. Ezinga *et al*. (2018) [146] using weight loss method. The results indicated that this extract acted as an efficient inhibitor for corrosion of zinc in sulphuric acid solution. The protective action of the extract was discussed in view of Langmuir adsorption isotherm.

II.5. Plant extracts as inhibitors for corrosion of nickel and its alloys

Nickel and its alloys are used extensively in industry, air craft, marine, chemical, petrochemical, oil and gas, nuclear and conventional power generating, textile, desalination and food processing industries. Because their resistance to aqueous corrosion and to oxidative corrosion at elevated temperature. Nickel is an important constituent of super alloys, stainless steel and other alloys as well as providing protective coating for less corrosion resistance materials, these has been an ever increasing demand for nickel in the last four decades resulting in higher rates of production [147].

Nickel has good resistance to many corrosive solutions like slightly acidic and alkaline solutions so that it forms a good base for alloys requiring strength at high temperature. Nickel occupied an intermediate position in the electrochemical series $E_0Ni^{2+}/Ni = –0.25$ V, it is more noble than Sn, Pb and Cu [148].

Many parts of new machines are made of pure nickel [149]. Nickel electrodes are important materials for petrochemical technology and energy conversion devices. In aqueous environments nickel electrodes are mainly covered by electrochemically active $Ni(OH)_2$ layers, although in acid solution nickel corrosion and passivation takes place [150].

The aqueous extract of the leaves henna (*Lawsonia*) was tested as corrosion inhibitor for C-steel, nickel, and zinc in acid, neutral and alkaline solution by A.Y. El-Etre (2005) [151] using the polarization technique. It has been found that the extract acted as a good inhibitor for the corrosion of the three metals in all tested media. The extract acted as mixed-type inhibitor and its inhibitive effect was discussed on the basis of the adsorption of its ingredients molecules on the surface of the metal.

The inhibitive action of the extract of *Ficus nitida* leaves for general and pitting corrosion of C-steel, nickel and zinc in different aqueous media was studied by A.Y. El-Etre (2006) [152] using weight loss, potentiostatic and potentiodynamic polarization techniques. The results indicated that this extract inhibit the corrosion of the three metals in acidic, neutral and alkaline media. The inhibitive action of the extract was discussed on the basis of the adsorption of its ingredient molecules on the surface of the metal. The adsorption process obeyed the Langmuir adsorption isotherm.

Inhibition of the corrosion of nickel, Inconel 600 and Inconel 690 in HCl solution by natural Clove oil was studied by M. Abdallah *et al*. (2009) [153] using potentiodynamic polarization measurements. The inhibitive action of the oil was attributed to its adsorption onto the metal surface. The experimental results of inhibition obeys Langmuir isotherm.

The inhibitive effect of natural black cumin oil on the corrosion of nickel in 0.1 M HCl was studied by M. Abdallah *et al*. (2010) [154] using galvanostatic and potentiodynamic polarization techniques. The extract act as effective inhibitor for corrosion of nickel in the acid solution and its inhibition effect was discussed on the basis of the adsorption of its constituents on the surface of the metal. The adsorption process was controlled by Langmuir adsorption isotherm.

Cannabis plant extract as inhibitor for the corrosion of nickel in 0.5 M H_2SO_4 was studied by B.A. Abd-El-Naby *et al*. (2012) [155] using potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The results indicated that this extract act as mixed-type inhibitor and serve as effective inhibitor for corrosion of nickel in sulphuric acid solution. Theoretical fitting of different adsorption isotherms, Langmuir, Flory-Huggins and the Kinetic-thermodynamic model were tested to clarify the nature of adsorption.

The inhibitive effect of some natural oil *e.g.* sesame oil, water cress oil, wheat germ oil and almond oil on the corrosion of nickel in NaOH solution was studied by M. Abdallah (2014) [156] using open circuit measurements, galvanostatic and potentiostatic polarization techniques. The inhibitive effect of these oils was discussed on the basis of adsorption of these constituents on the surface of the metal. The adsorption process followed Freundlich isotherm.

III. Plant extracts as scale inhibitors

Scaling is the deposition of a mineral salt on processing equipment. It is a result of super saturation of mineral ions in the process fluid. The mineral salt can be either Calcium Carbonate or Barium Sulphate and so on [157].

Mineral scale deposits in industrial waters that support a number of processes represent a major problem leading to unexpected shutdowns and costly chemical or mechanical cleaning actions. There are many strategies to control scale formation including acidification, ion exchange softening, and scale inhibitors addition [158].

The environment concerns lead the research to develop new environmental friendly chemicals, currently named "Green" as scale inhibitors. The challenge is to determine the mechanisms of action of these chemicals against scale deposition to optimise their doses. There is already a need to develop a range of innovative antiscalants from materials of bio-origin because these have in-built multifunctional ion complexing and solubilizing functional groups such as carboxylic, hydroxyl, amine, or others [159]. Plant extracts have been recently used as new green antiscalants. Indeed, as they can be easily extracted and are environmentally friendly, they represent an interesting alternative source of "natural" organic molecules.

III.1. Plant extracts for inhibition of Carbonate crystallization

Miksic *et al.* (2005) [160] investigated the scale inhibition effect of soy bean extract and polysaccharides extracted from sea weeds using test methods described in a NACE Standard [161]. The authors reported that these extracts were more effective than polyaspartic acid in preventing calcium carbonate formation. Indeed, the percentage of inhibition was 16.7% for both soy based polymer and polysaccharides from sea weeds, whereas it was only 6.6% for polyaspartic acid.

Abdel-Gaber *et al.* (2008) [162] reported inhibition of $CaCO_3$ scale performed with fig leaf extract. Stock solution of inhibitor was obtained from grinded dried leaves of fig tree by extraction in boiling distilled water. The $CaCO_3$ deposition from an alkaline brine solution which models natural seawater was studied by chronoamperometry at 40°C on a steel electrode. Electrochemical Impedance Spectroscopy (EIS) measurements were also used to observe the nucleation, growth and total coverage at the surface electrode. Measurements were also used to observe the nucleation, growth and total coverage on the steel electrode surface. According to impedance data, the concentration of the solution necessary to obtain an inhibition efficiency of 85% was 75 mg/L. In addition, optical microscopic examination indicated that, even at low concentration (5 mg/L), fig leaf extract could prevent total coverage of the steel electrode surface. The authors suggested that fig leaf extract may complex the cations present in the brine solution, or to disperse the suspended solids through adsorption. Also, the authors carried out the same study concerning the olive leaf extract

(2001) [163]. Indeed, olive leaves contain many phenolic molecules including oleuropein, the most abundant biophenols in olive leaves [164] and caffeic acid [165, 166]. Stock solution of inhibitor was obtained from grinded dried leaves of olive tree by extraction in boiling distilled water. According to micrographic photos, the authors concluded that olive leaf extract acts as antiscalant by inhibiting nucleation step. These phenolic molecules may complex Ca^{2+} *via* their carboxyl and hydroxyl groups. A concentration of 50 mg/L of the olive leaf extract was found to be most effective for inhibition of calcium carbonate scaling. Even at low concentration (5 mg/L), addition of olive leaf extract is able to prevent total coverage stage of the steel electrode surface.

Moreover, Abdel-Gaber *et al.* (2012) [167] studied the antiscaling properties of *Punica granatum* hull and leaf extract in alkaline brine at 25°C using conductivity measurements, electrochemical impedance spectroscopy and chronoamperometry in conjunction with SEM, EDX and optical microscopic examinations. This work showed that the hull extract exhibited better antiscaling properties than leaf extract. Two major constituents of hull, *i.e.* the polyphenols punicalin and punicalagin, might be involved in the inhibition process. Microscopic examination of the filmformed over the steel electrode surface indicated that this process might take place via surface modification.

Castillo *et al.* (2009) [168] reported inhibition results of calcium carbonate scale performed with *Aloe vera* in Venezuelan oilfields. The scale inhibitor was obtained by dissolving *Aloe vera* gel in water at concentration in the range 5%–50% wt/wt. This solution contains polysaccharides [169] that can complex with Ca^{2+} ions. Some field tests were carried out on Venezuelan oil wells with water containing high bicarbonate ions (total calcium concentration of 535.4 mg/L). Information on the inhibitory performance of *Aloe vera* are unfortunately limited. Weekly inspections of coupons performed during field tests with inhibitors (20 or 30 days of duration) led the authors to define the recommended concentration for the inhibitor. The *Aloe vera* solution was reported to provide very effective scale inhibition with a concentration of 15.2 mg/L (Barinas field test). Pressure and temperature were also recorded through the entire field test, and remained almost constant during the tests. This indicated the absence of precipitated solids in the system in the presence of *Aloe vera*.

Suharso *et al.* (2011) [170] reported Gambier extract from *Uncaria gambier* Roxb leaves as green inhibitor of $CaCO_3$ scale formation. The addition of the Gambier extract (*Uncaria gambier* Roxb leaves) modification (Gambier:benzoic acid:citric acid / 2:1:2) as a green inhibitor on the formation of $CaCO_3$ scale at various concentrations was carried out using a seeded experiment method. The experiments were performed with observing the precipitation change of the $CaCO_3$ crystals growth obtained. In order to prove the efficiency of the inhibitor in inhibiting the formation of the $CaCO_3$ crystals, the changes of the crystal morphology were investigated by scanning electron microscopy (SEM) and the changes of the crystal size distribution were analyzed by particle size analyzer (PSA).The research results showed that the Gambier extract modification was able to inhibit the formation of the $CaCO_3$ scale indicated with the morphology change of the $CaCO_3$ crystals and smaller crystal size distribution after the addition of this inhibitor. The modification of the Gambier extracts with the addition of benzoic and citric acid with the composition ratio of Gambier extract: benzoic acid:citric Acid / 2:1:2 in 1 L of water solution as green inhibitor increased the quality of this mixture. This mixture was able to inhibit the formation of $CaCO_3$ scale at various inhibitor concentrations of 50–300 ppm and growth solution concentrations of 0.1–0.6 M using seeded experiment method with the inhibitor efficiency (%*IE*) of 12–92%. The inhibitor used also changed the morphology of $CaCO_3$ crystals and it made the crystal size of $CaCO_3$ crystals to be smaller in their crystal size distribution after addition of this inhibitor.

Belarbi *et al*. (2014) [171, 172] carried out chronoamperometry experiments to test the scale inhibition performance of aqueous extract of *Paranichia argentea*. Stock solution of inhibitor (20% w/v) was obtained from grinded dried leaves and flowers of *P. argentea* by infusion in boiling distilled water. *P. argentea* acts as good antiscalant of $CaCO_3$ formation on a copper surface. For inhibition of calcium carbonate scale, a concentration of 70 mg/L of the *P. argentea* extract was found to be the most effective concentration in carbonically pure water (concentration of Ca^{2+} 120 mg/L). The authors indicated that the efficiency of this inhibitor was decreased at 60°C, which could limit the interest for some industrial processes. Finally, SEM micrographs indicated, in the presence of low concentrations of *P. argentea*, highly deformed crystals of calcite and vaterite. This could be due to a partial covering of the crystal surface by the inhibitor. Different fractions were separated by HPLC from the stock solution; some of them were more efficient towards scale inhibition than the whole solution. Unfortunately, information about this purification process is limited [173]. Moreover, different commercially available biocides named B310, B320, B330 and B340 were also tested. The biocide B340 was the only found not compatible with green inhibitor.

Abd-El-Khalek, *et al*. (2016) [174] studied the antiscale properties of palm leaves extract (*Phoenix dactylifera L*) using electrochemical impedance spectroscopy (EIS) and chronoamperometry techniques, in addition to microscopic examination. Calcium carbonate scales were deposited from the brine solution by cathodic polarization of the steel surface at −0.9 V (*vs*. SCE). Chronoamperometry curves and EIS measurements displayed that palm leaves extract increased the time of $CaCO_3$ nuclei formation and consequently, retarded the growth step. The optical micrographs of the steel electrode clarified that the surface area occupied by the scale particles decreased with increasing plant extract concentrations. The results showed that palm leaves extract could be considered as efficient inhibitor for $CaCO_3$ precipitation.

The anti-scale property of tobacco rob extract (TRE) in artificial seawater was studied by Wang *et al*. (2016) [175] using static tests for scale and the scale deposits such as X-ray diffraction (XRD) and SEM, respectively. The results showed that the scale deposits surface morphology and size were changed in the presence of TRE. The corrosion and scale inhibition results indicated the potential use of TRE as an efficient corrosion and scale inhibitor in artificial seawater.

Bendaoud-Boulahlib *et al*. (2017) [176] studied the effect of orange peel on the $CaCO_3$ precipitation in Algerian ground water of Bounouara having a hardness of 58°F. The main objective was to reduce the scaling power and then to prevent the fouling phenomenon met in the equipments supplied by this water. Chronoamperometry tests showed that Bounouara water is extremely scale-forming water by scaling time of ($tE = 9.17$ min) and an index scaling ($IE = 107$ min^{-1}). The accelerated scaling curves registered at different temperatures of raw water of Bounouara showed that scaling time decreases by increasing the temperature, and Bounouara water become more scale-forming in high temperatures. The anti-scale treatment with orange peel is more efficient in low temperature, because the total inhibition of hardness of Bounouara water was realized at 20°C with an addition of 0.75 g/L of orange peel but at 40°C, 1 g/L of orange peel must be added to Bounouara water to totally block the precipitation of tartar. So orange peel is an effective inhibitor for the scaling treatment of drinking water and for the valorization of orange peel and protection of the environment from food industry waste that reject 60% of citrus fruits in the form of waste.

The addition of the Gambier extract (*Uncaria gambier* Roxb leaves) modification (Gambier:benzoic acid:citric acid = 2:1:2) as a green inhibitor on the formation of $CaCO_3$ scale at various concentrations was carried out by Suharso *et al.* (2017) [177] using a seeded experiment method. The experiments were performed with observing the precipitation change of the $CaCO_3$ crystals growth obtained. In order to prove the efficiency of the inhibitor in inhibiting the formation of the $CaCO_3$ crystals, the changes of the crystal morphology were investigated by scanning electron microscopy (SEM) and the changes of the crystal size distribution were analyzed by particle size analyzer (PSA). The research results showed that the Gambier extract modification was able to inhibit the formation of the $CaCO_3$ scale. indicated with the morphology change of the $CaCO_3$ crystals and smaller crystal size distribution after the addition of this inhibitor.

Mohammadi *et al*. (2018) [178] examined *Bistorta officinalis* extract as an effective green inhibitor for prevention of scale formation problems in cooling water system. Electrochemical impedance spectroscopy, DC polarization, weight loss measurement and UV-Vis spectroscopy have been conducted to investigate the mechanism and performance of the proposed inhibitor. The *Bistorta officinalis* extract exhibited an excellent scale inhibition performance of 99.5% which make it an ideal material to prevent the scale formation in cooling water. Accordingly, the *Bistorta officinalis* extract could be proposed for effective treatment of cooling water systems.

The aqueous extract of *Gypsophila aretioides* roots as a "green" or eco-friendly inhibitor of calcium carbonate formation was investigated by Hajizadeh *et al.* (2019) [179]. The presence of chelating factors such as polyphenol compounds and foaming agents in the plant extract was assessed. Atomic absorption spectroscopy (AAS) demonstrated the ability of *G. aretioides* extract for dissolution of the calcium carbonate precipitates. Moreover, conductivity measurements revealed that the *G. aretioides* extract could efficiently retard sedimentation of calcium carbonate from a brine solution of calcium ions exposed to carbonate ions by adding Na_2CO_3 to the system solution. In addition, chronoamperometry was performed for a period of 3 h by polarizing the steel electrode to –0.9 V (*vs*. SCE) at 40°C. The presence of *G. aretioides* extract at a concentration of 10% (w/v) in chronoamperometry prevented the precipitation of $CaCO_3$ on the steel electrode surface. This was confirmed by scanning electron microscopy (SEM) and energy dispersive X-ray (EDX) analyses which showed an absence of precipitates and lack of calcium ions on the steel electrode after 3 h chronoamperometry, respectively.

Vasyliev *et al.* (2020) [180] prepared *Raphanus sativus* L. ethanol extract by radish cake maceration in ethanol and tested this extract as a scale and corrosion inhibitor of mild steel in tap water. Antiscalant efficiency was tested with electrochemical and thermal scaling techniques, and changes in hardness content were determined titrimetrically. No deposits were found on the metal surface at the extract concentration of 10 mL/L in chronoamperometry test, and scaling suppression was established 5 times in thermal scaling conditions. Corrosion Inhibition efficiency was found to be 75% in thermal scaling conditions. Formation of the surface film was responsible for both scaling and corrosion suppression on mild steel surface as was established with FT-IR spectroscopy and SEM. Surface film was found to contain polymerization products of isothiocyanates.

III.2. Plant extract for inhibition of sulfate crystallization

Abd-El-Khalek *et al.* (2019) [181] studied the antiscale properties of sunflower (*Helianthus annuus*) seed extract for $CaSO_4$ and $BaSO_4$ scales using NACE and conductivity tests, respectively. Comparative studies between the extract and 1-hydroxyethane-1,1-diphosphonic acid (HEDP), as commercial antiscalant, were done. The results revealed that the inhibition of $CaSO_4$ scales using sunflower seed extract reached 100%, while HEDP achieved a maximum inhibition of 88%. Moreover, the maximum inhibition of $BaSO_4$ scale in the presence of the extract was 84% compared with 86% in the presence of HEDP. Also microscopic examination showed that both inhibitors modified $CaSO_4$ and $BaSO_4$ crystals.

The influence of *Sargassum sp*., *Corallina mediterranea*, and *Avicennia marina* on RO membrane mineral scaling was evaluated by Hamdona *et al.* (2019) [182] using gypsum as a model scalant. The degree of inhibition of gypsum scale in the presence of these inhibitors are in the following order: *Avicennia marina* (M) > *C. Mediterranean* > *Sargassum sp.* extract at different dosage. data confirmed that the anti-scalant properties by 100 ppm of *Avicennia marina* leave extract giving 85% of scale inhibition

Suharso *et al.* (2019) [183] studied the effect of the addition of gambier extract modified with kemenyan extract on the growth of calcium sulfate ($CaSO_4$) scale formation as a green inhibitor. The crystallization experiments were carried out by using unseeded experiment method at temperature of 90°C. The $CaSO_4$ crystals obtained with and without the addition of inhibitor were analyzed by scanning electron microscopy (SEM), particle size analyzer (PSA), and X-ray diffraction (XRD). The results of this experiment show that the addition of the combination of gambier and kemenyan extract with ratio of 5:9 can inhibit the growth of $CaSO_4$ crystals with the inhibition effectivity of 39.88%. These results were supported from the SEM and PSA data showing that the crystal size and particle size distribution of the $CaSO_4$ in the addition of the inhibitor are smaller than without the addition of inhibitor. In addition, analysis using XRD showed that $CaSO_4$ crystals undergo a change in crystalline phase with the addition of inhibitors.

The addition of *Piper betle* leaf extract with the concentration of 450 ppm was tested by Santoso *et al.* (2019) [184] as a green inhibitor for $CaSO_4$ scale formation at the concentration of growth solution of 0.05 M and temperature of 90°C using a seeded experiment method. The experiments were performed with observing the precipitation change of $CaSO_4$ crystals growth obtained. In order to prove the efficiency of the inhibitor in inhibiting the formation of $CaSO_4$ crystals, the changes of the crystal morphology were investigated by scanning electron microscopy (SEM). The research results showed that *Piper betle* leaf extract was able to inhibit the formation of $CaSO_4$ scale which was indicated with the morphology change of the $CaSO_4$ crystals after the addition of this inhibitor. The ability of *Piper betle* leaf extract as an inhibitor of the formation of $CaSO_4$ is 47.07%.

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