Green Plants Extracts Corrosion Inhibition of Aluminum – A Review

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Abstract- The importance of the use of green inhibitors on aluminum in both acidic and alkaline media cannot be over - emphasize due to the growing cost of corrosion mitigation processes. This review article examines various green plants and their effectiveness on aluminum and its alloys. The methods used to study their effectiveness were also examined. Room and elevated temperatures situations were examined. Density functional theory (DFT)/B3LYP and Monte Carlo computational approaches were used for the interpretation of inhibition process under electronic and atomic scales. The review concludes that the use of green plant extracts as an inhibitors on aluminum will continue to grow as most of the plant extracts proves to be effective particularly in the packaging industry.

Indexed Terms- Aluminum, Corrosion Inhibition, Green Plants, Plants Extracts, Versatile Metal

I. INTRODUCTION

Aluminum is a versatile metal due to its light weight, ease in manufacture, seeming corrosion resistance at mild situations, availability of its ore etc (Nanan, 2020). It is used in the transportation, building, electrical, packaging, machinery, consumer and others industries (Woodford, 2021). In the consumer and packaging industries where aluminum and its alloys are used especially in pipes, reservoirs, packaging of foods, etc. The salt used in seasoning foods, salt water in ballast tank; acids used in pickling, flushing of pipes etc., are causing corrosion (wikipedia, 2022).

Corrosion usually occurs in pipes, reservoirs, food cans in the food industry (Kingsley, *et al.*, 2022) and transportation industry (Buzalsky, nd). It is the loss of chemical and physical qualities of metallic materials due to environmental attack on the surfaces of metals. These attacks cause destruction of metal leading to catastrophic failures. The effects are complete shutdown of plants, product contamination, lost of product when it is a storage facility, plant efficiency reduction and sometimes loss of lives (Harsimran, et al., 2021). The economic impact is approximately 5% of gross domestic products (GDPs) of economies especially in the middle east areas (Mazumder, 2020; Harsimran, et al., 2021). General attack, localized corrosion, galvanic, environmental cracking, flow assisted, inter-granular, de-alloying, fretting, high temperature etc., are types of corrosion (Bell, 2019) the most common being general attacks. These situations need to be avoided or mitigated so that process industries can operate seamlessly without sudden short downs.

Farh, et al. (2023) classified corrosion mitigation processes into passive, active and hybrid methods. The passive methods according to them include electrical isolation, material design, barriers, linings, coatings, inhibitors, and multi-passive techniques. In their statement impressed current and sacrificial anode cathodic protection systems are active ones. They continued by saying that better comprehensive protective system can be achieved by combining passive and active techniques.

Amongst the techniques mentioned, inhibitors are chemicals either organic or inorganic added to corrosive environment in small dosages to alter the corrosive behavior positively without altering the chemistry of the environment (Desai, et al., 2022). There are inorganic, organic and mixed type inhibitors. Inorganic inhibitors are further divided into anodic, cathodic and mixed type inhibitors (Ma, et al., 2021). Organic inhibitors are natural substances either synthesize or in their natural state used to mitigate corrosion by chemisorptions or physisorption forming a film that causes barrier thereby protecting the system. Oil and gas; petrochemical; marine and food industries widely uses organic inhibitors (Al-Amiery, et al., 2023). They are ionic molecules and green inhibitors. Ionic molecules are synthetic liquids that contain organic compounds with double and triple bonds that are used for corrosion inhibition. They are ions whose melting point below 100^{o}C (Verma, et al., 2017).

Green inhibitors (plant extracts) are also used to inhibit corrosion in acid and alkaline media. They have effectively performed the inhibition function (Fazal, et al., 2022; Hossain, et al., 2023 etc. Our concern in this article is to review the use of green inhibitors (plant extracts) on aluminum and their alloys in corrosive media.

II. GREEN LEAVES USED IN CORROSION INHIBITION ON ALUMINUM

The presence of chlorophyll which is a colour pigment contained in chloroplast in plants made leaves to be green. Blue and red wavelengths are usually absorbed and reflect green light. Green leaves are medicinal (Meeks & Tee-melegrito, 2021). These medicinal constituents in plants are called phytochemicals. It is proven that phytochemicals inhibit corrosion and environmentally friendly and therefore widely used (Donkor, et al., 2022). The following are recent studies on the effectiveness of phytochemicals on aluminum alloys corrosion inhibition.

Abakedi (2017) studied aluminum (Al) corrosion inhibition by microdesmis puberula leaf extract (MPLE) in 2 M hydrochloric (HCl) acid solution using thermometric and hydrogen evolution methods. Increase in MPLE concentration increases corrosion inhibition efficiency of Al in 2M HCl media. Increase in temperature in corrosion led to decrease in inhibition efficiency of MPLE even with increasing concentration. Thermodynamic parameters calculated from the data obtained from hydrogen evolution experiment revealed spontaneous reaction of MPLE with Al surface especially from the change in the standard Gibbs free energy of adsorption ($\Delta G°_{ads}$) values. The adsorption curves obtained obeyed the Langmuir adsorption isotherm. Physisorption was suspected from the various calculations.

Obruche, et al., (2019) studied the corrosion inhibition effects of tagetes erecta l. leaf extract on aluminum in acidic (0.5 M solution of HCl) medium using gravimetric method. The results revealed that increase in the concentration of tagetes erecta l. leaf extract led to the reduction of corrosion rate of aluminum in 0.5M HCl solution. Maximum inhibition efficiency of 72.2% was obtained at the concentration of 4.8ml.

Chadili, et al., (2021) studied the corrosion inhibition of 3003 aluminum alloy in molar hydrochloric acid solution by olive oil mill liquid (OML) by-product using gravimetrical, thermometric and electrochemical techniques. DFT/B3LYP and Monte Carlo computational approaches were used for the interpretation of inhibition process under electronic and atomic scales, respectively. The results of the experiments show that inhibition efficiency increased up to 89% at 6.0 ppm concentration. At a rising temperature, efficiency reduced. The tafel plots revealed that the inhibitor is a mixed type and Langmuir adsorption isotherm was obeyed. The computational analysis confirmed OML components effectively interacted with the metal surface, hence the formation of a protective layer, which justified the observed inhibition behaviors. Conferring to the present study, OML can be used as a good green corrosion inhibitor for AA3003 alloy in the acidic medium.

Fouda, et al., (2021) studied the *conocarpus erectus* extract (CEE) as an eco-friendly corrosion inhibitor on aluminum in hydrochloric acid (1M HCl) solution using gravimetric and thermometric methods and also morphological study was carried out.

Conocarpus Erectus extract was examined as a green corrosion inhibitor for aluminum in 1M HCl and evaluated by different methods (chemical &

electrochemical methods). The study revealed that corrosion rate of aluminum reduction was observed as CEE concentration was increased but increased when temperature was increased. Maximum efficiency at 91.1% was attained at 300ppm of CEE. Anodic and cathodic (mixed) type of inhibition was observed from the thermodynamic parameters calculated from the thermometric studies. Adsorption curves obeyed Temkin isotherm. The morphological studies revealed that effective adsorption occurred on the aluminum surface from the 1M HCl medium by the CEE which confirmed its' effectiveness in inhibiting aluminum corrosion by 1M HCl.

Onen and Buba (2018) studied ficus polita (bush fig) leaves extract as corrosion inhibitor of aluminum in alkaline medium. Gravimetric method and electrochemical measurements were used at 303,313 and 323K temperatures. Corrosion parameters were calculated from the results of the gravimetric method electrochemical measurements. Increase in inhibitors concentration increased inhibition efficiency (%IE). Increase in the environmental temperature increased the rate of corrosion and efficiency was reduced. The increase in activation energy E_a values indicated physisorption process. Increase in efficiency and surface coverage (θ) values confirmed that the inhibitor is effective in inhibiting corrosion in sodium hydroxide (NaOH) medium. Since Langmuir isotherm was obeyed by adsorption curves (θ) the inhibitor acted as mixed – type inhibitor. The presence of – C =O and –N=N-, etc groups in extracts contributed greatly to the inhibition process by interacting with aluminum surface.

Abakedi and Moses (2016) studied the aluminum corrosion inhibition by *maesobatrya barteri* root extract in hydrochloric acid solution. Weight loss and at different temperatures method were used. Corrosion rate of aluminum was reduced as inhibitor concentrations increases. Physisorption was observed from the activation energy calculations as Langmuir adsorption isotherm was obeyed. Spontaneous and endothermic reactions were observed from the thermodynamic parameters calculation in the corrosion inhibition process.

Raghavendra and Bhat (2017) studied red arecanut seed extract as a sustainable corrosion inhibitor for aluminum submerged in acidic corrodent: an experimental approach towards zero environmental impact. Gravimetric and electrochemical methods and morphological studies were carried out on aluminum in 0.5M hydrochloric acid environment. The gravimetric studies revealed that the inhibition efficiency is indirectly proportional to solution temperature and directly proportional to inhibition concentration and contact time. Mixed type corrosion inhibition was observed from the Tafel plots and Langmuir adsorption isotherm was obeyed showing physisorption of red arecanut seed extract constituents on aluminum surface in 0.5 M HCl system. Morphological studies show adsorption of plant constituents on the surface of the aluminum metal.

Kumar and Mathur (2013) studied the corrosion inhibition and adsorption properties of ethanolic extract of *calotropis* (leaves, latex and fruit)for corrosion of aluminum in acidic media gravimetric and thermometric methods. The efficiency of corrosion inhibition increased and rate of corrosion reduced as inhibitor concentration increased. This was due to the adsorption of the plant parts on the surface of aluminum.

Ayuba and Abdullateef (2020) studied corrosion inhibition potentials of *strichnos spinosa* leave on aluminum in 0.9M HCl medium: experimental and theoretical investigations using gravimetric, Fourier transform infrared spectroscopy (FTIR), quantum chemical parameters and molecular dynamic stimulations of some compounds isolated from the plant in literatures. As the inhibitor concentration increase in this order: 0.2g/l < 0.4g/l < 0.6g/l respectively, corrosion rate reduced and efficiency increased (84.7%) but when the temperature increased from 303-323K, corrosion rate increase and efficiency reduced. Fourier transform infra-red (FTIR) examination revealed functional groups that participated in the inhibition process. Physisorption was apparent as revealed by activation calculations and adsorption curves fitted into Freundlich isotherm. Ursolic acid>Betulinic acid>Erythrodiol adsorption/binding strength order was observed from the quantum chemical parameters and molecular dynamic stimulations results.

Obot and Obi-Egbedi (2009) studied the effectiveness of ginseng root as corrosion inhibitor for aluminum alloy of type AA 1060 in hydrochloric acid solution using gravimetric method at 30-60^oC. Ginseng root extract showed to be effective and excellent inhibitor in the acid medium. Increase in temperature increase the corrosion rate both in the absence and presence of inhibitor. Efficiency of 93.1% was observed when the inhibitor concentration increase and corrosion rate decrease in the acid solution. Addition of iodide ions to the root extracts of ginseng enhances the inhibition efficiency considerably and the effect is more pronounced at higher temperatures. Spontaneous process was observed in the reaction and it followed the Freundlich adsorption isotherm. The thermodynamic parameters revealed physical adsorption of the root components on the surface of the metal.

Bhaskara et al., (2021) studied the evaluation of corrosion inhibition efficiency of aluminum alloy 2024 by diaminostilbene and azobenzene schiff bases in 1M hydrochloric acid. Synthesis of Schiff base compounds: N,N′-bis(salicylidine)-4,4′–diaminostilbene(SDS) and N,N′-bis(salicylidine)-4,4′-diamino azobenzene (SDA) were carried out and FT-IR and 1H NMR were carried out to determine their molecular structures. Gravimetric method, potentiodynamic polarization, impedance and morphological studies were done. Cathodic inhibition process was in the potentiodynamic polarization (PDP) examination of SDS and SDA compounds. Physisorption was revealed by the electrochemical impedance spectroscopic (EIS) study. Efficiency was observed to be increased as corrosion inhibitor concentration increased and decreases as environment temperature increased. Langmuir adsorption isotherm model was obeyed by the inhibitor. The morphological study revealed that the deterioration of the alloy surface is minimal in the presence of an inhibitor. Both Schiff base molecules exhibited superior corrosion inhibition for aluminum alloy 2024 alloy in HCl medium.

Chaudhary and Tak (2022) studied the natural corrosion inhibition and adsorption characteristics of tribulus terrestris plant extract (TTPE) on aluminum in hydrochloric acid (1.0 N HCl) environment using gravimetric, thermometric and morphological methods. The outcome of the empirical studies shows that increase in TTPE concentration reduces the rate of corrosion and increase in temperature worked against the efficiency of TTPE in 1N HCl medium on aluminum. The thermodynamic parameters obtained from the thermometric studies revealed physical adsorption on the surface of aluminum by TTPE in 1N HCl medium. Adsorption curves obeyed Langmuir, Temkin and Freundlich adsorption isotherms. The morphological studies show that there was effective adsorption on the surface of aluminum by TTPE in 1N MHCl solution and the impedance studies confirmed it. The electrochemical studies show a mixed type and supported the formation of a protective layer of inhibitor on a metal surface.

3.1 Reaction Mechanism

Aluminum reacts with acid in the following manner according to Prabhu and Padmalatha (2017):

$Al + H_2O = AlOH_{ads} + H^+ + e^-$ (1)

$AlOH_{ads} + 5H_2O + H^+ + e^- = Al^{3+} + 6H_2O + e^-$ (2)

$Al^{3+} + H_2O = (AlOH)^{2+} + H^+$ (3)

$(AlOH)^{2+} + X^- = AlOHX^+$ (4)

The series of the above reactions lead to the formation of the soluble product as shown in equation four (4).

They said again that in sodium hydroxide, the reactions will be:

$Al + 3(OH)^- = Al(OH)_3 + 3e^-$ (5)

$AlOH_3 + OH^- = Al(OH)_4^- + 3e^-$ (6)

$3/4O_2 + 3/2H_2O = 3OH^-$ (7)

$3H_2O + 3e^- = 3/2H_2 + 3OH-$ (8)

$Al + OH^- + 3/4O_2 + 3/2H_2O = Al(OH)_4^-$ (9)

$Al + 3/2H_2O + OH^- = 3/2H_2 + Al(OH)_4^-$ (10)

The above reactions include, the dissolution of aluminum leading to anodic and cathodic corrosion due to formation of hydroxide which dissolves continuously.

3.2 Corrosion Study Methods

3.2.1 Gravimetric study: this is a corrosion study method whereby the corrosion products are quantitatively measured. It rely on measuring the weight difference between initial weight and final weight of the mass of the metal being measured (Gravimetric Analysis, nd)

3.2.1.1. Weight loss measurement

This is the simplest corrosion study method which involved immersing the carefully cut and cleaned sample (coupon) in a corrosive environment with or without inhibitor in the solution after initial weight (W_i) have been measured. The immersed sample is expected to last for predetermined time and final weight (W_f) taken. From there, weight loss, corrosion rate, efficiency and other corrosion parameters calculated

Weight loss, $W_L = W_i - W_f$ (11)

Corrosion Rate$\left(\frac{MM}{Y}\right) = \frac{87.6W_L}{DAT}$ (12)

Where:

W_L = weight loss in milligrams
D = metal density in g/cm^3
A = area of the sample in cm^2
T = time of exposure of the metal sample in hours

$$Efficiency(\%E) = \frac{W_i - W_f}{W_i}\% \quad (13)$$

3.2.2 Atomic absorption spectroscopy

This method involved metallic ion concentration measurement in a corrosion solution with or without inhibitors. This method is carried out with atomic absorption spectroscopy (AAS) machine. It is a quantitative one (Al-Bagawi, 2021). The principal equation necessary to calculate the metal ions concentration present in the sample:

$$C = \epsilon l A \quad (14)$$

where C = metal ion concentration in the sample, A= sample absorbance, ϵ is the molar absorptivity of the metal, and l is the path length of the light through the sample.

3.2.3 Gasometrical measurements (hydrogen evolution measurements)

This is another very important method for corrosion study whereby hydrogen gas evolution from the acid-metal reaction to determine the rate of corrosion. The evolved gas can be measured using secundomer in displaced water in calibrated cylinder. The rate of evolution of the gas (RV_H) is determined from the slope of the graph of volume of gas evolved (v) versus time (t), according to equation (15)

$$RV_H = \frac{\Delta V}{\Delta t} \quad (15)$$

and the inhibitor surface coverage (θ) and efficiencies (IE%) determined using equation (16) and (17) respectively.

$$\theta = \frac{RV_{H\,blank} - RV_{H\,inhibior}}{RV_{H\,blank}} \quad (16)$$

$$IE = \left(\frac{RV_{H\,blank} - RV_{H\,inhibior}}{RV_{H\,blank}}\right) x\, \frac{100}{1} \quad (17)$$

Where $RV_{H\ blank,}$ and $RV_{H\ inhibitor,,}$ and the rate of hydrogen evolution in the absence and presence of the inhibiting molecules respectively. This gasometric technique has been corroborated by other well established corrosion rate determination techniques, including weight loss, thermometric and electrochemical techniques (Okafor *et al*., 2012; Umoren *et al*, 2013).

3.2.4 Thermometric measurements

This method is like the weight loss method except that it is carried out above room temperature. It is to determine how temperature affects the rate of corrosion. Arrhenius equation is important model to use.

$$\frac{InCR_2}{InCR_1} = x = \frac{E_a}{R}\left(\frac{1}{T_2} - -\frac{1}{T_1}\right) \quad (14)$$

CR_1 is corrosion rate at temperature T_1 and CR_2 is corrosion rate at temperature T_2 with Ea as activation and R as gas constant

3.2.5 Acidification method

In this method, acid concentration is measured with pH meter at the different inhibitor concentration before and after immersion of metallic coupons

3.2.6 The Potentiodynamic Polarization (PDP)

It involves electrode potential selected rate variation from the application of current in an electrolyte. It is used to study corrosion behaviour in corrosive environment. Both reduction and oxidation occurs on the metal surface in the electrolyte (Potentiodynamic, 2019; Mehr, 2022) . A platinum wire was used as a counter electrode and a saturated calomel electrode (SCE) as the reference electrode to which all potentials are referred (Al-Bagawi, 2021). Corrtest machines (CS100E, CS350 etc), Gamry instruments are some equipments used for testing potentiodynamic of corrosion systems.

3.2.7 Microscopic Examination

Metallurgical, optical and scanning electron microscopic examinations are carried out to examine the level of corrosion of the metallic materials subjected to corrosive environment. After cleaning the surfaces, these microscopes are used to examine the surfaces to confirm the level of corrosion. It is also very important to use this method to confirm the particular type of corrosion

Ebert (2012) also listed in American society of testing materials (ASTM) G 31 as procedures and other methods as ASTM Test Method for static leaching of monolithic waste forms for disposal of radioactive waste (C 1220), ASTM Standard Test Method for Diffusive Releases from Solidified Waste and a Computer Program to Model Diffusive, Fractional Leaching from Cylindrical Waste Forms (C 1308), modified ASTM C 1220 test and vapor hydration test (VHT)

CONCLUSION

Continuous development of the packaging industries will lead green corrosion inhibition studies of aluminum to grow from strength to strength as they are humanly and environmentally friendly, affordable and available. Several green plants have proven to have the capacity to inhibit aluminum and its alloys corrosion with efficiencies ranging above 60%. Gravimetric (weight loss) method is the most popular. Others include atomic absorption spectroscopy (AAS), potentiodynamic polarization (PDP), electrochemical impedance spectroscopy (EIS), scanning electron microscope (SEM) etc. These methods are used to determine weight, surface coverage, efficiency, corrosion rate, surface polarization, amount of ions in solution and surface morphology. Lastly, phytochemical screening is carried out to know the plants chemicals performing the inhibition.

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