International Journal of Biosciences | IJB |
ISSN: 2220-6655 (Print), 2222-5234 (Online)
http://www.innspub.net
Vol. 12, No. 2, p. 126-135, 2018

RESEARCH PAPER OPEN ACCESS

Sustainable Solution to reduce corrosion rate in mild steel using aqueous and organic extracts of Plants

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Key words: Corrosion, Inhibition, Plants, Mild steel, Factorial design

http://dx.doi.org/10.12692/ijb/12.2.126-135 Article published on February 26, 2018

Abstract

Three plants *Sageretia thea*, *Calotropis procera* and *Cucumis melo* were selected to investigate the corrosion inhibition potential against mild steel. Different test concentrations (0.05gm, 0.1gm and 0.3gm) of aqueous, methanol and ethanol extracts of the selected plants were tested in 2 Molar HCL Solution. The main and interactive effects of Plant type sample concentration and solvent used were studied using 3^3 factorial design of experiment with three replicates by taking readings at 12 hrs, 24 hrs and 48 hrs systematically. Weight loss measurements were considered enough to determine corrosion inhibition efficiency of the selected plants. Statistical results indicate that Main effect of plant type; Joint effect of plant type and extract used; and interaction effect of plant type, extract used and concentration level were found significant at 12 hrs and 24 hrs readings: Also Main Effect of Plant type; Joint effect of Plant type and extract used were found significant factors at 48 hrs readings in the corrosion control via used green resources. Percent weight loss was calculated for all the test samples. Our results indicate that aqueous and organic extracts of *Sageretia thea, Calotropis procera* and *Cucumis melo* have impressive anti-corrosion potential against mild steel resulting in remarkable inhibition efficiencies for the included plants.

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Introduction

Materials corrosion presents a massive problem in industries and therefore remained a popular area of surge for material scientists and engineers. Different ideas have been proposed to control the process of corrosion, while the mostly adopted method has been isolation of material from the corrosive media (Ferreira *et al.* 2004). Researchers are continuously looking for cheap, effective and economical ways to control corrosion of metals. Carbon steel has a number of uses mainly as a used as structural and pipeline materials in the oil and gas industry. Mild steel is represented by low carbon content. For cleaning of such pipelines and gigantic carbon steel sheets, acidification is commonly performed (Migahed and Nassar 2008). However, the use of abrasive media such as acids can significantly induce and accelerate the rate corrosion. In order to protect mild steel for the abrasive effects of acidification, corrosion inhibitors are used (Soltani *et al.* 2016). Many organic chemical entities have been successfully used as corrosion inhibitors. These compounds possess strong affinity for the metal surface and can become adsorbed on their surface of the metal because of their bounded functional groups(Ferreira *et al.* 2004). Organic chemicals containing Nitrogen, Oxygen and Sulphur like 1H-pyrrole-2,5-dione derivatives (Morales-Gil *et al.* 2014), 1,4-di[1′- methylene-3′-methyl imidazolium bromide]-benzene (Hanza *et al.* 2016), N-alkyl-sodium phthalamates (Flores *et al.* 2011), 1-(2-ethylamino)-2-methylimidazolin (Cruz *et al.* 2004), Schiff bases (Yurt *et al.* 2004), tris (benzimidazole-2-ylmethyl) amine (Roque *et al.* 2008), pyrimidothiazine derivative (Belayachi *et al.* 2015), 2-mercaptobenzimidazole (Morales-Gil *et al.* 2015) and3-amino-1,2,4-triazole-5-thiol (Mert *et al.* 2011), has been used as corrosion inhibitors in acidic media. However, in spite of being effective, the widespread applications of these compounds are limited because of certain disadvantages like cost and being hazardous to the environment. In order to overcome these problems of hazardous waste greener methods have been proposed and therefore importance has been given to the plants with rich phytochemistry. In addition to being eco-friendly and environmentally acceptable, they are inexpensive, easily available and also renewable (Soltani *et al.* 2016). Therefore, scientists have continuously investigated greener, sustainable, effective and environmental friendly method for corrosion control.

Over the previous few years, numerous plants have been reported to have impressive corrosion inhibition potential. *Osmanthus fragran* (Li *et al.* 2012), *Ferula gumosa* (Behpour *et al.* 2009), *Hibiscus rosasinensis* (Anuradha *et al.* 2007), *Dacryodis edulis* (Oguzie *et al.* 2010), *Argania spinose* (Mounir *et al.* 2016), *Cheilocostus speciosus* (Hassan *et al.* 2016), *Gossipium hirsutum* (Abiola *et al.* 2009), *Thymus vulgaris* (Ehsani *et al.* 2017), *Withania somnifera* (Saxena *et al.* 2016), *Aloe vera* (Ajayi *et al.* 2016), *Cordia dichotoma* (Khandelwal *et al.* 2016), *Heliotropium indicum* (Sharma and Sharma 2016), *Sida acuta* (Umoren *et al.* 2016) etc. has been already reported to have significant inhibition of corrosion. The proposed reason for the corrosion inhibition properties of plants is the presence of active organic compounds that can form a layer or film over the surface of metal s subsequently lowering the contact between abrasive media and metal surface which limits the rate of corrosion (Gerengi and Sahin 2011). Main objective of this study is to study the corrosion inhibition potential of few medicinal plants in Pakistan. It is a comprehensive study reported for the first time on the corrosion inhibition efficiency of three medicinal plants of Pakistan i.e. *Sageretia thea*, *Cucumis melo* and *Calotropis procera*. It is noteworthy to mention that earlier studies mostly included only aqueous plant extracts while the novelty of the present report lies in involving different parameters. Inhibition efficiency was monitored with respect to solvent type, concentration and time. In addition, significant factors have been investigated by employing various statistical tools.

Objective of this research is basically to Identify and characterize cheap and eco-friendly solution to control corrosion that must be sustainable as well. For Screening and characterizing of the aqueous and organic (methanol and ethanol) extracts of selected plants *Sageretia thea*, *Calotropis procera* and *Cucumis melo* were tested for corrosive inhibitor property against mild steel. Different factors were also identified that are significant and play major rolein corrosion inhibition. This work also include to study the significance of main effects along with interaction effects as discussed in results.

Material and method

Selection of plants

Extensive and rigorous review of literature was performed for the selection of plants. Plants were selected based upon their medicinal uses, rich phytochemistry, novelty and availability. Based on the inclusion parameters, *Sageretia thea*, *Cucumis melo* and *Calotropis procera* were selected.

Collection, Identification and Processing

Plant specimens were collected and identified in Department of plant sciences, Quaid-i-Azam University, Islamabad, Pakistan, followed by proper collection. All plants were collected from Islamabad, washed in running distill water and kept for shade drying for about 2 weeks. After drying phase, the left over material was excised and grounded to fine powder in a Willy mill. Leaf powder was stored for further use.

Extraction

Grounded plant material of about 30g was added to 200 ml of solvents i.e. distill water, ethanol and methanol in an air tight reagent bottle. The solution was kept air tight for 2 weeks with daily stirring thrice a day. The resultant solution was filtered twice to remove the residual waste and the test samples were dried by heating them at ~ 60°C for 4 hours. The leftover, was considered as plant extract and test dilutions were made as 0.3 mg/ml, 0.1 mg/ml and 0.05 mg/ml in 2M HCL.

Experimental Data

Experiment was carried out in molecular systematic and applied ethno botany lab of QAU Islamabad. Experiment was run in randomized order in order to eliminate any biasness and specific trend that could have contributed in to results obtained.

Statistical graphs of the experiment performed shows that data follow normal distribution with no specific trend in the experiment same as for 12 hrs, 24 hrs and 48 hrs as shown from the Figure (1).

Experimental procedure

2 M HCL solution was prepared using 37% analytical grade HCL. Mild steel strips with 2 mm thickness and 50 mm length were used to check the inhibition efficiency. Before experiments, the steel coupons were aggressively abraded using emery papers with grit of 240, 400, 800 and 1200. Samples were degreased in xylene and washed with distill water for experiment. Weight loss experiment.

Steel coupons were carefully placed in 2M HCL media and 2M HCL supplemented with the test plant samples. Weight loss measurements were taken at different times during the experiment i.e. 12 hrs, 24 hrs and 48 hrs. Steel coupons in 2M HCL were used as control. The Percentage Inhibition efficiency was calculated using the following equation (Okafor *et al.* 2008);

$$IE = \frac{W0 - W1}{W0} \times 100$$

Where W0 and W1 are the weight of sample in absence and presence of inhibitor.

Procedure

Experimentation procedure was designed in Minitab, a tool used for statistical analysis. Three factors were defined Plant type. Extract used and Concentration level with all having three levels with three replicates. A total of 81 runs were executed in a random order in order to eliminate the effect of any specific trend for the experiment. Random run order was generated using Minitab and experiment was preceded with the same order. Weight loss values were taken with the help of digital balance with accuracy to 3 digits and reading were systematically recorded after 12 hrs, 24 hrs and 48 hr respectively.

Results

Significant factors

Different factors directly or indirectly influencing the rate of corrosion were investigated and the significant factors were identified.

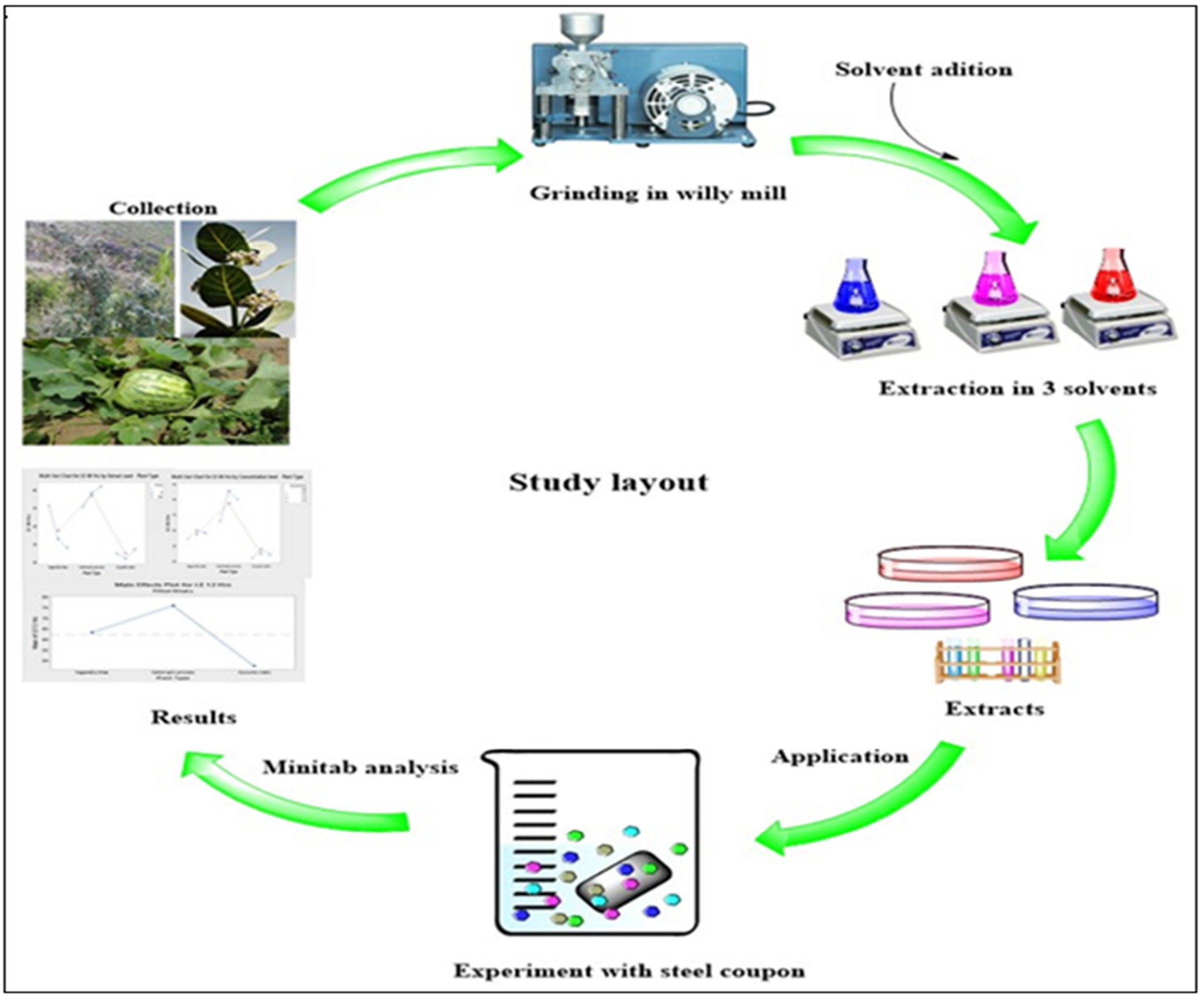
Different parameters investigated are summarized in table 1. Plant type and Plant type*extract used were identified as the most significant factors driving the corrosion inhibition in the abrasive media.

Table 1. Different significant factors studied in the present work.

Factors	Effect		
	12 Hrs	24 Hrs	48 Hrs
Plant type	Y	Y	Y
Extract Used	-	-	-
Concentration level	-	-	Y
Plant Type*Extract Used	Y	Y	Y
Plant Type*Concentration level	Y	-	-
Extract Used*Concentration Level	-	-	-
Plant type*Extract Used* Concentration Level	Y	Y	-

"Y": Significant; "-": Non Significant.

The effect was indicated as significant across 48 hours. From the table, it can also be concluded that concentration is as a significant factor in limiting the rate of corrosion uptil an optimum level however its role shows a decreasing trend with concentration level when used beyond the optimum level.



Graphical abstract.

Inhibition efficiencies relative to time

Mean inhibition efficiencies were calculated with the respect to time as indicated in Figure 2 indicate the calculated inhibition efficiencies with respect to time. It can be concluded that *Calotropis procera* was most effective in inhibiting the rate of corrosion in mild steel in an abrasive 2M HCL media. The inhibition efficiency for *Calotropis procera* was found to be > 70% across 48 hours that indicates their effectiveness and stability in the abrasive media. *Sageretia thea* was also found to be stable across 48 hours, however the inhibition efficiency calculated was approximately around 50%. *Cucumis melo* was found to be least effective in corrosion inhibition in mild steel.

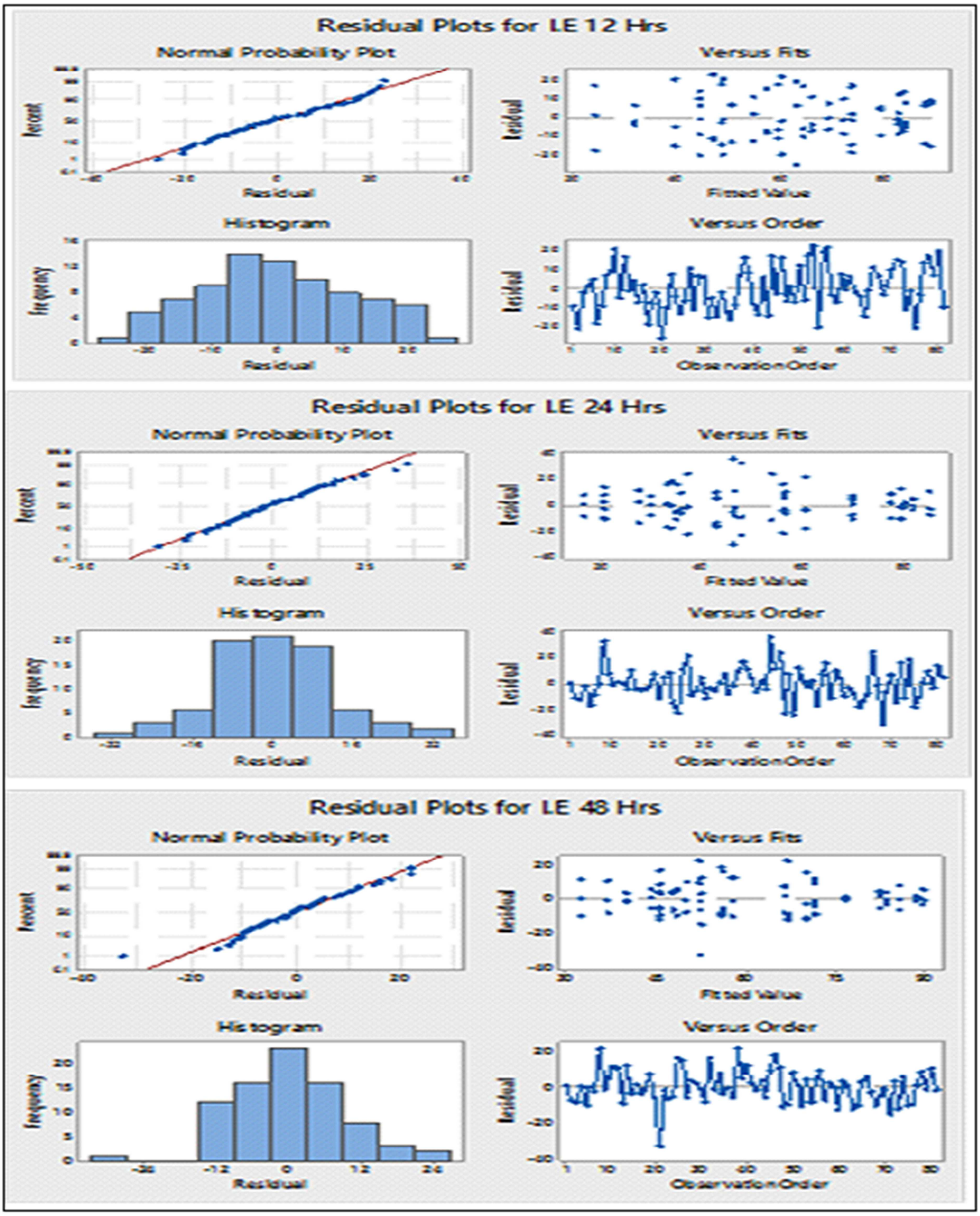


Fig. 1. Data Distribution and Normality Plots of experiment.

Inhibition efficiency relative to extract type

IE were calculated for plant extracts prepared in 3 different solvents i.e. water, methanol and ethanol as shown in Fig 3. To find out the most effective solvents the results of 3 plants were compared. Our results indicated mixed type results depending on the plant sample used.

For *Sageretia thea*, the order of corrosion inhibition efficiencies was investigated as water > ethanol > methanol while for *Caltropis procera* the order was found as methanol > ethanol > water. In *Cucumis melo*, the increase in inhibition efficiencies were investigated as methanol > water > ethanol.

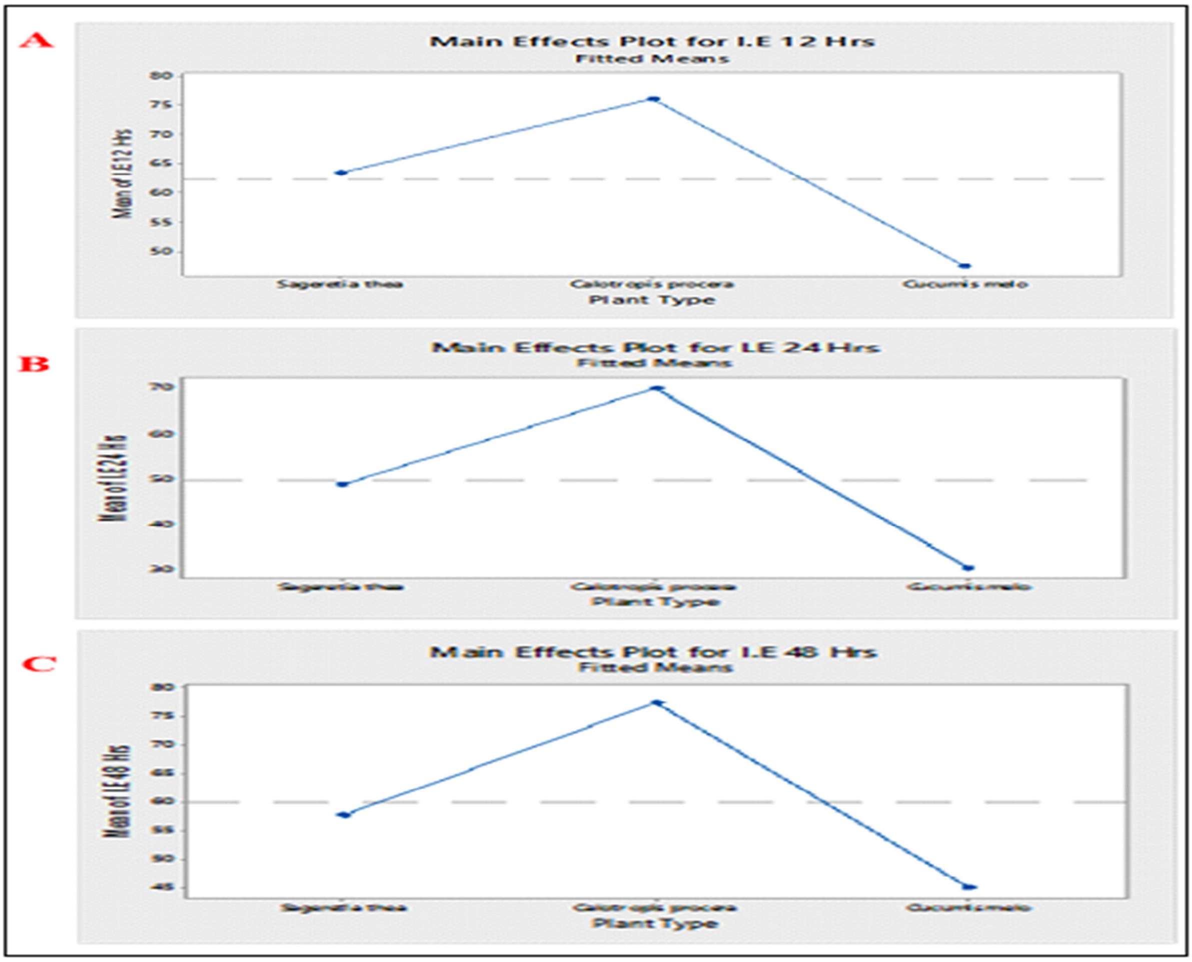


Fig. 2. IE in response to immersion time.

Inhibition efficiency relative to concentration

Concentration of the plant was also identified to have a significant role in corrosion inhibition potential. The relationship between various concentration of the test sample and their respective inhibition potential is indicated in Figure 4.

The optimum concentration for each plant was identified. Our results indicate an optimum result at concentration level of 0.1 g/ml. Increasing concentration beyond 0.1 g/ml decreased the inhibition efficiency of plant extracts.

Discussion

Corrosion inhibition is the result of interaction between the surfaces of the material and the active phytochemical components/functional groups present in plants that can adsorbed to the surface of the material and alienate the material surface from abrasive media. A plausible mechanism is suggested in figure 5. Weight loss readings were recorded by involving various parameters such as concentration of the plant extract based corrosion inhibitor, effect of their concentration and type of solvent used were studied.

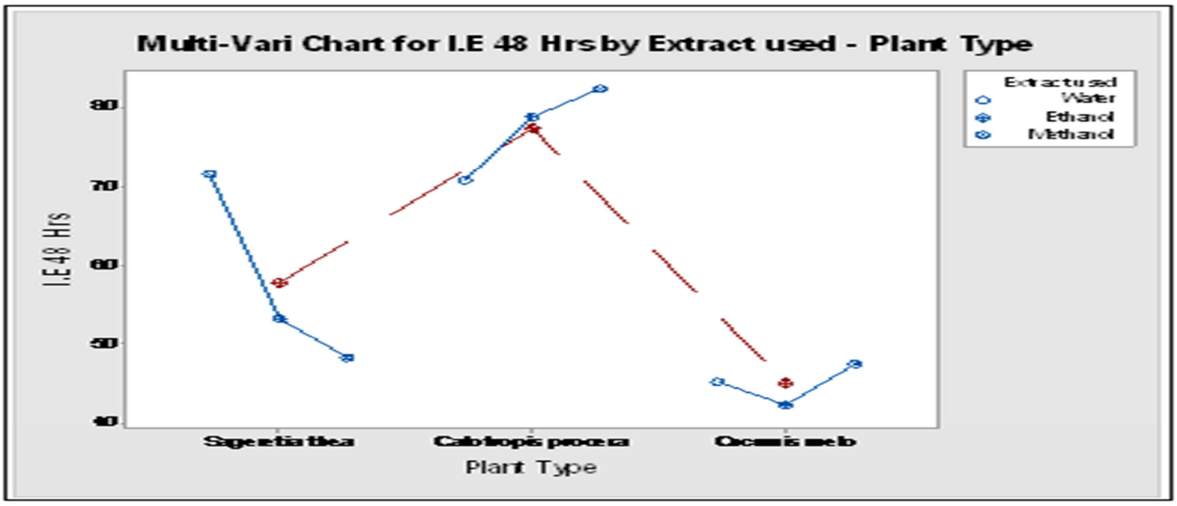


Fig. 3. IE in response to extract used.

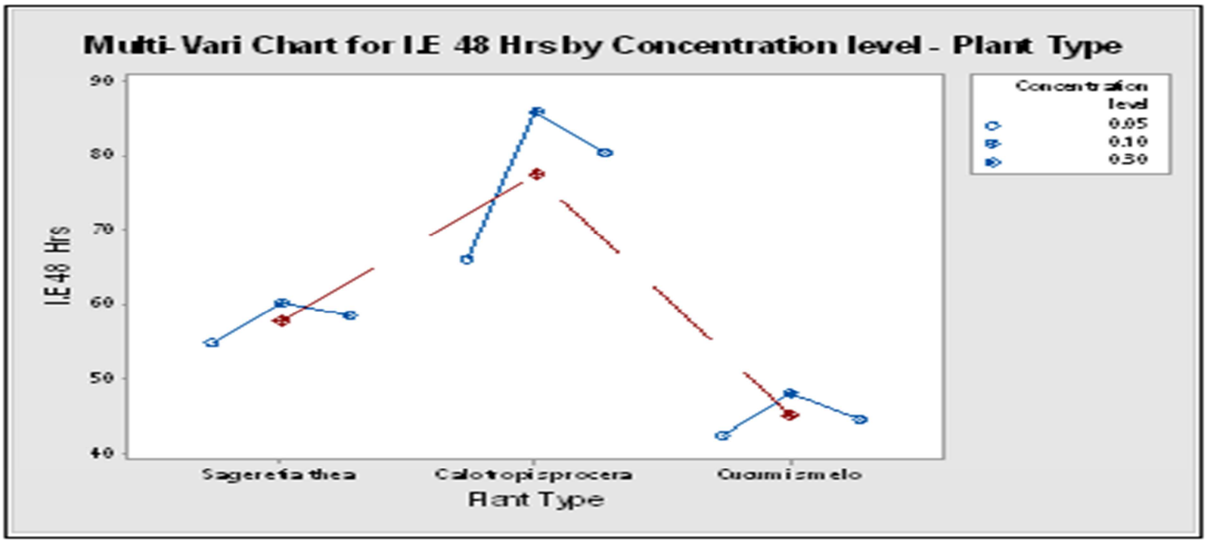


Fig. 4. IE in response to concentration level.

It was investigated that after initial increase in the applied concentration (0.05 g/ml to 0.1 g/ml) the inhibition potential increased, however, increasing concentration beyond 0.1 g/ml decreased the inhibition potential of the plant extracts. Decrease in the inhibition potential with increase in concentration has been reported previously (Ajani *et al.* 2014; Arockiasamy *et al.* 2014).

It has been proposed that increasing concentration can weaken the metal-inhibitor interaction subsequently leading to the replacement of corrosion inhibitor molecules with water or chlorine ions resulting in decreased inhibition efficiency. From our results it is cleared that the adsorbed film of the phytochemicals serve as barrier against abrasive media, and hence contribute to corrosion resistance in the mild steel.

The mild steel gets blocked and the adsorbed film of inhibitor acts as a physical barrier between mild steel and corrosion medium.

Analyzing all the weight loss measurements for the test samples suggested that *Calotropis procera* was found as most effective as compared to *Sageretia thea* and *Cucumis melo*. The precise order that followed is *Calotropis procera*> *Sageretia thea*> *Cucumis melo*. The active components in plants that can inhibit the corrosion varies from plant to plant and type of the extract used and within plant extracts (Chigondo and Chigondo 2016).

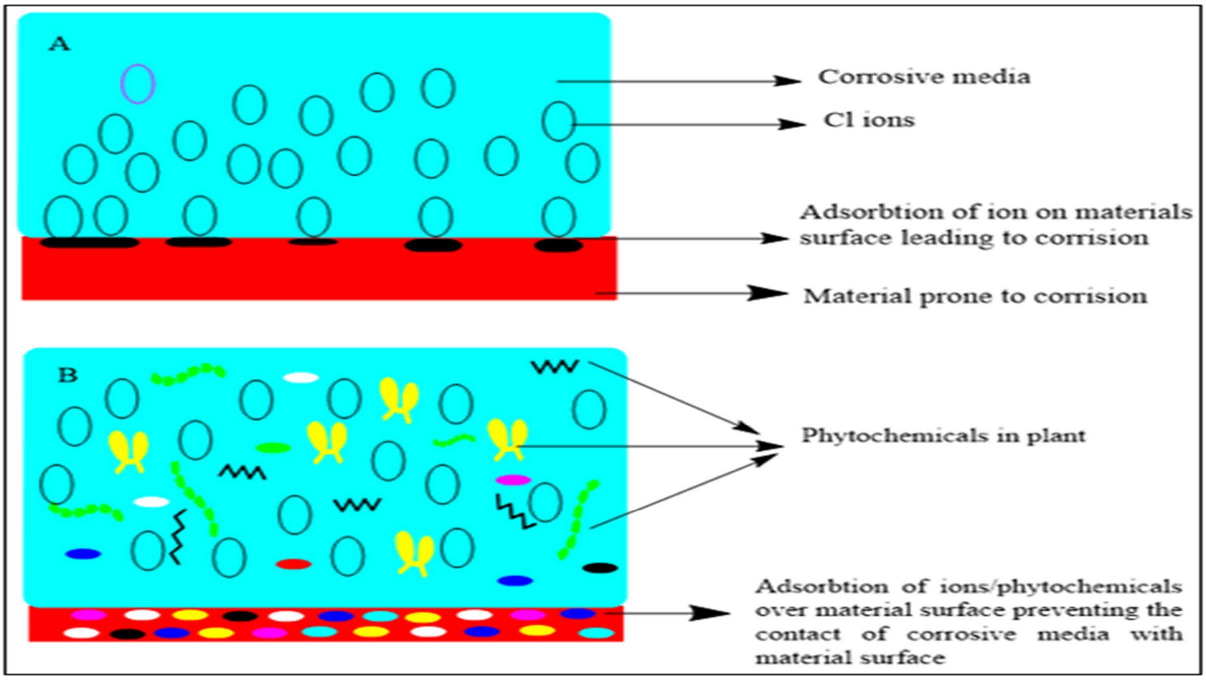


Fig. 5. Plausible mechanism of green corrosion inhibition.

Conclusions

Natural plant extracts provides an ideal platform to investigate the green and eco-friendly corrosion inhibitors. The potential of plants is still largely unexplored for potential corrosive inhibitors. Although being effective in abrasive acidic media, their corrosion inhibition studies needs to be extended to other corrosion mediums like in hydrogen sulfide, carbon dioxide and, sulphur dioxide. We further recommend studies through impedance spectroscopy, Tafel polarization and surface analysis through high resolution microscopy on the material surfaces to get insights into the mechanism aspects of green corrosion inhibition.

Acknowledgments

The authors are indebted to Department of Industrial Engineering, UET-Peshawar and Molecular Systematics and Applied Ethnobotany lab, Quaid-i-Azam University, Islamabad for providing necessary assistance to carry out the research project.

Conflict of interests

The authors declare no conflict of interest.

Authors contribution

Ali Hamza Khalil conceived the idea and executed the experimental. Misbah Ullah provides assistance in carrying out research work. Ali Hamza Khalil and Misbah Ullah prepared the draft manuscript.

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