Review

A critical review on the recent studies on plant biomaterials as corrosion inhibitors for industrial metals

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ARTICLE INFO

ABSTRACT

Plant biomaterials as inexpensive, nontoxic, biodegradable materials are found abundantly in nature. They contain heteroatoms and/or pi electrons that make them candidate for metals corrosion inhibitor. In recent years, numerous research works have been undertaken on plant biomaterials as metals corrosion inhibitor in different corrosive media. This review critically looks at the researches done in the years 2013–2018. Corrosive environments covered include acidic, basic, neutral, aqueous, geothermal fluid and artificial saliva. The major findings and the mechanism of inhibition has been elucidated. The missing gap in this area of research has been pointed out and future direction proposed.

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Background

Preamble

Metals corrosion, defined as the deterioration of desired metals properties on interaction with certain elements that are present in the environment is a normal and unavoidable process. Besides gold, platinum and a few others, metals exist in nature in impure form mostly as sulphides or oxides and are stable [1]. To obtain pure metals, energy is expended (e.g. in a blast furnace) and this make the pure metals to exist in a higher energy state than the ore. Consequently, metals are in constant strive to revert to their most stable and lowest energy state (the ores) and the easiest way is by corrosion. Certain factors such as metals reactivity, impurities, solution pH, and temperature as well as industrial practices like acidizing, acid cleaning, pickling, etc. facilitate metals corrosion. Highly reactive metals have the tendency to corrode more than the less reactive ones. Impurities help in setting up voltaic cells and this increase the rate of corrosion. Increase in temperature and pH equally increases corrosion rate of metals.

Forms of corrosion

Metals corrosion comes in different forms. Fontana and Greene [2] grouped them into eight forms namely; uniform or general attack, galvanic or two-metal corrosion, crevice corrosion, pitting, intergranular corrosion, selective leaching or parting, erosion corrosion, and stress corrosion. The general corrosion, which is the most common form of corrosion, is often characterized by a chemical or electrochemical reaction that proceeds evenly over the entire exposed surface or over a large area. Galvanic corrosion occurs between two dissimilar metals when they are exposed to a corrosive environment [2]. Localized corrosion is a form of corrosion that occurs within crevices and other shielded areas on metal surfaces exposed to aggressive environment. Pitting corrosion gives rise to holes or pits on the metal surface [2]. Intergranular corrosion is a form of corrosion where the boundaries of crystallites of material are more prone to corrosion than their insides [2]. Selective leaching is the dislodging of one element from a solid alloy by corrosion processes [2]. A typical example is the selective removal of zinc in brass alloys. Erosion corrosion is the acceleration in the rate of dissolution of metal due to relative migration between a corrosive system and the metal surface. Finally, stress-corrosion cracking is the form of corrosion that is caused by the simultaneous presence of tensile stress and a specific corrosive medium [2].

Impact of metals corrosion on economy and life

Metals corrosion is a serious challenge that has bitten hard on the world economy. The annual global corrosion cost is estimated at USD $2.5 trillion [3]. Recent reports revealed that the indirect cost due to corrosion for the United States of America (USA) is over $ 1.1 trillion in 2016 [4]. The indirect corrosion cost was put at USD $ 90.4 billion in 1999 [5], USD $ 9.2 billion in 1977 [6], and USD $ 90.4 billion in 1999 [5] for Japan. China and Saudi Arabia recorded~ USD $ 310 billion in 2015 [3] and USD $ 900 million in 2003 [7] respectively. This cost is expected to increase since the world is witnessing technological advancement. Metals corrosion has also resulted in serious catastrophes. For instance, on November 22, 2013, due to corroded under-ground Sinopec pipeline, the Donghuang II oil pipeline unexpectedly exploded in Qingdao, eastern China killing 62 people and injuring 136 [8]. Beside the tragic loss of human life, the explosion caused the company a direct economic loss of 750 million yuan (USD $124.9 million) [8]. On August 19, 2000, a 30-inch natural gas pipeline owned by El Paso

Natural Gas (EPNG) exploded and released gases that burnt for 55 min [9]. Twelve fatalities, including children and infants were reported as well as destruction of three vehicles [10]. According to the National Transportation Safety Board, the cause of the explosion was due to a significant reduction in the pipe wall thickness arising from severe internal corrosion on a 50-year-old pipeline [11]. Again, on August 2013, because of corrosion around faulty seals, 300 metric tons of contaminated water leaked from a storage tank of the Fukushima nuclear plant into the sea [12]. Fishing in the waters was banned and clean-up was estimated to cost over USD $100 billion and may last up to 40 years before it could be completed [13].

Corrosion inhibitor/Selection requirements

Several strategies have been adopted in the fight against corrosion. These include design, materials selection, electrochemical (anodic and cathodic protection), coatings, and the use of corrosion inhibitors. Of all these techniques, the use of corrosion inhibitor is considered the most cost effective and the easiest to apply [14]. Corrosion inhibitors are chemicals that can decrease the rate of metals dissolution when present in minimal concentration in aggressive environment [14]. The inorganic compounds, particularly chromates and its derivatives are known for their potent inhibitive strength [15]. Nevertheless, their negative impact on human lives and the natural environment has necessitated the discontinuation of their use [15]. Legislative bodies like the Registration, Evaluation, Authorization and Restriction of Chemicals (REACH) and Paris Commission (PARCOM) have clearly spell out the requirements for a substance to be accepted as a green metals corrosion inhibitor and they are: (i) must be non bioaccumulative, (ii) must be biodegradable, and (iii) must have zero or very low marine toxicity level [16]. According to these bodies, toxicity is to be defined as both LC_{50} and EC_{50}. EC_{50} implies the effective concentration of chemical needed to adversely affect 50% of the population while LC_{50} is the quantity of chemical required to kill 50% of the population [16]. For biodegradation acceptance, PARCOM requires that environmental assessment be undertaken to ascertain the length of time a chemical can persist in the natural environment and the allowable limit is that 60% must degrade after 28 days [16]. The bioaccumulation evaluation takes into cognizance the extent of chemical buildup in the body [16]. It is examined by partition coefficient – a test of the distribution of the chemical between an octanol and water mixture expressed as Log (Po/w). For a chemical to be accepted, it must have log (Po/w) <3.

The choice of natural products as corrosion inhibitors

Natural products including plant extracts, apart from satisfying the above listed requirements are easily accessible and economically viable. Hence, the 21st century has witnessed tremendous research activities on natural products [17–20]. In the area of corrosion inhibition, there has been outpour of researches utilizing extracts from various parts of plant. In fact, it has been found that the bioactive compounds in plant extracts are as effective as synthetic inhibitors [19,20]. Some authors have carried out reviews on plant extracts as corrosion inhibitor in diverse kinds of corrosive media. Dar [17], in 2011 reviewed plant extracts and oils as corrosion inhibitors for different substrates and diverse kinds of aggressive media. Similar reviews had been undertaken almost on a yearly basis; 2012 [19], 2013 [18,20,21], 2015 [22], 2016 [23–25], 2017 [26], and 2018 [27,28]. Nevertheless, most of these reviews only highlighted what has been reported on plant extracts as corrosion inhibitor without actually identifying the research gap and probably forecasting the future perspective. Motivated by the

volumes of research articles published on plant extracts as metals corrosion inhibitor in the recent years, precisely between 2013–2018, we deemed it necessary to carry out a critical review so as to provide an update-to-date information, identify the research gap, and propose the future direction on this area of research. This review article is designed to serve as a lead-way material for the scientists working on this area, industries that are into the production of green inhibitors, and students studying corrosion and corrosion inhibition.

Application of plant extracts for metals protection

HCl environment

Hydrochloric acid is a famous industrial acid ranked topmost in pickling and oil wells acidization practices [29]. The preference of HCl over other acids for pickling exercise is due to the fact that, less pickling time and better surface quality can be achieved at low temperatures [29]. In fact, the practice does not require temperature above 30 °C as this will cause excessive amount of hydrogen chloride gas to be liberated. The recommended concentration of HCl acid for pickling is 5–15% while higher concentrations up to 32% are for acidization [29]. It appears from recent reports on plant parts extracts as anticorrosive agent in HCl environment that metals corrosion retardation during pickling is the target. For instance, most reports are on HCl concentration of 0.1 M [30–41], 0.5 M [42–65], 1 M [66–169], and 2 M [170–174]. Beyond these concentrations, there seems to be no report. The reason could be that, extracts from plant parts might not withstand the severity of acidization process (i.e the remarkably high acid concentration and operational temperatures).

In the acid concentration range (0.1–2 M) studied, plant parts extracts are effective corrosion inhibitors behaving mostly as mixed type corrosion inhibitor although few reports have shown that they could also act as anodic as well as cathodic type inhibitors (Table 1). The corrosion inhibition is by adsorption mechanism occasioned by the presence of active organic moieties in them. Actually, plant parts are embodiment of phytochemicals such as tannins, polyphenols, flavonoids, phlobatannins, anthroquinones, saponins, alkaloids, organic sugars, etc and each of this phytocompound can interact with a metal surface and obstruct either the anodic, cathodic, or both corrosion reactions. For example, El Hamdani et al. [78] demonstrated that alkaloids in Retama monosperma (L.) Boiss. seeds extract was responsible for the mitigation of carbon steel corrosion in 1 M HCl medium. Raja et al. [136] showed that the alkaloids isolated from Alstonia angustifolia var. latifolia leaves were good inhibitor for mild steel in 1 M HCl. The alkaloids exhibited inhibition efficiency higher than 80% at concentrations between 3 and 5 mg/L. Bouknana et al. [154] examined the effect of the phenolic (OOMW-Ph) and non-phenolic (OOMW-NPh) fractions of the extract of olive oil mill wastewaters on the dissolution of steel in 1 M HCl using weight loss measurements and electrochemical polarization. It was found that, the referred compounds reduced the corrosion rate of the substrate attaining 88.9% and 89.1% inhibition efficiency by OOMW-Ph and OOMW-NPh, respectively. Odewunmi et al. [80] linked the corrosion inhibition of mild steel in HCl environment by watermelon rind extract to the presence L-citrulline (an amino acid). Inhibition of Al corrosion in HCl environment by Cashew nut testa extract was linked by Nnaji et al. [63] to the presence of tannins. For the inhibition of steel corrosion by morus Alba pendula leaves extract, Jokar et al. [81] associated it with the presence of flavonoids such as morusin, kuwanonC and kuwanonG, phenolic acids and pyrrole alkaloids in the extract. However, corrosion scientists have been so curious to know the actual component

responsible for the corrosion inhibition of a particular extract and probably isolate the component. Such knowledge would allow the most favorable condition for the component to be established and when necessary, for appropriate modification approach to be designed. It will also be easy to propose the mechanism of inhibition. Most recent reports are therefore focused on the isolation of the active component (s) responsible for the anticorrosive property of plant parts extracts. Chevalier et al. [72] investigated the corrosion inhibition property of Aniba rosaeodora alkaloidic extract on the corrosion of C38 steel in 1 M HCl acid using electrochemical techniques. The authors found that the extract, which acted as a mixed type corrosion inhibitor, was efficient in retarding the dissolution of the metal in the studied corrosive environment. Studies on the phytochemical constituents of the total alkaloids extract using Nuclear magnetic resonance spectroscopy (NMR) and X-ray photoelectron spectroscopy (XPS) revealed that anibine was actually the major alkaloid responsible for the anticorrosion property of Aniba rosaeodora alkaloidic extract. Raja et al. [119] linked the corrosion inhibition by the alkaloid extracts of leaves and bark of Ochrosia oppositifolia for mild steel in 1 M HCl medium to the presence of isoreserpiline (ISR). In fact, 25 mg/L of ISR isolated from the plant parts was found to possess 93% inhibition efficiency while the crude extract from the plant leaves and stem had 87% and 94% respectively. M'hiriet al. [30] however, observed that the retardation strength of crude orange peel extract and selected antioxidant compounds (neohesperidin, naringin, ascorbic acid) evaluated separately by electrochemical approaches was remarkably different. M'hiri et al. noticed that, for all the concentrations studied, the crude extract exhibited significant corrosion inhibition than the selected antioxidant compounds (Fig. 1). In the report of Ghazouani et al. [99], polyphenols mainly neochlorogenic acid, rutin, and chlorogenic acid present in quince pulp extract were the active compounds that inhibited the corrosion of carbon steel in 1 M HCl solution. In Anthemis pseudocotula extract, the highly potent anticorrosive compound isolated was luteolin-7-O-b-D-glucoside of which 0.446 mM was found to offer 94.8% protection to mild steel surface in 1.0 M HCl solution [110]. In addition, Alvarez et al. [73] tested the extract of Rollinia occidentalis and pure solutions of two acetogenins (Rolliniastatin-1 and Motrilin) isolated from the extract as corrosion inhibitors for mild steel in 1.0 M HCl solutions. It was found that 1.0 g/L of the crude extract afforded corrosion inhibition of 79.7% while 0.007 g/L of Rolliniastatin-1 and Motrilin provided 72% and 59% protection respectively to the steel surface. Cai et al. [175] isolated amide alkaloids namely, piperine (CL1), piperanine (CL2), pipernonatine (CL3), and (2E,4E,10E)-N-11-(3,4-methylenedioxyphenylhmdecatrienoyl)-piperidine (CL4)) from a traditional Chinese medicine, Piper longum L. (Anguo, Hebei) and tested them as corrosion inhibitor for Cu in 1 M HCl solution. From the test, CL1, CL2, CL3, and CL4 were found to have 78.4%, 67.4%, 44.4%, and 18.5% respectively as inhibition efficiency. In a similar study, 1000 ppm of Momilactone A isolated from rice hulls (Oryza sativa L.) was found to exhibit 88% corrosion inhibition efficiency for mild steel in 1 M HCl solution [74]. Raja et al. [176] had equally identified 3β-isodihydrocadambine as the most potent anticorrosive component in Neolamarckia cadamba bark and leaves extracts. Liu et al. [177] deployed the liquid chromatography-mass spectrometry to characterize ginger extract and confirmed 6-Gingerol, 8-Gingerol, Cyclocurcumin, 1-hydroxy-1, 5-bis (4- hydroxy- 3-methoxyphenyl) pentan-3-one, 1-hydroxy-1, 5-bis (4- hydroxy-3, 5- dimethoxyphenyl) pentan- 3- on, and Arginine as the main phenolic compounds of ginger extract. Fig. 2 presents some of the active components isolated from various plant parts and tested for anticorrosive property for metals in HCl medium.

Table 1
Plant extracts investigated as metals corrosion inhibitor in HCl medium at pickling temperatures.

S/N	Plant name	Part of plant	Substrate studied	Acid concentration studied (M)	Highest inhibition efficiency (%)	Extract conc. with the highest efficiency	Inhibitor type	Extracting solvent	Reference
1	*Punica granatum* Linne	Husk	Q235A steel	1.0	95.0	1000 mg/L	Mixed	Water; Alcohol	[66]
2	*Musa paradisica* (Banana)	Peel	Mild steel	1.0	90.0 for raw; 80 for ripe; 78 for over ripe	300 mg/L	Anodic	Water-acetone	[67]
3	*Silybum marianum*	Leaf	304 stainless steel	1.0	96.4	1.0 g/L	Mixed	Petroleum ether	[68]
4	Aloe Vera gel	–	Mild steel	1.0	92.6	200 ppm	Mixed	–	[69]
5	*Thymus vulgaris*	Leaf	304 stainless steel	1.0	62.2	2%	Mixed	–	[70]
6	Longan	Seed and peel	Mild steel	0.5	92.4	600 mg/L	Mixed	Water	[42]
7	Lychee	Seed and peel	Mild steel	0.5	98.0	600 mg/L	Mixed	Ethanol; Water	[43]
8	*Euphorbia falcata*	Fruit	Carbon steel	1.0	93.2	3.0 g/L	Mixed	Methanol	[71]
9	*Aniba rosaeodora*	wood	C38 steel	1.0	95.3	200 mg/L	Mixed	Acid-base	[72]
10	*Rollinia occidentalis*	Fruit	Mild steel	1.0	85.7	1.0 g/L	Mixed	Methanol	[73]
11	*Oryza sativa* L.	Hull	Mild steel	1.0	88.0	1000 ppm	Mixed	Methanol	[74]
12	Egyptian licorice	Powder	Copper	0.1	86.8	8.0% (v/v)	Mixed	Ethanol/Water	[31]
13	Orange (*Citrus sinensis*)	Peel	Carbon steel	0.1	95.0	10.0 %	Mixed	Ethanol/Water	[30]
14	*Capsella bursa-pastoris*	Fruit	Q235 carbon steel	1.0	97.0	60 mg/L	Mixed	Ethyl alcohol	[75]
15	*Pisum sativum*	Peel	Mild steel	1.0	91.0	400 mg/L	Mixed	Water	[76]
16	*Eleusine aegyptiaca and Croton rottleri*	Leaf	Cast iron	1.0	91.3 for *E. aegyptiaca* and 94.5 for *C. rottleri*	2400 ppm	Mixed	Methanol	[77]
17	*Retama monosperma* (L.) Boiss	Seed	Carbon steel	1.0	94.4	400 mg/L	Mixed but with predominant effect on the cathodic reactions	–	[78]
18	*Pimenta dioica*	Leaf	Mild steel	0.5 and 1.0	97.4 in 0.5 M and 95.0 in 1.0 M	4% (v/v)	Mixed	Ethanol	[44]
19	Argan	Oil	C38 steel	1.0	81.0	3 g/L	Mixed	–	[79]
20	Watermelon	Rind	Mild steel	1.0	90.2	1000 ppm	Mixed	HCl	[80]
21	*Ligularia fischeri*	Whole plant	Mild steel	1.0	92.0	500 ppm	Mixed	Water	[81]
22	*Tinospora crispa*	Stem	Mild steel	1.0	87.9 for water extract and 83.9 for acetone-water extract	800 ppm for water extract and 1000 ppm for acetone-water extract	Anodic	Water; Acetone	[82]
23	*Lycium shawii, Teucrium oliverianum, Ochradenus baccatus, Anvillea garcinii, Cassia italica, Artemisia sieberi, Carthamus tinctorius, and Tripleurospermum auriculatum*	Fruit	Mild steel	0.5	62.7 – 91.0	0.01 g/100 ml	Mixed	Chloroform and alcohol	[45]
24	*Ficus hispida*	Leaf	Mild steel	1.0	90.0	250 ppm	Mixed	Absolute ethanol	[83]
25	*Gentiana olivieri*	Fruit	Mild steel	0.5	93.7	800 mg/L	Mixed	Methanol	[46]
26	*Nicotiana tabacum*	Leaf	Aluminium alloy AA3003	0.1	84.8	1200 mg/L	Mixed	Ethanol	[32]
27	Watermelon	Waste	Mild steel	1.0	83.67	2 g/L	Mixed	Boiling water	[84]
28	*Diospyros kaki* (Persimmon)	Leaf	St37 steel	0.1	91.0	225 ppm	Cathodic	Ethanol	[33]
29	*Diospyros kaki* L.	Leaf	Q235A steel	1.0	94.3	1000 mg/L	Mixed	Water; Alcohol	[85]
30	*Tragia plukenetii*	Leaf	Mild steel	1.0	88.0	500 ppm	Mixed	Methanol	[86]
31	*Reutera lutea* (Desf.) Maire	Aerial parts	Carbon steel	1.0	85.0	800 mg/L	Mixed	Methylene dichloride; Butanol	[87]
32	*Ptychotis verticillata*	Aerial parts	Mild steel	1.0	75; 86,	0.5 g/L	Mixed	Diethyl ether; Ethyl acetate	[88]
33	Pelargonium	Oil	Mild steel	1.0	90.6	4 mL/L	Mixed	Water vapour	[89]
34	*Ruta chalepensis*	Aerial parts	Carbon steel	1.0	87.0	3.5 g/L	Mixed	Ethanol	[90]
35	*Blighia sapida*	Seed	Mild steel	2.0	86.9	0.5 g/L	–	Water	[170]
36	*Olea europaea* L.	Root; Stem; Leaf	Mild steel	1.0	89.2 for root, 88.8 for stem, and 89.8 for leaf	0.000014 mL/ L	Mixed	Methanol; Water	[91]
37	*Chrysophyllum albidum*	–	Mild steel	1.0	96.2	1000 mg/L	Mixed	n-hexane	[92]

Table 1 (Continued)

S/N	Plant name	Part of plant	Substrate studied	Acid concentration studied (M)	Highest inhibition efficiency (%)	Extract conc. with the highest efficiency	Inhibitor type	Extracting solvent	Reference
38	*Saccocalyx satureioides*	Aerial parts	Carbon steel (X52)	1.0	86.8	900 mg/L	Mixed	Boiling methyl alcohol (70 %)	[93]
39	*Tilia cordata*	Leaf	Carbon steel	1.0	96.0	300 mg/L	Mixed	Water	[94]
40	*Phoenix dactylifera*	Leaf	Aluminum and aluminum-silicon alloy	0.5	89.1 for Al and 91.8 for Al-Si alloy	1000 ppm	Mixed	Petroleum ether; Methanol	[47]
41	Egyptian *Schinus_terebinthifolius*	Aerial parts	Carbon steel	0.5	93.3	900 ppm	Mixed	Methanol	[48]
42	*Juglans regia* L.	Leaf	Stainless steel; Aluminum	1.0	Ethyl acetate and water extracts exhibited 50.2 for 64.5 for stainless steel; 77.1 and 88.8 for Al respectively	0.14 mg/mL	Mixed	Water; Ethyl acetate	[95]
43	*Psidium guajava*	Leaf	Mild steel	0.5 N, 1 N, 1.5 N, 2 N	Water extract: 10 ml = 87.8 in 0.5 N; 8 ml = 97.1 in 1.0 N; 10 ml = 81.6 in 1.5 N; 8 ml = 56.2. Ethanol extract: 10 ml = 94.9 in 0.5 N; 10 ml = 93.8 in 1.0 N; 10 ml = 80.9 in 1.5 N; 10 ml = 77.2.	–	Mixed	Water; Ethanol	[49]
44	Olive	Mill wastewater	C35E carbon steel	1.0	92.9	4.0 g/L	Mixed	Ethyl acetate	[96]
45	*Mentha rotundifolia*	Leaf	Steel	1.0	92.9	35% (v/v)	Mixed	HCl	[97]
46	*Marrubium vulgare*L.	Fruit	C38 steel	1.0	86.5	8.0 g/L	Mixed	HCl	[98]
47	*Commiphora pedunculata*	Gum	Aluminum alloy AA 3001	0.1	73.0	0.5 g/L	–	–	[34]
48	Roselle	Aerial parts	Carbon steel	0.5	91.0	500 ppm	Mixed	Petroleum ether; Methanol	[50]
49	Quince	Pulp	Carbon steel	1.0	88.0	0.5 g/L	Mixed	Water; Cold acetone	[99]
50	*Argemone Mexicana*	Root	Mild steel	1.0	94.0	400 mg/L	Mixed	Water	[100]
51	Tender arecanut	Seed	Aluminum	0.5	94.4	4.5 g/L	Mixed	Water	[51]
52	Verbena	Oil	C38 steel	1.0	85.5	5.0 g/L	Mixed	Water	[101]
53	*Petersianthus macrocarpus*	Leaf	Mild steel	1.0	93.5	1000 mg/L	Mixed	Water	[102]
54	*M. piperita; M. pulegium*	Oil	Mild steel	1.0	86.0 for *M. piperita*; 90.0 for *M. pulegium*	1 g/L	Mixed	Water	[103]
55	*Nigella sativa*	Seed	Mild steel	0.5	74.0	0.4 g/L	Mixed	HCl	[52]
56	*Asparagus Racemosus*	–	Aluminum	1.0	72.2	4000 ppm	–	–	[104]
57	*Diospyros Kaki* L.f	Husk	Q235A steel	1.0	65.1	1000 mg/L	Mixed	Water; Alcohol	[105]
58	*Acalypha torta*	Leaf	Mild steel	1.0	91.0	1000 ppm	Mixed	Chloroform – methanol (2:1)	[106]
59	*Momordica charantia*	Fruit	Carbon steel	2.0	99.4	34.6 g/L	Mixed	Water; Ethanol	[171]
60	UAE *Rhazya stricta* Decne	Leaf	Mild steel	1.0	87.5	2.0 g/L	–	Water	[107]
61	*Andrographis paniculata*	Leaf	Low carbon steel	0.1	96.3	1 g/L	Anodic	Water	[35]
62	*Sesbania grandiflora*	Leaf	Mild steel	1.0	98.0	10,000 ppm	Anodic	Methanol	[108]
63	Date palm (*Phoenix dactylifera*)	Leaf	Carbon steel	1.0	88.1	2 g/L	Mixed	HCl	[109]
64	*Gloriosa superba*	Gum	Aluminum	0.1	84.9	0.5 g/L	–	Water; Ethanol	[36]
65	*Anthemis pseudocotula*	Whole plant	Mild steel	1.0	94.8	0.446 mM Luteolin-7-O-b-D-glucoside	Mixed	Methanol	[110]
66	*Terminalia chebula*	Fruit	Mild steel	1.0	88.9	800 mg/L	Mixed	–	[111]
67	Ginger	Root	Carbon steel	1.0	94.0	200 ppm	Mixed	Methanol	[112]
68	*Matricaria recutita*	Flower	Mild steel	1.0	93.3	7.2 g/L	Anodic	Water	[113]
69	*Kola nitida*	Seed	Mild steel	0.1	94.2	1200 mg/L	Mixed	Ethanol	[37]
70	Nettle (*Urtica dioica* L.)	Leaf & Stem	Mild steel	1.0	92.2	0.3 g/L	Anodic	Water	[114]
71	Safflower (*Carthamus tinctorius*)	–	Mild steel	1.0	94.2	0.14 g/L	Mixed	Water	[115]
72	*Aloe vera*	Leaf	Mild steel	0.5	97.1	2.5% (v/v)	Mixed	Methanol	[53]
73	*Tectona grandis*	Leaf	Stainless steel 304 (SS304)	2.0	71.7	1 g/L	Mixed	Ethanol	[172]
74	Atlas cedar (*Cedrus atlantica*)	Oil	S300 steel	1.0	88.0	1 g/L	Mixed	Diethyl ether	[116]

Table 1 (Continued)

S/N	Plant name	Part of plant	Substrate studied	Acid concentration studied (M)	Highest inhibition efficiency (%)	Extract conc. with the highest efficiency	Inhibitor type	Extracting solvent	Reference
75	*Opuntia-Ficus Indica*	Fruit	C38 steel	1.0	91.0	5 g/L	Mixed	–	[117]
76	*Cleome droserifolia* (Samwah)	–	Mild steel	1.0	91.1	300 ppm	Mixed	Methanol	[118]
77	*Ziziphus mauritiana*	Fruit	Aluminum; Copper	0.5 N	76.8 for Al; 88.6 for Cu	1.288 g/L	–	ethyl alcohol	[54]
78	*Ochrosia oppositifolia*	Leaf; Stem	Mild steel	1.0	94 for stem extract and 87 for leaves extract	25 mg/L	Mixed	Dichloromethane	[119]
79	*Michelia alba*	Leaf	Industrial pure iron; 303 stainless steel; Q235 carbon steel	1.0	96.7 for industrial pure iron; 98.1 for 303 stainless steel; 98.4 for Q235 carbon steel	1.686 g/L	Mixed	Water	[120]
80	Red apple (*Malus domestica*)	Fruit	Mild steel	0.5	90.0	5.0 g/L	Mixed	HCl	[55]
81	Black tea	Leaf	Mild steel	0.1, 0.5, 1.0	97 for 0.1 M; 92 for 0.5 M; 87 for 1 M	10 drops	–	Water	[56]
82	Opuntia	Stem	Mild steel	2.0	95.2	1000 ppm	Mixed	–	[173]
83	Cherry	Stem	Mild steel	1.0	89.5	0.005 g/L	–	Water	[121]
84	Aloes	Leaf	Mild steel	1.0	77.0	800 mg/L	Mixed	Water	[122]
85	Punica	–	Carbon steel	1.0	91.4	300 ppm	Mixed	Water	[123]
86	*Senna cana* (Nees & Mart.) HS Irwin & Barneby, *Byrsonima sericea* DC, *Dimorphandra gardneriana* Tul., *Mangifera indica* L., *Zanthoxylum syncarpum* Tull.	Leaf; Branch.	1020 carbon steel	0.1	Between 80.8 – 82.2	0.041 – 0.058 g/L	Mixed	Ethanol	[38]
87	*Cucumis sativus* (cucumber)	Peel	Carbon steel	1.0	82.7	50% (v/v)	Mixed	HCl	[124]
88	Jasmine tea	Leaf	Carbon steel	1.0	96.1	1 g/L	Mixed	HCl	[125]
89	*Launaea nudicaulis*	Whole plant	Mild steel	1.0	92.5 for methanolic extract; 87.2 for acidic extract	400 ppm	Mixed	Methanol; HCl	[126]
90	*Atropa belladonna*	Aerial parts	Carbon steel	1.0	96.6	500 ppm	Mixed	Petroleum ether; Methanol	[127]
91	*Ephedra alata*	Leaf	Mild steel	1.0 N	86.9	1.65 % v/v	Mixed	HCl	[128]
92	*Salvadora persica*	Leaf, Roots, Fruits	Low chromium-molybdenum steel (ASTM A213) grade T2 (0.5Cr-0.5 Mo)	1.0	94.4	500 ppm	Mixed	Water	[129]
93	*Juniperus procera*	Leaf	Carbon steel	1.0	88.8	2.5 g/L	Mixed	HCl	[130]
94	*Tiliacora accuminata*	Leaf	Mild steel	1.0	93.0	320 ppm	Mixed	Absolute ethanol	[131]
95	*Ocimum basilicum* L.	Oil	Aluminum	0.5	78.4	5.7 g/L	Cathodic	Ethanol	[57]
96	*Pistacia atlantica*	Gals	X52 steel	1.0	97.1	10 ppm	Cathodic	Acetone: water: 7/3 v/v	[132]
97	Garlic	Oil	Carbon steel	1.0	95.8	2.5 g/L	Mixed	Water	[133]
98	Brown onion	Peel	Carbon steel	1.0	94.0	300 mg/L	Mixed	Water	[134]
99	*Asteriscus Imbricatus*	Leaf	C38 steel	1.0	96.3	0.6 g/L	Mixed	Water	[135]
100	*Strychnos nux-vomica*	Seed	Carbon steel	4%; 8%	91.1 in 4% and 88.1 in 8%	2.0 g/L	Mixed	Methonol	[58]
101	*Camellia sinensis* (Green tea)	Leaf	Aluminum	0.5	90.5	100% (v/v)	–	Sulphuric acid	[59]
102	*Avicennia marina*	Leaf	Zinc	0.5	83.7	500 ppm	Mixed	Water	[60]
103	*Alstonia angustifolia* var. *latifolia*	Leaf	Mild steel	1.0	88.0	5 mg/L	Mixed	NH₄OH; CH₂Cl₂	[136]
104	Akee apple (*Blighia sapida*)	Seed	Mild steel	2.0	86.9	0.5 g/L	–	Ethanol	[174]
105	*Luffa aegyptiaca*	Leaf	Mild steel	1.0	96.6	0.28% (v/v)	Mixed	Methanol	[137]
106	*Chlorophytum borivilianum*	Root	Mild steel	1.0	91.0	500 mg/L	Mixed	Water	[138]
107	*Funtumia elastic*	Leaf	Q235 mild steel	1.0	90.5	600 ppm	Mixed	Water	[139]
108	Sunflower	Seed hull	Mild steel	1.0	98.0	400 ppm	–	Water; Ethanol	[140]
109	*Glycyrrhiza glabra*	Leaf	Mild steel	1.0	88.0	800 ppm	Mixed	–	[141]
110	*Kola nitida*	Seed	Aluminium alloy AA3003	0.1	85.0	1200 mg/L	Mixed	–	[39]
111	*Thymus algeriensis*	Leaf; Seed	2024 aluminium alloy	1.0	78.7	0.75 g/L	–	HCl	[142]

Table 1 (*Continued*)

| S/N	Plant name	Part of plant	Substrate studied	Acid concentration studied (M)	Highest inhibition efficiency (%)	Extract conc. with the highest efficiency	Inhibitor type	Extracting solvent	Reference
112	*Phyllanthus amarus*	Leaf	Mild steel	1.0	96.1	4% (v/v)	Mixed	Ethanol	[143]
113	*Mentha Pulegium*	Oil	Carbon steel	1.0	81.0	3 mL/L	Mixed	Water	[144]
114	*Opuntia ficus indica*	Fruit	Mild steel	1.0	91.0	5 g/L	Mixed	–	[145]
115	*Eugenia caryophyllata*	Flower	A60 carbon steel	1.0	91.1	1 g/L	Mixed	Water	[146]
116	*Morinda tinctoria*	Leaf	Mild steel	1.0	Aqueous extract = 96.0; HCl extract = 81.4	12% (v/v) for aqueous extract; 15% (v/v) for HCl extract	Mixed	Water; HCl	[147]
117	*Ananas sativum*	Leaf	Aluminum	0.1	92.5	0.5 g/L	–	Ethanol	[40]
118	*Luffa aegyptiaca*	Leaf	Mild steel	1.0	97.2	0.24 g/L	Mixed	Methanol	[148]
119	*Ricinus communis*	Bark powder	AISI 1020 carbon steel	0.5	83.0	1.77 g/L	Mixed	–	[61]
120	*Theobroma Cacao*	Bark	ASTM 1020 carbon steel	18.23 g/L	89.0	1.112 g/L	Cathodic	Ethanol	[62]
121	Mango and orange	Peel	Carbon steel	1.0	97.0 for Mango extract; 95.0 for Orange extract.	400 mg/L	Mixed	Hexane, ethyl acetate, and ethanol	[149]
122	*Coffea canephora* (coffee)	Seed	Carbon steel	1.0	94.0	1000 mg/L	Mixed but with predominate cathodic character	Water	[150]
123	Turmeric (*Curcuma longa*)	Rhizome	Mild steel	1.0	92.0	0.03 g/L	Anodic	Water	[151]
124	Date palm (*Phoenix dactylifera*)	Seed	Mild steel	1.0	86.8	2.5 g/L	Mixed	–	[152]
125	*Mentha Spicata*	Oil	Aluminum	1.0	83.0	1800 ppm	Mixed	Water	[153]
126	Olive	Oil	C38 steel	1.0	Phenolic (OOMW-Ph) fraction = 88.9; Non-phenolic (OOMW-NPh) fraction = 89.1	0.05 mL/L of OOMW-Ph and 0.006 mL/L OOMW-NPh	Mixed	–	[154]
127	Cashew nut testa tannin	Nut	Aluminum	0.1; 0.5; 2.0	Between 50.0 – 80.7	0.5 g/L	–	Acetone	[63]
128	*Mentha Pulegium*	Aerial parts	Carbon steel	1.0	88.3	33% (v/v)	Mixed	HCl	[155]
129	Lupine	–	Aluminum	0.1	71.4	100 ppm	Anodic	Water	[41]
130	Linseed	Oil	Carbon steel	1.0	98.2	3 g/L	Mixed	–	[156]
131	Pawpaw	Leaf	Mild steel	1.0	83.0	1.0 g/L	–	–	[157]
132	*Psidium guajava*	Seed	Aluminum	0.5 N	85.7	1.206 g/L	–	Ethanol	[64]
133	*Grewa Venusta* (Wild Jute Tree)	Root	Mild steel	1.0	97.9	8% (v/v)	Mixed	Ethanol	[158]
134	*Trigonellafoenum-graecum L*	Seed	Aluminum	1.0	86.6 for DCM extract, 84.41 for ethanolic extract, and 75.77 for pentane extract	1400 ppm	Mixed	Pentane; Dichloromethane (DCM); Ethanol	[159]
135	*Juniperus phoenicea* (Cupressaceae)	Oil	Mild steel	1.0	83.0	1500 ppm	Mixed	–	[160]
136	Ginkgo	Leaf	X70 steel	1.0	92.5	200 mg/L	Mixed	Alcohol	[161]
137	*Polygonatum odaratum*	Leaf	Aluminum	1.0	67.5	500 ppm	Mixed	Methanol	[162]
138	Black pepper	Seed	Pure Aluminum	1.0	99.7	0.243 g/L	Mixed	Methanol	[163]
139	Nettle (*Urtica dioica* L.)	Whole plant	Mild steel	0.5	97.0	0.0015 g/L	Mixed	Water	[65]
140	*Mentha rotundifolia*	Oil	Carbon steel	1.0	89.0	2 mL/L	Mixed	Water	[164]
141	*Boscia Senegalensis*	Leaf	Mild steel	1.0	81.5	0.5 g/L	Mixed	Ethanol	[165]
142	*Piper nigrum* L.	–	Mild steel	1.0	97.0	6000 mg/L	–	Alcohol	[166]
143	*Schreabera swietenioids*	Leaf	Mild steel	1.0 N	92.7	15 ppm	Mixed	Water	[167]
145	Geissospermum laeve	Leaf	C38 steel	1.0	92.0	100 mg/L	Mixed	–	[168]
146	*Glycine max* leaves (GMLE), *Cuscuta reflexa roxb.* (CRRE) and *Spirogyra* algae (SGAE)	–	Mild steel	1.0	73.6 for GMLE, 81.9 for CRRE, and 94.0 for SGAE.	2 g/L	Mixed	–	[169]
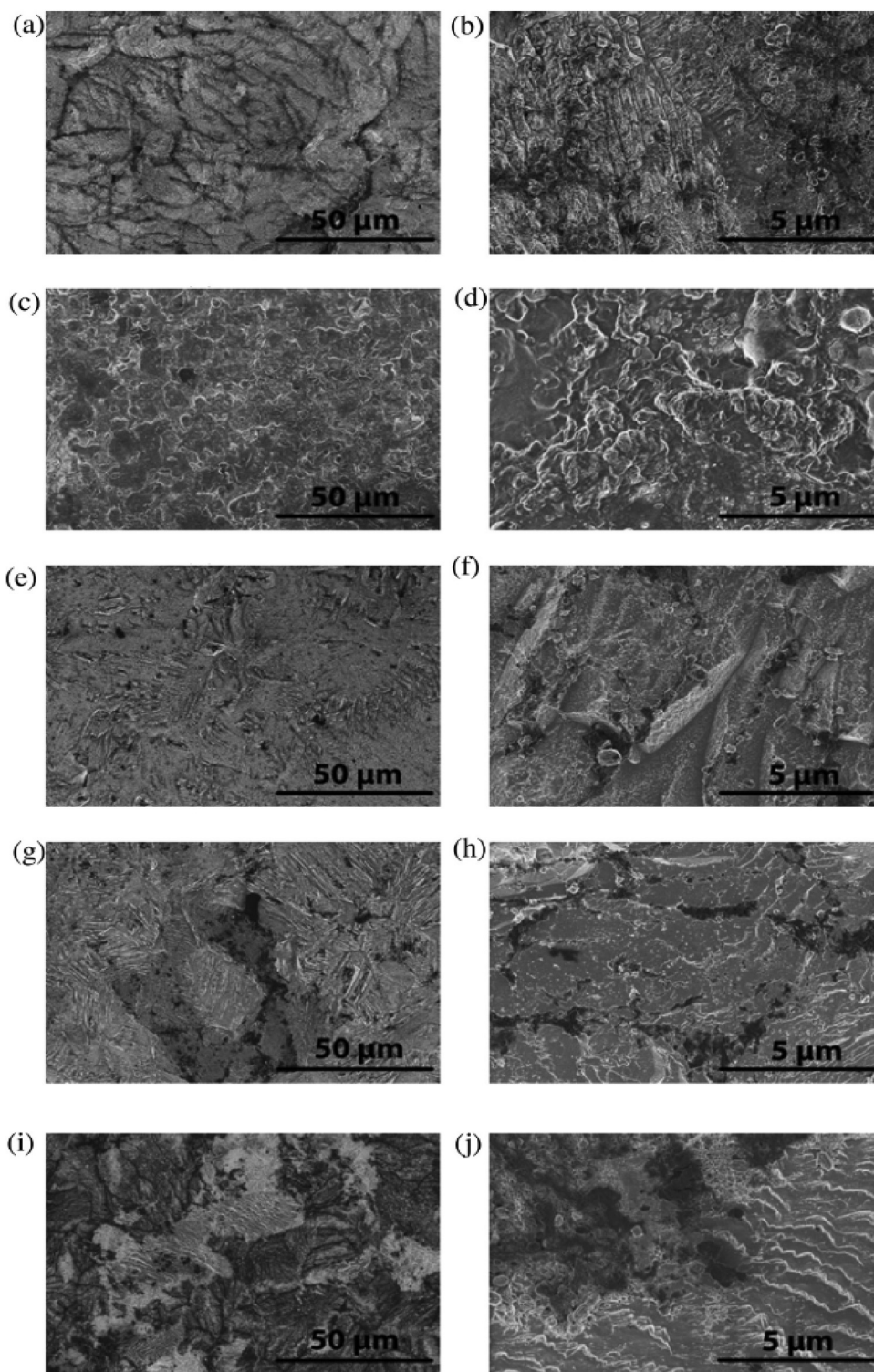
Fig. 1. SEM surface morphologies of steel immersed for 10 days in a water/ethanol solution containing 0.1 M HCl (a, b). Effects of the addition of orange peel extract 10% (c, d), and selected pure antioxidant compounds: neohesperidin 10^{-5} M (e,f), naringin 10^{-5} M (g, h) and ascorbic acid 10^{-5} M (i, j). Extracted with permission from M'hiri et al. [30]. Copyright 2015 Elsevier Ltd.

H₂SO₄ medium

Sulphuric acid is another industrial workhorse chemical like the hydrochloric acid. Compared with HCl acid, H_2SO_4 is cheaper. Primarily, H_2SO_4 acid is used for the manufacturing of phosphate fertilizers, calcium dihydrogenphosphate, and the ammonium

phosphates [178]. The second largest application of sulphuric acid is in metal processing like acid cleaning, pickling, as well as in the manufacture of copper and zinc. For oil well acidizing, the use of H_2SO_4 acid is minimal because of insoluble sulphate by-products with calcium and the fact that the acid might modify some oils to sludges [29]. The efficiency of H_2SO_4 acid as a pickling agent is
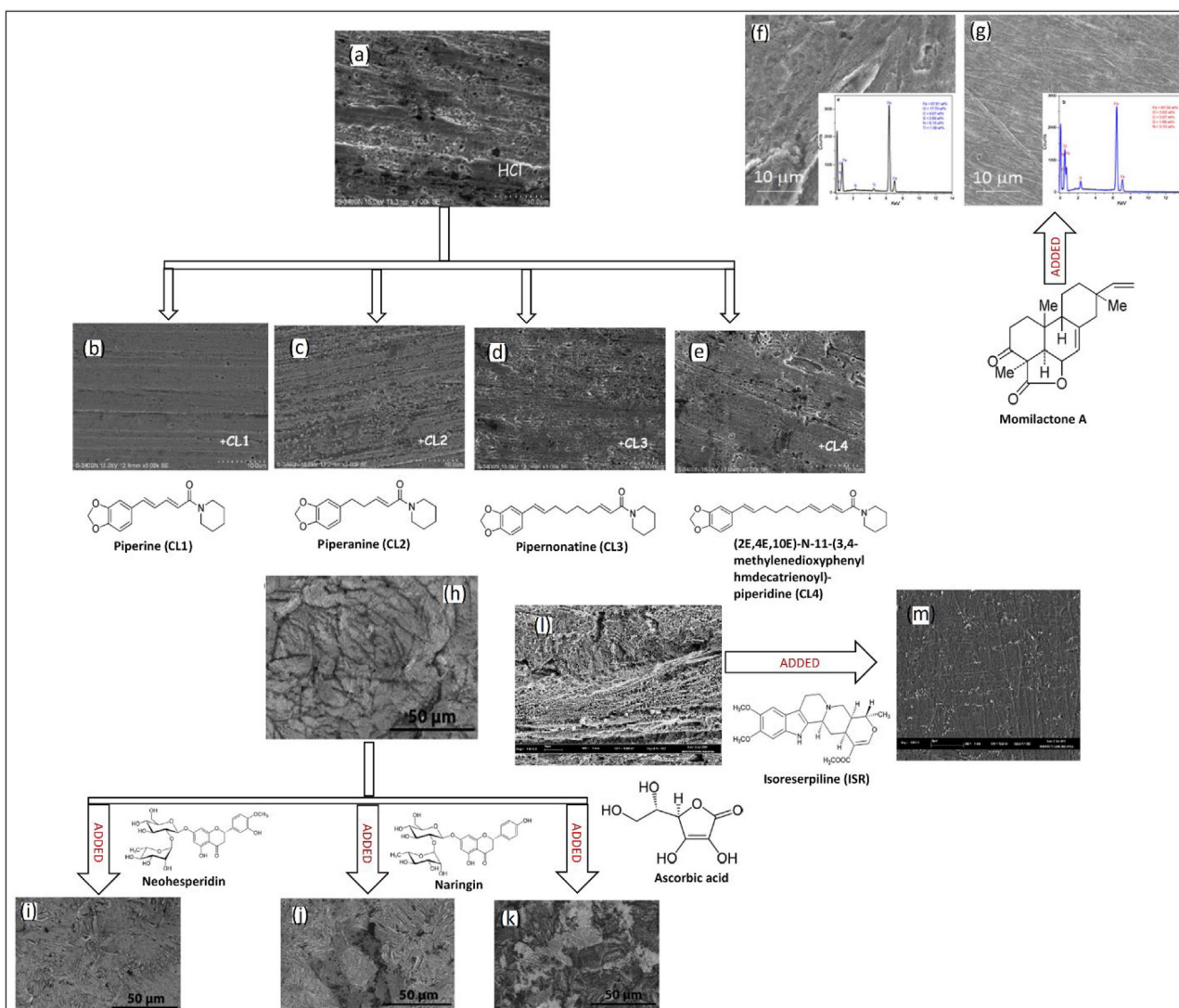
Fig. 2. Inhibitive effect of some active components isolated from various plant parts in HCl medium. The addition of 10^{-3} M piperine, piperanine, pipernonatine, and (2E, 4E, 10E)-N-11-(3, 4-methylenedioxyphenylhmdecatrienoyl)-piperidine to 1 M HCl solution inhibited copper corrosion by 78% (b), 67% (c), 44% (d), and 19% (e) respectively [175]. Inhibition efficiency of 88% (g) was achieved with 1000 ppm Momilactone A [74]. The presence of 10^{-5} M Neohesperidin, Naringin, and Ascorbic acid in 0.1 M HCl solution protected carbon steel against corrosion by 87% (i), 56% (j), and 92% (k) respectively [30]. Similarly, 25 mg/L Isoreserpiline inhibited mild steel corrosion in 1 M HCl solution by 93% (m) [119]. Figs. 2(a–e) were extracted with permission from Cai et al. [175] (Copyright 2013 American Chemical Society). Figs. 2(f, g) were reproduced with permission from Prabakran et al. [74] (Copyright The Korean Society of Industrial and Engineering Chemistry). Figs. 2(h–k) were reprinted from M'hiri et al. [30] (Copyright 2015 Elsevier Ltd). Figs. 2(l, m) were extracted with permission from Raja et al. [119] (Copyright 2012 Elsevier Ltd).

extremely temperature dependent as illustrated in Fig. 3. The acid concentration in the range of 5–10% is recommended for pickling [29]. As earlier mentioned, addition of corrosion inhibitor(s) to acid solution deploy for industrial application is essential to protect the metals used in such an environment. Within the years covered in this review, an appreciable number of scientific reports have been documented on the use of extracts from different plant parts as corrosion inhibitor in H_2SO_4 environment (Table 2). The concentration range covered includes 0.1 M [179,180], 0.25 M [181,182], 0.5 M [183–215],0.75 M [182], 1.0 M [183–235], 1.5 M [236], 2.0 M [237,238], and 5.0 M [239–242]. It is obvious from Table 2 that plant extracts are effective metals corrosion inhibitor in H_2SO_4 medium. However, some findings indicate that plants extracts are more effective in HCl solution than in H_2SO_4 environment. For example, Benahmed et al. [188] reported that 500 ppm Bupleurum lancifolium (Apiaceae) inhibited carbon steel (API 5L Gr B) corrosion in H_2SO_4 solution by 80.0% but 92.2% in HCl medium. Umoren et al. [243] equally demonstrated the inhibitive

effectiveness of strawberry fruit extracts respectively for mild steel in HCl medium than in H_2SO_4 solution (Fig. 4). These reports align with most results in the literature on the comparative studies of corrosion inhibition of organic species in HCl and H_2SO_4 solutions [244–246]. This clearly reflects the influence of chloride and sulphate anions on the adsorption process of organic inhibitors. As is it known, Cl^- ions are less hydrated than SO_4^{2-} ions [40,247] and has a stronger tendency to adsorb on metal surface compared to SO_4^{2-} ions. Hence, Cl- ions have the capacity to create excess negative charge toward solution phase than SO_4^{2-} ions and this favors Cl^- ions–organic species co-adsorption on metal surface [247,248].

We had in our previous review [249] highlighted the effect of certain parameters like inhibitor concentration, pH, and temperature on the adsorption behavior of organic species including plants extracts. Besides these parameters, another parameter studied recently is the influence of rotation speed on the corrosion
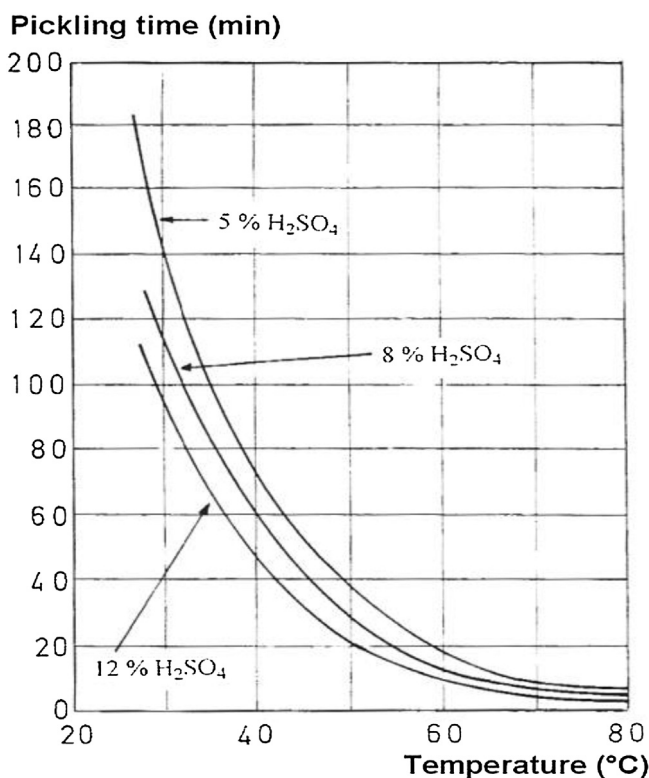
Pickling time (min)

Fig. 3. Plots showing the dependency of pickling on temperature. Extracted from Maanonen [29].

inhibition efficiency of plant extracts. The essence is to mimic a non-static environment like oil transportation pipelines. Lopes-Sesenes et al. [203] studied the effect of Buddleia perfoliata leaves extract on the corrosion behavior of carbon steel corrosion in 0.5 M H_2SO_4 acid using potentiodynamic polarization, electrochemical impedance spectroscopy, and electrochemical noise techniques. To established the influence of rotation speed on the components of Buddleia perfoliata leaves extract adsorbed on the metal surface, the authors conducted the experiments at different rotation speeds (0, 250, 500, 1000, and 2000 rpm). It was found that the corrosion of the metal was a function of rotation speed. The metal corrosion rate increased up to 500 rpm but thereafter decreased with a further increase in the rotation speed. In the system containing the plant extract, the lowest corrosion rate was obtained under static conditions. However, the corrosion rate varies directly with the rotation speed. In fact, the authors found that the inhibitor only remain on the metal surface for a few hours. As the rotation speed was increased, the adsorbed inhibitor species detached from the surface exposing active sites and making the substrate to be susceptible to localized type of corrosion.

Unlike in HCl medium, very few works have been devoted to the isolation of the active components in plants extracts for application in H_2SO_4 solution/corrosive environment. Prabakaran et al. [250] isolated β-sitosterol-3-O-b-D-glucoside (β-Sitosterol) from rice hulls and examined the inhibiting effect on the corrosion of mild steel in 1 M H_2SO_4 solution in the temperature range of 303–333 K. In ^{1}H NMR spectrum, β-Sitosterol displayed a single proton multiplet at δ = 3.515 and was assigned to a carbinol3 α-proton. In the ultra violet-visible spectrum, the isolated compound showed absorption bands at 212 nm and 240 nm which was due to π–π* transitions [251]. β-Sitosterol exhibited excellent anticorrosion property with 500 ppm protecting the metal surface by 95% at 303 K. The inhibition efficiency of β-Sitosterol varied directly with concentration but inversely with temperature (Fig. 5).

NaCl and Na_2SO_4 environments

Sodium chloride is a multipurpose salt and its use is almost endless. In the oil and gas industries, NaCl is an essential component of drilling fluids. It serves as a flocculant increasing the density of drilling fluid and bringing down downwell gas pressures [252]. During drilling, if salt formation is encounter, NaCl salt is often introduced into drilling fluids to saturate the solution and decrease or prevent dissolution within the salt stratum [253]. NaCl salt also gains application as a curing agent in cemented casings [254]. In the textiles and dyeing industries, this salt serves as a brine rinse to separate organic contaminants and enhance "salting out" of dyestuff precipitates. It also find usage in metals processing (specifically in aluminium, copper, beryllium, steel, and vanadium) and in the bleaching of wood pulp. Despite the versatility of NaCl salt, it is corrosive both in molten and in solution forms. In fact, corrosion of marine metallic structures, storage and cooling tanks, etc.is mainly cause by the presence of chloride ions, which facilitate initiation, and propagation of pits. Many research investigations have been devoted to metallic corrosion in NaCl environments [255–257]. Among the plant extracts studied as corrosion inhibitor in NaCl solution include Santolina chamaecyparissus [258], Azadirachta indica (neem) [259], Coptis chinensis [260], Mansoa alliacea [261], Laurus nobilis L. [262], and Prosopis juliflora [263]. Others are cactus mucilage and brown seaweed [264], Matricaria recutita chamomile [265], myrrh [266], Linum usitatissimum [267], Brassica campestris [268], Nettle [269,270], palm oil and corn Oil [271], Anise (Pimpinella anisum) [272], Caraway (Carum Carvi) [272], Cumin (Cuminum cyminum) [272], and Hibiscus (Hibiscus sabdarriffa) [272]. Laurus nobilis L. oil, Linum usitatissimum seeds, and Coptis chinensis root extracts were applied as inhibitor for AA5754 aluminium alloy, aluminium alloy AA2024, and aluminium 7075 alloy in 3% NaCl and 3.5% NaCl solutions respectively. The plant extracts were found to be adsorbed on the substrate surface and behaved predominantly as cathodic inhibitor retarding the chloride induced pitting corrosion above 80%. Santolina chamaecyparissus, Prosopis juliflora, cactus mucilage, brown seaweed, Matricaria recutita chamomile, Brassica campestris, palm oil and corn oil, as well as Nettle leaves extracts were effective corrosion inhibitor for various classes of steel in the considered aggressive environment. For Mansoa alliacea, the ethanolic extract was examined as corrosion inhibitor for zinc in 3% NaCl solution using electrochemical techniques. Mansoa alliacea extract acted as mixed-type inhibitor and gave inhibition up to 90% to the zinc surface. The extracts from A. indica (neem), Anise (Pimpinella anisum), Caraway (Carum carvi), Cumin (Cuminum cyminum), Hibiscus (Hibiscus sabdarriffa), and myrrh plant extracts were capable of suppressing the dissolution of copper in NaCl solutions. Fig. 6 and 7 summarize the corrosion inhibiting ability of plant extracts for the two most studied substrates – steel and aluminium respectively in NaCl environment.

Hot corrosion, an accelerated oxidation or sulfidation attack due to salt deposit (mostly solid or liquid mixed environments of NaCl and Na_2SO_4) has been widely studied [273–275]. It has been classified as type I and II [276,277,278]. Type I hot corrosion occur at operating temperatures higher than the melting point of the salt/acid mixture [276,277] whereas the type II which is more moderate than the type I takes place at temperatures below the melting temperature of the salt/acid mixture [276,278]. The study on possible application of plant part extracts as corrosion inhibitor has been extended to this type of corrosion. Chellouli et al. [279] studied the hot corrosion inhibition of iron by oil extracted from the seeds of Nigella sativa L. by means of potentiodynamic polarization, electrochemical impedance spectroscopy, and weight loss measurements. The composition of the aggressive

Table 2
Plant extracts studied as metals corrosion inhibitor in H_2SO_4 environment at pickling temperatures.

S/N	Plant name	Part of plant	Substrate studied	Acid concentration studied (M)	Highest inhibition efficiency (%)	Extract conc. with the highest efficiency	Inhibitor type	Extracting solvent	Reference
1	*Tagetes erecta* (Marigold flower)	Flower	Mild steel	0.5	96.1	1.0 g/L	Mixed	Ethanol	[183]
2	Coconut	Coir dust	Mild steel	0.5	94.3 for ethanolic extract; 41.7 for water extract	0.5 g/L	Mixed	Ethanol; Water	[184]
3	Aloe Vera	Leaf	Stainless steel	1.0	98.0	30% (v/v)	Mixed	Water	[216]
4	*Cryptostegia grandiflora*	Leaf	Mild steel	1.0	87.5	500 ppm	Mixed	Methanol	[217]
5	*Aster koraiensis*	Leaf	Mild steel	1.0	90.5	2000 ppm	Mixed	Methanol	[218]
6	*Sida acuta*	Leaf; Stem	Mild steel	1.0	85.0 for leaves extract; 52.0 for stem extract	0.5 g/L	–	Absolute ethanol	[219]
7	*Nauclea latifolia*	Leaf; Bark; Root	Mild steel	1.0	75.3 for bark extract; 91.6 for leaf extract; 94.3 for root extract	4.0 g/L	–	Absolute ethanol	[220]
8	*Lannea coromandelica*	Leaf	Mild steel	1.0	93.8	250 mg/L	Mixed	Water	[221]
9	Litchi (*Litchi chinensis*)	Peel	Mild steel	0.5	97.4	3.0 g/L	Mixed	Water	[185]
10	*Ficus Racemosa*	Stem	Mild steel	1.0	90.5	2500 ppm	Mixed	Methanol	[222]
11	*Sida cordifolia*	Leaf	Mild steel	0.5	99.0	500 mg/L	Mixed	Water	[186]
12	*Anacyclus pyrethrum* L. extracts	Leaf; Stem; Flower	Mild steel	0.5	87.0 for leaf and stem extract; 88.9 for flower extract; 79.3 for root extract	350 mg/L	Anodic	–	[187]
13	*Morinda tinctoria*	Leaf	Copper	0.25	52.6	15% (v/v)	Mixed	Water	[181]
14	*Bupleurum lancifolium* (Apiaceae)	Aerial parts	Carbon steel (API 5 L Gr B)	0.5	90.0	700 ppm	Mixed	Methyl alcohol	[188]
15	*Hyptis suaveolens*	Leaf	Mild steel	1.0	95.0	250 mg/L	Mixed	Ethanol	[223]
16	*Rothmannia longiflora*	Leaf	Mild steel	0.5	96.5	1000 mg/L	Mixed	–	[189]
17	*Thapsia villosa*	Aerial parts	Carbon steel	1.0	60.9 for EAE; 73.6 for BE	800 ppm	Mixed	Ethyl acetate (EAE); n-butanol (BE)	[224]
18	*Gongronema latifolium*	Leaf	Mild steel	5.0	94.2	10 g/L	–	Water	[239]
19	*Calotrpis procera; Calotropis gigantea*	Leaf; Latex; Fruit	Mild steel	0.5 N	*Calotropis procera:* Leaf 74.9; Latex 72.3; Fruit 79.8. *Calotropis gigantea:* Leaf 81.9; Latex 77.5; Fruit 79.8.	0.60% (v/v)	–	Ethanol	[190]
20	*Hemidesmus indicus*	Leaf	Mild steel	5.0	90.6	4.0 g/L	Mixed	Ethanol	[240]
21	*Syzygium cumini*	Seed	Mild steel	0.5	81	300 ppm	Mixed	Water	[191]
22	*Calotropis gigantiea* latex	Stem and leaf	Carbon steel	0.5	90.0	50 ppm	Mixed	–	[192]
23	*Hydroclathrus clathratus* marine alga	–	Mild steel	1.0	77.6	500 ppm	Mixed	Methanol	[225]
24	*Adhatoda vasica*	Leaf and flower	Mild steel	0.5	98.8	3.0 g/L	Mixed	Water	[193]
25	*Dodonaea viscosa*	Leaf	Mild steel	0.5	76.1	0.6 g/L	Mixed	H_2SO_4	[194]
26	*Opuntia ficus-indica*	Peel	Carbon steel	0.5	94.0	1000 ppm	Cathodic	Methanol	[195]
27	Barley	Whole plant	Steel	1.0	94.2	0.8 g/L	Mixed	Water	[226]
28	*Camellia sinensis* (green tea)	Leaf	Mild steel	0.5	90.4	100% (v/v)	–	Acid	[196]
29	*Acacia senegalensis*	Leaf	Aluminum alloy (Al–Si–Fe) reinforced with 15% SiC	0.5	92.7	8% (v/v)	Mixed	Water	[197]
30	*Morinda tinctoria*	Leaf	Aluminum	0.25; 0.5; 0.75	74.4 in 0.25 M; 79.1 in 0.5; 80.9 in 0.75	16% (v/v)	Mixed	Water	[182]
31	Mustard	Seed	X60 pipeline steel	1.0	76.0	1000 mg/L	Mixed	H_2SO_4	[227]
32	*Aframomum melegueta*	Seed	Mild steel	0.5	96.4	800 mg/L	Mixed	H_2SO_4	[198]
33	*Setaria verticillata*	Leaf	Mild steel	1.0	94.3	250 mg/L	Mixed	Water	[228]
34	*Luffa Aegyptiaca*	Leaf	Mild steel	0.5	93.3	0.24 g/L	Mixed	Methanol	[199]
35	*Commiphora caudate*	Leaf	Mild steel	2.0	67.0	0.7 mg/L	Mixed	H_2SO_4	[237]
36	*Tagetes erecta* (Marigold flower)	Flower	Mild steel	0.5	98.1	1.0 g/L	Mixed	Ethanol	[200]
37	*Morinda tinctoria*	Leaf	Mild steel	0.5	87.9 for acidic extract; 91.8 for aqueous extract.	40% (v/v) acid extract; 22% (v/v) aqueous extract	Mixed	Water	[201]

Table 2 (*Continued*)

S/N	Plant name	Part of plant	Substrate studied	Acid concentration studied (M)	Highest inhibition efficiency (%)	Extract conc. with the highest efficiency	Inhibitor type	Extracting solvent	Reference
38	*Wrightiatinctoria, Clerodendrumphlomidis, Ipomoeatriloba*	Leaf	Mild steel	0.5	97.0	200 ppm	Mixed	H_2SO_4	[202]
39	*Nauclea latifolia*	Root	Carbon steel	5.0	89.0	2.5 g/L	–	Ethanol	[241]
40	*Fenugreek*	Seed	Copper; Nickel	2.0	85.6 for Cu; 83.5 for Ni	20% (v/v)	Mixed	Water	[238]
41	*Buddleia perfoliata*	Leaf	Carbon steel	0.5	–	–	–	Methanol	[203]
42	*Phyllanthus fraternus*	Leaf	Mild steel	1.0; 5.0	85.8 in 1.0 M and 80.0 in 5.0 M	4.0 g/L	–	H_2SO_4	[229]
43	*Argemone mexicana*	Leaf	Mild steel	0.5	87.0	600 mg/L	Mixed	Water	[204]
44	*Eriobotrya japonica*	Leaf	Mild steel	0.5	42.0	250 ppm	Anodic	Water	[205]
45	Barley	Bark and non-germinated barley	Stainless steel AISI 304	1.5	92.9	4.0 g/L	Mixed	H_2SO_4	[236]
46	*Origanum Compactum*	Aerial parts	Carbon steel	0.5	93.3	3.0 g/L	Mixed	Water; Methanol	[206]
47	Rice	Husk ash	Mild steel	1.0	86.0	5.0% (v/v)	–	H_2SO_4	[230]
48	*Malus domestica* (MD); *Caesalpinia bonducella* (CB)	Leaf	Mild steel	0.5	78.0 for CB; 70.2 for MD	0.7 of CB; 0.25 of MD.		Water	[207]
49	*Gundelia tournefortii*	Leaf	Mild steel	1.0	90.0	150 ppm	Mixed	Methanol/ Water	[231]
50	*Houttuynia cordata*	Leaf	Mild steel	0.5	90.0	0.7 g g/L	Mixed	H_2SO_4	[208]
51	*Saraca ashoka*	Seed	Mild steel	0.5	95.5	100 mg/L	Mixed	Ethanol	[209]
52	*Acacia nilotica*	–	Mild steel	1.0 N	–	2% (v/v)	Mixed	Ethanol	[232]
53	*Myrtus Communis*	–	Copper	0.5	90.7	100 mg/L	Mixed	Ethanol	[210]
54	Date palm (*Phoenix dactylifera*)	Seed	Mild steel	0.5	84.1	1.5 g/L	Mixed	H_2SO_4	[152]
55	*Eichhornia crassipes*	Root	Mild steel	0.5	–	10% (v/v)	Mixed	H_2SO_4	[211]
56	*Ficus tricopoda*	Gum	Aluminum	0.1	85.2	0.5 g/L	–	–	[179]
57	*Ficus Benjamina*	Gum	Aluminum	0.1	83.7	0.5 g/L	–	–	[180]
58	*Phyllanthus muellerianus*	Leaf	Steel reinforced concrete	0.5	91.7	0.3% (v/v)	–	Methanol	[212]
59	*Polyalthia longifolia*	Leaf	Mild steel	0.5	82.7	0.4 g/L	–	Water	[213]
60	*Rotula aquatica*	Leaf; Stem; Root	Mild steel	1.0	79.7 by leaf extract; 70.3 by stem extract; 86.6 by root extract	4.0 g/L	Anodic	Ethanol	[233]
61	Black liquor was from pulp and paper industry	–	Mild steel	0.5	89.0	1000 ppm	Mixed	H_2SO_4	[214]
62	Thymus vulgaris (TYV); *Xylopia aethiopica* (XYA); *Zingiber officinale* (ZGO)	Leaf; Fruit; Root.	Mild steel	5.0	88.0 for TYV; 71.0 for XYA; 93.0 for ZGO	10.0 g/L	–	Ethanol	[242]
63	*Rhus verniciflua*	Leaf	Mild steel	1.0	93.0	500 ppm	Mixed	Methanol	[234]
64	*Acacia cyanophylla*	Leaf	Mild steel	1.0	84.0	200 ppm	Mixed	Water	[235]
65	Marjoram	Leaf	Mild steel	0.5	84.7	13% (v/v)	Mixed	Water	[215]

environment was as follows: 0.2 g/L Na_2SO_4, 0.2 g/L $NaHCO_3$ and 0.2 g/L NaCl. From the findings, the extract was adjudged a good mixed type inhibitor and an inhibition effectiveness of 99% was achieved with the extract concentration of 2500 ppm. Varvara et al. [280] examined the influence of various concentrations of propolis on bronze corrosion in corrosive electrolyte comprising of 0.2 g/L Na_2SO_4 and 0.2 g/LNaHCO$_3$, acidified to pH = 5 by addition of dilute H_2SO_4 at 20°C using multiscale electrochemical techniques. High performance liquid chromatography was used to identify the major constituents of propolis. Varvara et al. found that propolis presented good anticorrosive properties on bronze, behaving as a mixed-type inhibitor, but with protection efficacy being time-dependent. The best inhibiting efficiency was 98.9% and was achieved with 100 ppm propolis after about 12 h of immersion in the inhibited system. In 0.1 M Na_2SO_4 solutions with pH 7 and pH 4, Ramde et al. [281] assessed the corrosion inhibition of brass by natural Camellia sinensis extract by electrochemical and scanning electron microscopy techniques. The results obtained indicate that

Camellia sinensis extract is a very effective corrosion inhibitor for brass corrosion process in both the acidic and neutral media.

NaOH environment

Sodium hydroxide (caustic soda) is a strong base with numerous industrial applications. It is used in pulping of wood for making paper or regenerated fibers, digestion of tissues, dissolving amphoteric metals and compounds, as esterification and transesterification reagent, and even in food preparation [282]. It is equally deployed in alkaline cleansing and pickling [29]. Another important application of NaOH is in Al-alkaline solutions used in the formulation of air/metal batteries [283,284] and in nuclear water reactors [285]. Aluminum, which is usually the anode in the air/Al batteries [283,284], is known for its excellent formability, high electrical and thermal conductivity, a low weight, and high reflectivity [286]. It also exhibits good corrosion resistance when exposed to the atmosphere and many aqueous
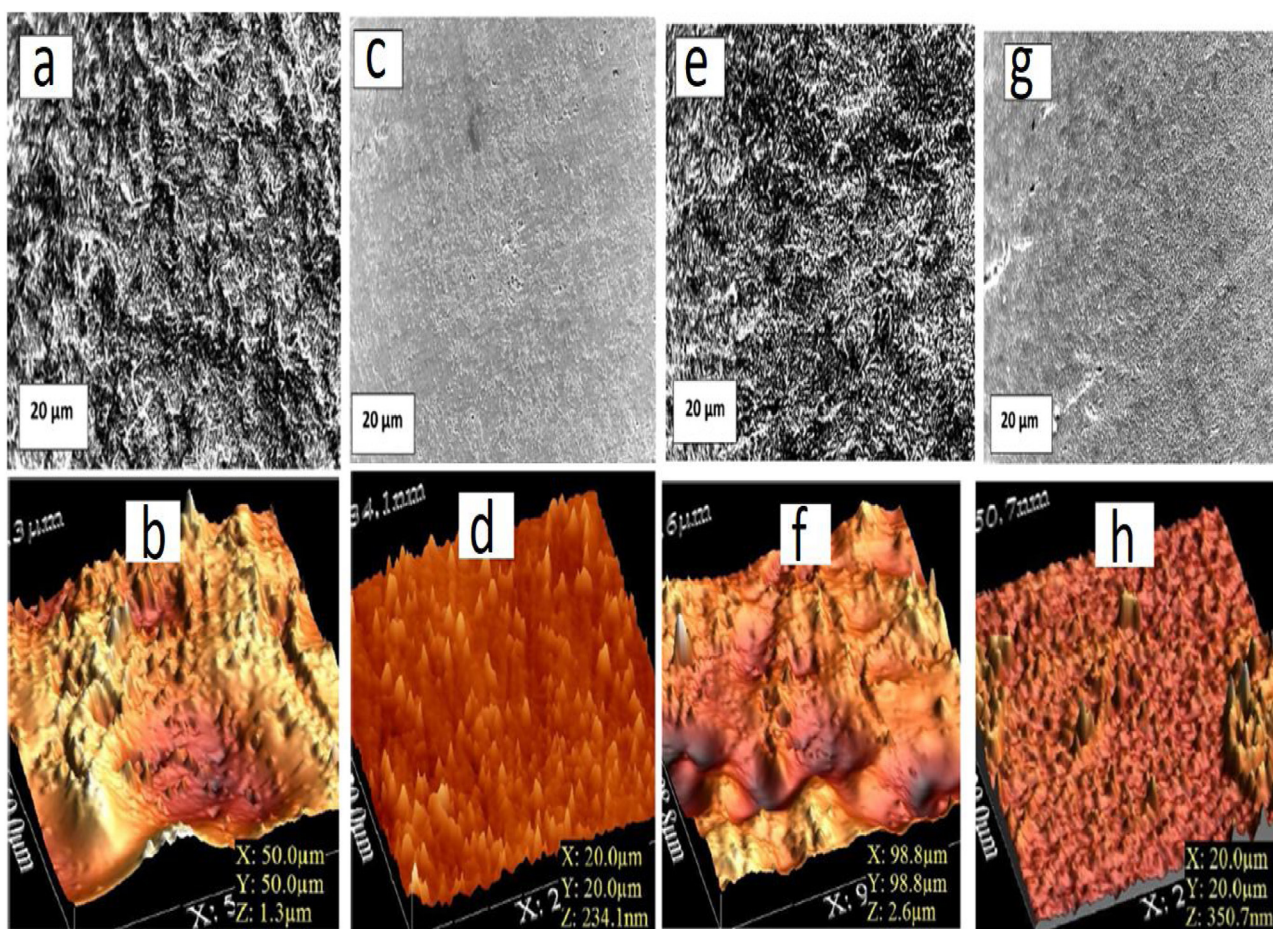
Fig. 4. Comparative results of plant extracts in HCl and H$_2$SO$_4$ environments; (a, b) 1 M HCl, (c) HCl + 2.0 g/L watermelon rind extract (WMRE), (d) HCl + 2.0 g/L strawberry fruit extract (SBFE), (e, f) 0.5 M H$_2$SO$_4$, (g) H$_2$SO$_4$ + 2.0 g/L WMRE, (h) H$_2$SO$_4$ + 2.0 g/L SBFE. In HCl medium, the corrosion inhibition performance of WMRE and SBFE was 83.3% [247] and 86.3% [243] but 77.3% [247] and 81.7% [243] respectively in H$_2$SO$_4$ environment. Figs. 4(a, c, e, & f) were extracted with permission from Odewunmi et al. [247] (Copyright 2014 The Korean Society of Industrial and Engineering Chemistry). Figs. 4(b, d, g, & h) were reproduced with permission from Umoren et al. [243] (Copyright Springer-Verlag Berlin Heidelberg 2014).

environments, due to the formation of a resistive oxide layer. Nevertheless, the oxide layer is destroyed when exposed to aggressive acidic and alkaline environments. In NaOH solution, protective oxide is dissolved by the OH$^-$ ions and the metal surface develops a negative potential [287–289]. The corroded Al surface produces hydrogen molecules and soluble species like aluminum hydroxide as the corrosion products [290,291]. According to Zhang and Jow [292] and Singh et al. [293], prevention of Al corrosion in air battery is a necessity as the metal corrosion could pose serious danger such as passivation of the cathode active material and increase of the electrical resistance by its solid products. It can also lead to the contamination of the electrolyte and increase in self-discharge rate by its soluble products, as well as reductive deposition of Al^{3+} ions in the counter anode. As a consequence of severe hydrogen gas liberation occasioned by Al corrosion, the practical use of Al/air battery despite the eco-friendliness and excellent energy density of this battery is minimal. No wonder keen attention is paid in recent times to the mitigation of Al corrosion in NaOH medium. Singh et al. [293] studied the inhibiting action of Piper longum seed extracts on the corrosion behavior of Al in 1 M NaOH solution using potentiodynamic polarization, electrochemical impedance spectroscopy, and weight loss methods. Based on the results reported by the authors, P. longum extract is an effective inhibitor being able to mitigate the metal dissolution up to 94% (400 mg/L concentration). The findings of Sing et al. was in conformity with the reports of Chaubey et al.

[294,295,296], Bataineh et al. [297], and Irshedat et al. [298]. According to Chaubey et al., Pisum sativum, Solanum tuberosum, Citrus reticulata, Kalmegh, and Neolamarkia Cadamba extracts can protect aluminium alloy surface above 80% in 1 M NaOH solution. For Bataineh et al. [297] and Irshedat et al. [298], 1.0% (v/v) Sinapis alba and 100% (v/v) Lupinus varius L. extracts respectively suppressed Al corrosion in 1 M NaOH medium by 97.98% and 93.73% at 298 K.

As contained in Awad et al. [299], the issue of the corrosion behavior of Al in NaOH solution has long been controversial. Some authors claimed that the corrosion rate is a linear function of NaOH concentration but the linearity does not apply at high concentrations. Others believed that, corrosion rate is proportional to the square root of NaOH concentration within the range of 0.03 M to 5 M. The argument that prompted Awad et al. to undertake a study into the corrosion behavior of Al in a wide range of NaOH concentration (0.001 M – 2.0 M) at 30 °C. Awad et al. had noted that the corrosion rate of Al was proportional to NaOH concentration within the range 0.001 M–0.005 M but almost proportional to the square root of concentration in the range 0.01 M–0.5 M. For concentrations higher than 0.5 M, the corrosion rate increased rapidly and was due to the formation of cathodic areas at the expense of the anodic ones. The recent investigation by Al-Moubaraki et al. [291] seems to verify Awad et al. findings. For instance, the corrosion rate obtained by Moubaraki et al. from weight loss measurements for Al in different concentrations was 1.560 g cm^{-2} min^{-1}, 2.798 g cm^{-2} min^{-1}, 4.156 g
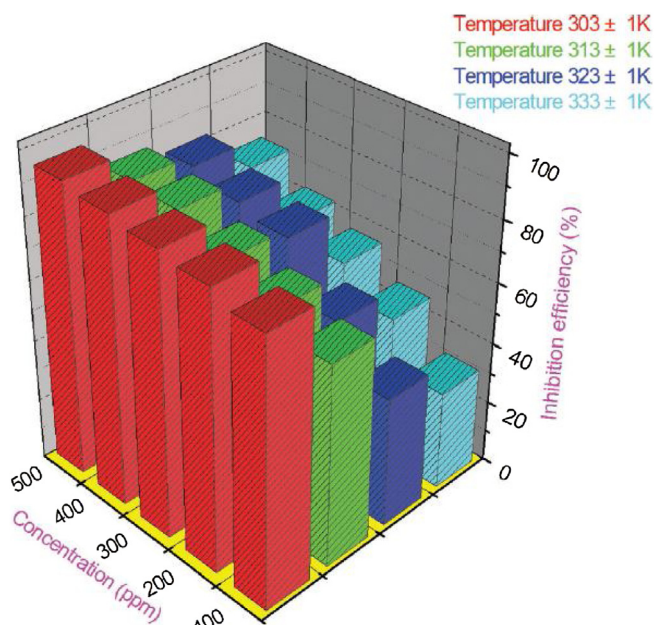
Fig. 5. The inhibition efficiency of β–sitosterol at different temperatures. Extracted with permission from Prabakaran et al. [250]. Copyright The Royal Society of Chemistry and the Centre National de la Recherche Scientifique 2017.

$cm^{-2} min^{-1}$, 9.007 g $cm^{-2} min^{-1}$, 14.368 g $cm^{-2} min^{-1}$, and 18.411 g $cm^{-2} min^{-1}$ for 0.025 M, 0.05 M, 0.10 M, 0.25 M, 0.50 M, and 1.00 M respectively. Moubaraki et al. decided to evaluate the role of an aqueous extract of celery (Apium graveolens L.) seeds (AECS) in inhibiting the corrosion of aluminum in the NaOH systems. It was found that at 0.50 g/L AECS, inhibition efficiency increased with increasing NaOH concentration up to 0.25 M and afterward declined. In 0.25 M NaOH solution, the authors found that, higher concentrations of AECS performed better and 1.50 g/L of the extract afforded the maximum inhibition efficiency of 93.33%. Fig. 8 illustrates the effectiveness of this concentration as inhibitor for Al in 0.25 M NaOH.

Copper is another metal in which the possibility of using plant parts extracts to control its corrosion has been examined. Cu and its alloys, because of their excellent corrosion resistance, ease of fabrication during installation, low installation cost, and friendliness to the natural environment are fast gaining preference in every aspect of industrial production as integral components in building construction, power generation and transmission, electronic products, industrial machinery and transportation, and predominantly in seawater system [300,301]. Nevertheless, the corrosion resistance property is lost when in long-term application in a corrosive environment. It was shown that utilizing 440 ppm of Capparis spinosa extract in 1.0 M NaOH solution where Cu is used can ensure 85% protection of the metal even at 45 °C [302]. Raghavendra and Bhat [303] also demonstrated that, mature areca nut husk (MAH) extract has the ability to protect Cu surface in 0.5 M NaOH medium up to 90.0%. MAH, which acted as a mixed type corrosion inhibitor, can as well be used as inhibitor for mild steel in 0.5 M NaOH solution and efficiency as high as 91.67% is guaranteed [303]. According to Etteyeb and Nóvoa [304], aqueous extracts of Eucalyptus globulus, Punica granatum and Olea europaea are effective corrosion inhibitors for steel bars in synthetic solution (0.1 mol/L NaOH + 0.5 mol/L NaCl).

Aqueous medium

Water, as often said, is life! It is very essential element and the human body is believed to contain up to 55% of water [305]. Beside the primary use of water, i.e drinking, water has numerous uses.

Water bodies serve as transportation medium. In industries, water is used in cooling systems, heat exchanger, air conditioning, in fossil and nuclear fuel steam plants, etc. High purity water is not corrosive. Water however becomes corrosive when dissolved salts and gases, bacteria, solid particles, and other contaminants are present. The presence of these contaminants in water bodies and water used for industrial purposes is somewhat unavoidable. Metals corrosion in water is a common problem. Plant parts extracts have been investigated as metals corrosion inhibitor in various kinds of water namely deuterium-depleted water (DD) [306], cooling water [307], well water [308], oil field water [309,310], rain water [311], saline formation water [312,313,314], seawater [315,316,317,318], and potable water [319]. In DD water, the effect of Citrus medica leaf on the corrosion of carbon steel was examined by mass loss measurements at different time. The leaf extract was noted to be an effective inhibitor with maximum inhibition efficiency of 81.58% at 300 ppm. The Citrus medica leaf extract was able to exert the corrosion inhibition due to interaction between the carboxylate anion and ring oxygen of the extract phytochemicals with the carbon surface. The interaction led to the formation Fe-Citrus medica leaf extract complex and this was confirmed by FTIR and UV–vis absorption results. Plants extracts are promising corrosion inhibitor in corrosive water environments. For instance, Mohammadi and Rahsepar [307] reported that 1000 ppm Mazuj gall extract provided corrosion inhibition efficiency of 94.3% and excellent scale inhibition efficiency of 97.2% to mild steel specimens exposed to simulated cooling water. 2.96 g/L and 5.25 g/L of Cucurbita and Ocimum respectively were sufficient for carbon steel to be protected by 82.02% and 77.78% at temperatures up to 40 °C [309]. Also, 5 g/L Ziziphus lotus (wild jujube) was capable of preventing copper corrosion in natural seawater by 93% [318]. From El-Taib Heakal et al. [312] report, very low concentration of 10 ppm aqueous cornflower (Centaurea cyanus) extract (CFE) mitigated carbon steel corrosion in harsh saline formation water by 69% and retained its inhibitive effect even at elevated temperatures. Tobacco rob extract was found to possess good anti-corrosion and anti-scaling properties for Q235 corrosion in artificial seawater [315]. Similarly, applying Seaweed extract on the corrosion of carbon steel in saline formation water [313], Neem extract on carbon steel API 5LX dissolution in hypersaline environments [314], and Psidium guajava leaf extract as corrosion inhibitors for double thermally-aged Al-Si-Mg (SSM-HPDC) alloy in simulated seawater [317] was a profitable venture as they were capable of suppressing the substrates deterioration above 80%. Nevertheless, there are some exceptions where plant extracts were found to moderately inhibited or poorly retarded metals corrosion in neutral medium. For example, Gerengi et al. [311] investigated the inhibition strength of mimosa extract on the corrosion of AA6060 aluminum alloy in acid rain solution deploying electrochemical impedance spectroscopy and dynamic electrochemical impedance spectroscopy techniques. It was found that the extract concentration as much as 2750 ppm could only afford 45% protection to the alloy surface. Deyab et al. [313] reported that, 400 ppm lemon grass (Cymbopogon citratus) extract (the optimum concentration) provided 60.1% corrosion inhibition to carbon steel in produced oilfield water at 293 K. Johnsirani et al. [316] observed that, 6 mL Eclipta alba extract produced 60% protection to carbon steel in sea water but upon combination with 25 ppm Zn^{2+} ions, 92% corrosion inhibition was accomplished. Factors such as the chemistry of the corrosive medium (hardness level, dissolved salts and gases, concentration of solids, bacteria etc.) and the concentration of the actual phytochemical responsible for corrosion inhibition of an extract may be the reason for the differences in the inhibiting behavior of plant extracts. In fact, the greatest challenge on the use of plant extract as metals corrosion inhibitor is the inability to clearly pinpoint the active component(s) that is actually responsible for corrosion inhibition.
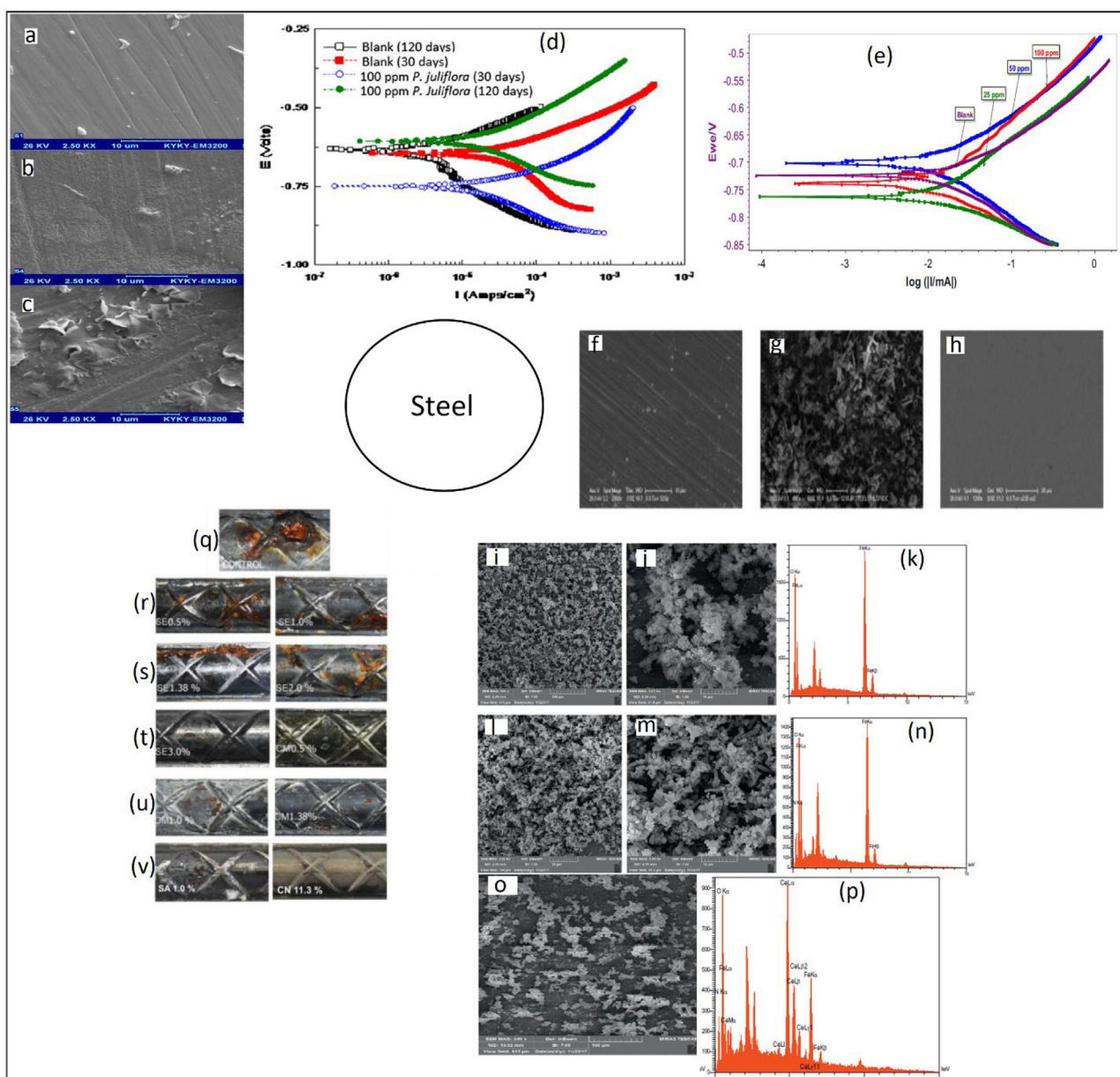
Fig. 6. The corrosion inhibiting strength of plant extracts for steel in NaCl environment; (a) freshly polished 304 stainless steel surface, (b) steel surface after 3 h immersion in 3.5% NaCl solution, (c) steel surface after 3 h immersion in 3.5% NaCl solution containing 1.0 g/L *Santolina chamaecyparissus* extract. The extract which acted as a mixed type corrosion inhibitor prevented the metal corrosion by 86.9% [258]. In Fig. 6(d) is shown the Tafel plots of embedded steel in concrete without and with *Prosopis juliflora* extract after 30 and 120 days. Similar plots but showing the performance of the different concentrations of *P. juliflora* extract after 30 days is shown in Fig. 6(e). The extract performed outstandingly affording inhibition efficiency of 91% after 120 days [263]. Figs. 6(f, g, & h) show the SEM micrographs of S235JR steel in abraded, after exposure to corrodent without and with *Matricaria reticutita* extract respectively. It was reported that 15% vol./vol of the extracted protected the steel surface from corrosion by 98.9% and this resulted in a smoother surface seen in Fig. Fig. 6(h) relative to Fig. 6(g) [265]. The extract from Nettle leaves was found to exhibit moderate corrosion inhibitive effect [269]. The FE-SEM images and EDAX spectra of steel specimens revealed that the surface morphology of the inhibited surface (l–n) was almost same as that of the unprotected (i–k). However, it was found that a mixture of 600 ppm Ce (III) ions with 200 ppm Nettle leaves extract significantly improved the performance (o, p) and 95% inhibition efficiency was reported [269]. The digital photographs taken from rebar active areas for various test solutions are displayed as Fig. 6(q–v). All control rebars presented larger pits compared to those in the organic additive solutions (CN = Cactus mucilage extract, CN = Calcium nitrite). Figs. 6(a–c) were extracted with permission from Shabani-Nooshabadi and Ghandchi [258] (Copyright 2015 The Korean Society of Industrial and Engineering Chemistry); (d) and (e) were reproduced from Palanisamy et al. [263] (Copyright Springer Science + Business Media Dordrecht 2016). Figs. 6(f–h) were adapted from Nasr et al. [265] (Copyright Emerald Publishing Limited) ; (i–p) were reprinted with permission from Ramezanzadeh et al. [269] (Copyrigt 2018 Elsevier B. V.); (q–v) were extracted from Hernández et al. [264] (Copyright 2017 Emerald Publishing Limited).

CO₂ environment

CO$_2$ corrosion otherwise called sweet corrosion is a serious corrosion problem in the oil and gas industry. It is estimated that approximately 60% of oilfield failures are related to sweet corrosion [320–322]. The source of CO$_2$ in the oilfield is both natural and anthropogenic [322,323]. Besides the natural presence in hydrocarbons formation, CO$_2$ in combination with hot water is usually injected into oil wells to enhance oil recovery [322]. However, CO$_2$ particularly when in high concentration dissolves in the water to form carbonic acid (H$_2$CO$_3$) thus posing corrosion challenge to metallic tubings and pipelines. The use of chemical corrosion inhibitors (mostly as amines and imidazolines and their derivatives) is one of the low cost techniques for abating sweet
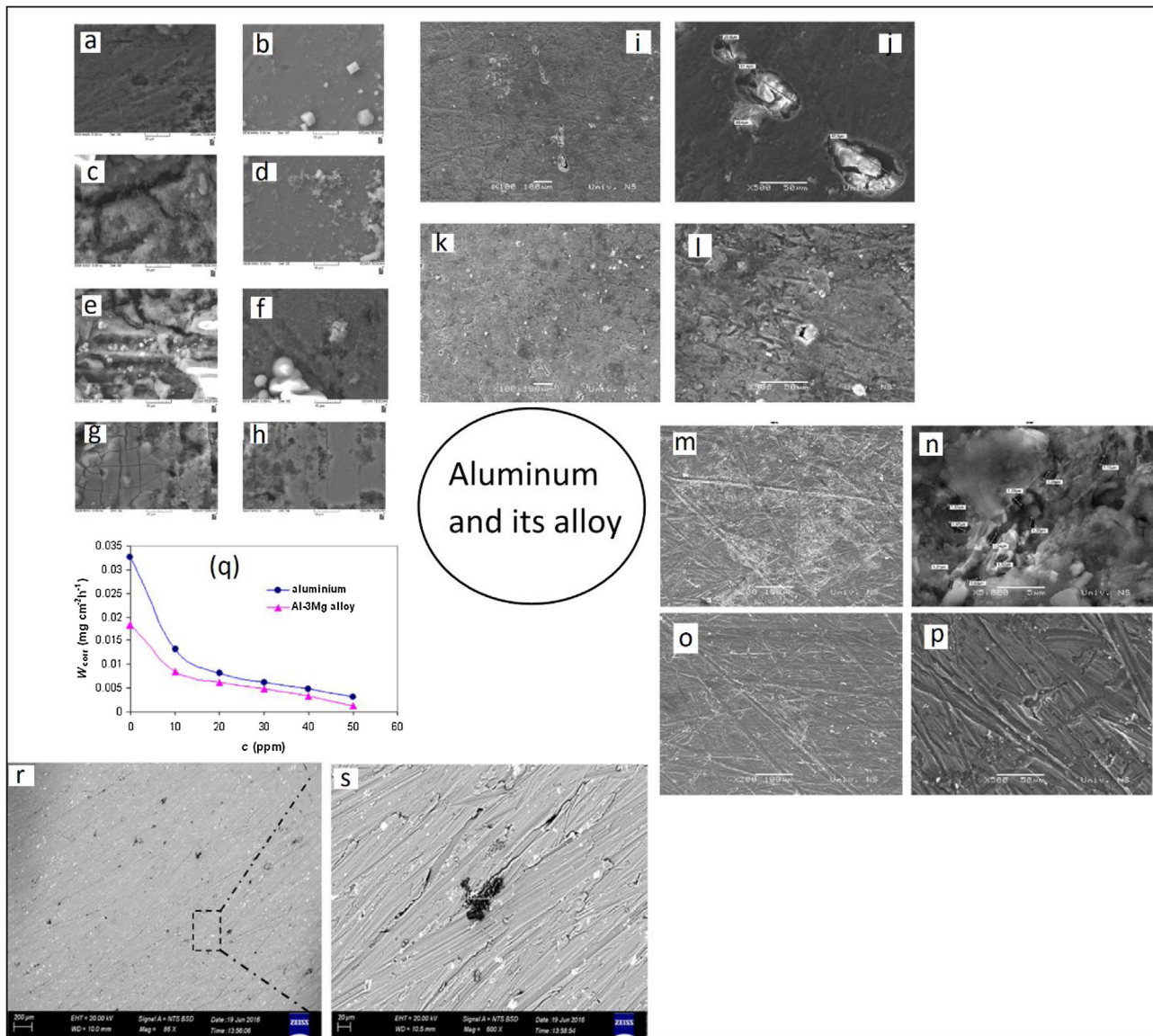
Fig. 7. The corrosion inhibiting strength of plant extracts for aluminum and its alloy in NaCl environment. The SEM micrographs of 7075 aluminum alloy surfaces in 3.5% NaCl solution without inhibitor at (a) 298 K, (c) 313 K, (e) 323 K, (g) 333 K and with 1000 ppm berberine at (b) 298 K, (d) 313 K, (f) 323 K, (h) 333 K [260]. The corrosion inhibition efficiency recorded was 95%, 79%, 39%, and 18% at 298 K, 313 K, 323 K, and 333 K respectively. The images of AA5754 aluminum alloy exposed to 3% NaCl solution without inhibitor for 24 h is shown in (i) and (j) while (k) and (l) show the metal surfaces inhibited with 20 ppm *Laurus nobilis* L. oil. The corrosion inhibition efficiency afforded by the 20 ppm of the extract was 54.1% but 84.4% was possible with 50 ppm concentration (q) [262]. The *Laurus nobilis* L. oil also effectively prevented pitting corrosion (o, p) compared to its absent (m, n) [262]. In addition, it was found that 1200 ppm *Linum usitatissimum* seeds extract was effective for aluminium in 3.5% NaCl solution and inhibited the metal surface by 82% (r, s) after 24 h of immersion. Figures were reproduced with permission from the original sources as follow: (a–g) were from Singh et al. [260] (Copyright 2014 The Korean Society of Industrial and Engineering Chemistry); (i–p) were from Halambek et al. [262] (Copyright 2012 Elsevier B. V.); (r, s) were from Elgahawi et al. [267] (Copyright Springer International Publishing AG 2017).

corrosion in oilfields [324–328]. Actually, these organic compounds are effective [325–328] but the areas of concern are the high cost of formulation and their toxicity level [324]. Most authors therefore justify their interest in exploring natural substances as sweet corrosion inhibitors on this backdrop. Gingko biloba fruit [322], pomelo peel [329], Saccharum sinense bagasse [330], Calotropis procera leaves [323], tangerine peel [331], and Ficus carica leaves [332] extracts are among the recently tested plant extracts for sweet corrosion inhibition. Gingko biloba fruit, Saccharum sinense bagasse, and tangerine peel extracts at a concentrations of 1000 ppm, 5% (v/v), and 4% (v/v) inhibited J55 steel corrosion in 3.5 wt.% NaCl solution saturated with CO_2 by 97%, 94.9%, and 83.3%. 0.4 g/L pomelo peel retarded N80 steel sweet corrosion by 86%. Calotropis procera and Ficus carica leaves

extracts at 50 ppm demonstrated 80% and 90% protection respectively to mild steel surface. The plants extracts in all cases behaved as mixed type corrosion inhibitors and inhibit by adsorptive mechanism. It can be said based on the above-cited results and those in Fig.9 that plant extracts are promising sweet corrosion inhibitors.

H_3PO_4 environment

Phosphoric acid is an important industrial acid used mainly in fertilizer production. It has also gain preference nowadays over HCl and H_2SO_4 acids in acid cleaning because the dissolution rate of metals in H_3PO_4 acid is slower than in HCl and H_2SO_4 acids [333,334]. Also, H_3PO_4 is used in electropolishing of Al [335]. In
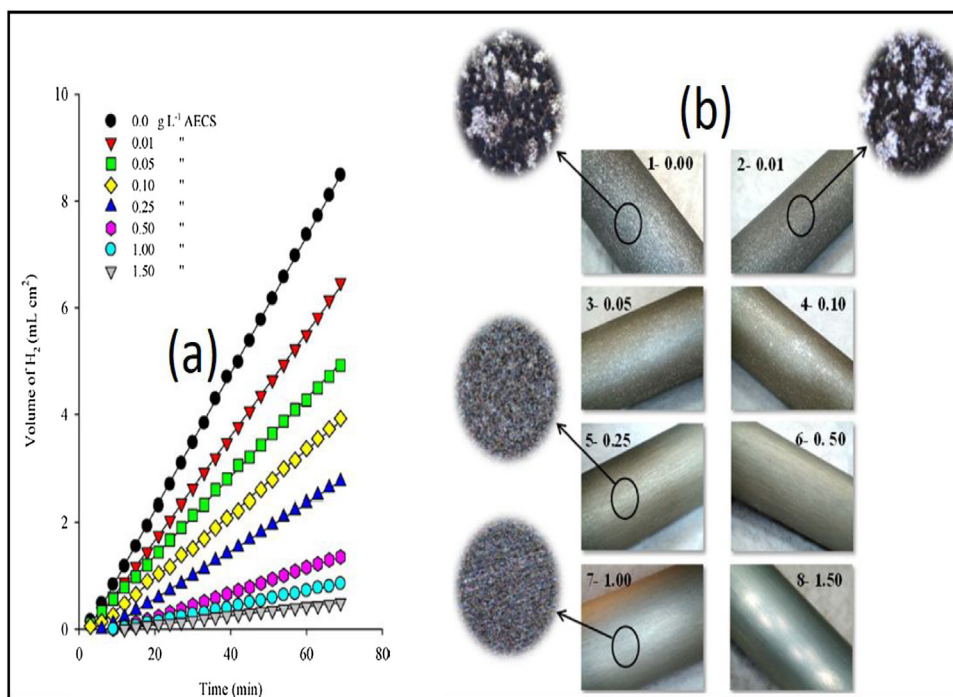
Fig. 8. Effective Al corrosion mitigation by various concentrations of aqueous extract of celery (*Apium graveolens* L.) seeds (AECS) in 0.25 M NaOH; (a) the variation of volume of H$_2$ evolved with exposure time and (b) the macroscopic and microscopic images for Al specimen in the absence and presence of different concentrations of AECS. The addition of an AECS decreased the corrosion of the metal and the volume of hydrogen gas evolved decreased (a). Increase in the concentration of the extract makes the inhibitor more efficient and reactive. At the higher concentration, the surface of the metal appeared brighter and smoother (b). The 1.50 mg/L afforded inhibition efficiency of 92.73% [291]. Extracted with permission from Al-Moubaraki et al. [291]. Copyright 2017 Elsevier Ltd.

pure state, the corrosivity of H$_3$PO$_4$ is mild when compare to that of nitric or sulphuric acids but becomes aggressive when impurities like chlorides, fluorides, and sulphides are present [334]. The impurities somehow are inevitable as they are often introduced during production. Predominantly (about 95%), H$_3$PO$_4$ is produced by wet process, which involves the reaction of phosphate ore with concentrated sulphuric acid followed by filtration [336]. Prevention of metals corrosion in environments where phosphoric acid is used is therefore imperative. Many organic compounds have been reported as effective inhibitor for H$_3$PO$_4$ corrosion [337,338]. Since the demand of the present is on green inhibitors, some plant parts extracts have been examined for anticorrosive property in aggressive H$_3$PO$_4$ medium. Prabhu and Rao [339] assessed the corrosion inhibition characteristics of aqueous extract of seeds of Coriandrum sativum L. (CSE) on aluminum in 1.0 M H$_3$PO$_4$ solution using electrochemical and surface analysis techniques. CSE was found to physically adsorb on Al surface following Langmuir adsorption model and retarded both the anodic and cathodic dissolution processes of the metal. The presence of 500 ppm CSE in the corrosive medium prevented Al corrosion by 72.75% at 30 °C but the inhibition efficiency decreased to 50.54% when the system temperature was raised to 50 °C. In the investigation on the adsorption and corrosion mitigation tendency of alcoholic Psidium guajava (guava) leaf extract on mild steel in 1 M H$_3$PO$_4$ by Victoria et al. [340], the inhibition efficiency of the leaf extract was found to increase with concentration up to 800 ppm and thereafter decreased slightly at 1200 ppm. 800 ppm as the optimum concentration gave inhibition efficiency of 89%. The decrease in inhibition efficiency after the critical concentration can be associated with desorption of the adsorbed inhibitor molecules into the bulk solution [340]. It has equally been shown that plant extracts can be deployed as effective corrosion inhibitor in highly aggressive H$_3$PO$_4$ environment. For example, Boudalia et al. [334] reported that 1.2 g/L oil of leaves of Lanvandula stoekas [L] could

suppress the corrosion of alloy UB6 stainless steel in 5.5 M H$_3$PO$_4$ solution by 87.3% at 298 K. The oil extract behaved as a mixed type corrosion inhibitor and equally showed capacity to inhibit at temperature as high as 353 K (62.8% inhibition had been achieved).

Other media

Other corrosive environments in which plant extracts have been tested in recent times as metals corrosion inhibitors include Na$_2$CO$_3$ [341,342], citric acid (H$_3$C$_6$H$_5$O$_7$) [343], biodiesel [344], geothermal fluid [345], trichloroacetic acid (Cl$_3$CCOOH) [346], and artificial saliva [347]. The essential oil of Artemisia herba alba [341] and eugenol oil [342] were tested as inhibitor for Pb and Al respectively in 0.1 M Na$_2$CO$_3$ medium. The oils were effective; 2000 ppm Artemisia herba alba oil showed 77% inhibition efficiency and 4.0 g/L eugenol oil demonstrated 81% protection to the considered substrate. Phyllostachys nigra Munro leaves' extract (PMLE) was applied as inhibitor for cold rolled steel and zinc in 0.2 M H$_3$C$_6$H$_5$O$_7$ solution [343]. PMLE, although effective was a better corrosion inhibitor for zinc than steel. For zinc, 2.0 g/L PMLE afforded 92.5% inhibition but 85.1% protection to steel surface. It was assumed by Li et al. [343] that, rutin was the primary contributor to the inhibitive property of PMLE extract and its adsorption was through chemical molecular skeleton structure of falvones backbone. Deyab [344] subjected Rosemary extract to a test as aluminium corrosion inhibitor in biodiesel. The author observed that as the concentration of Rosemary extract was increased in the corrosive medium, Al dissolution rate decreased but increased when the temperature of the corrosive system was raised. The extract acted as mixed type corrosion inhibitor and 0.5 g/L of the extract reduced Al corrosion rate from 1.785×10^{-5} mg cm^{-2} h^{-1} to 0.075×10^{-5} mg cm^{-2} h^{-1} with 95.7% as the inhibition efficiency. Two plants extracts had been reported as corrosion inhibitor for St37 steel in geothermal fluid: Quercus
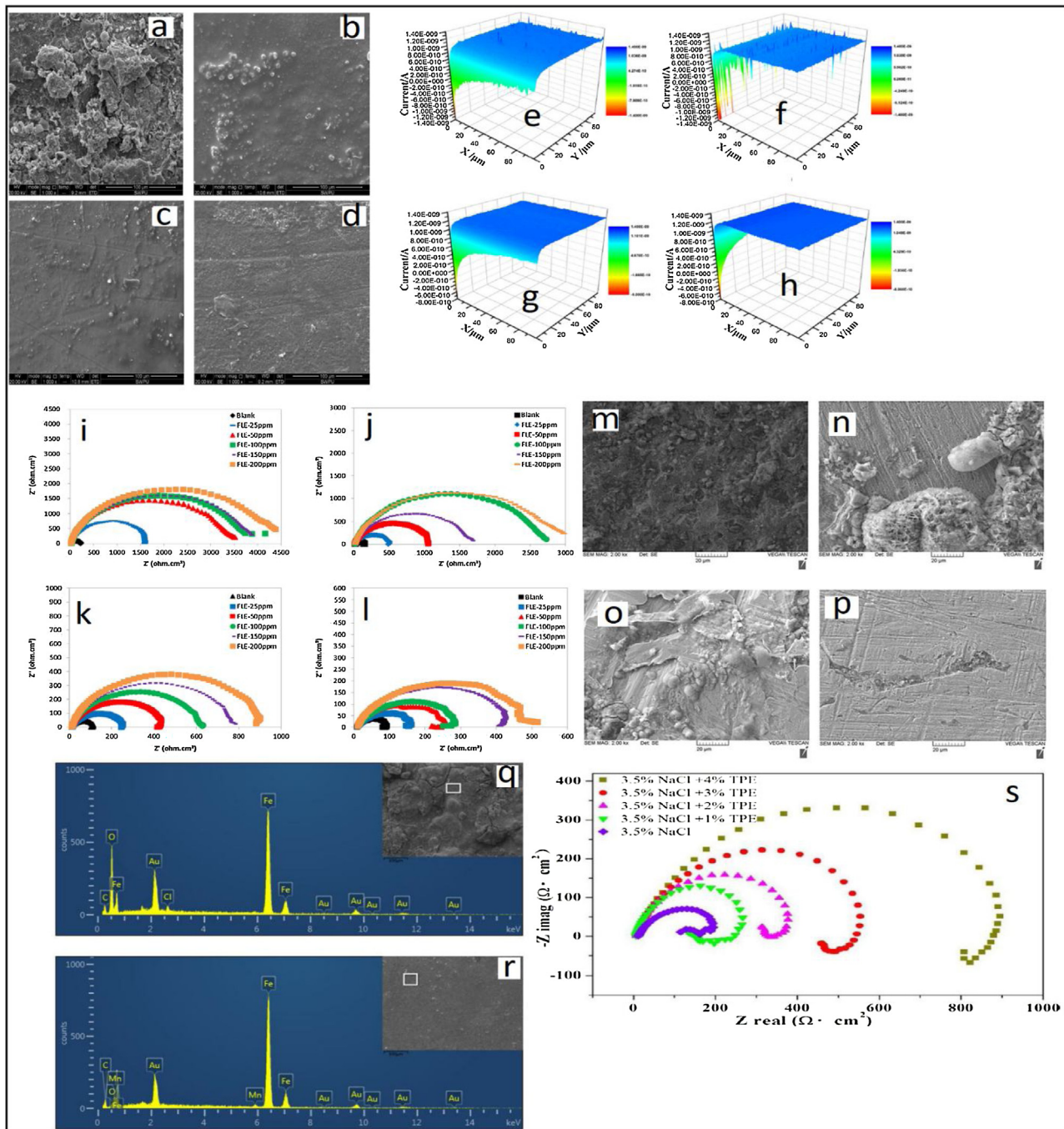
Fig. 9. Performance of plant extracts as sweet corrosion inhibitor. Shown are the SEM images for J55 in.(a, m) free 3.5 wt.% NaCl solution saturated with CO_2 and containing (b) 250 ppm *Gingko biloba* (GFE), (c) 500 ppm GFE, (d) 1000 ppm GFE. The protective ability of the extract is obvious as the surfaces in (b)-(d) are smoother than the one in (a). The 250 ppm, 500 ppm and 1000 ppm GFE afforded 87%, 95%, and 97% inhibition efficiency respectively [322]. The 3-D micrographs of scanning electrochemical microscopy of N80 steel in unprotected 3.5% NaCl solution saturated with CO_2 is shown in (e) (X-axis) and (f) (Y-axis). The X-axis and Y-axis 3-D micrographs recorded in the sweet solution fortified with 0.4 g/L Pomelo peel extract is displayed as (g) and (h) respectively. The surfaces of (g) and (h) are smoother compared to those of (e) and (f). From the electrochemical results, the protection effectiveness of the extract was 93.3% [329]. Shown in (i), (j), (k), and (l) are the Nyquist plots for mild steel in sweet environment without and with various concentrations of *Ficus carica* leaves extract (FCLE) at 25 °C, 40 °C, 50 °C, and 60 °C respectively. At 25 °C, 25, 50, 100, and 200 ppm FCLE afforded inhibition efficiency of 90.7%, 93.5%, 94.5%, 94.3%, and 95.0% respectively [332]. The inhibition efficiency declined upon increasing the system temperature. From the report of Huang et al. [330], the presence of 1% vol./vol., 2% vol./vol., and 5% vol./vol. *saccharum sinense* bagasse extract in 3.5 wt.% NaCl solution inhibited J55 steel corrosion by 94.5% (n), 93.3% (o), and 94.5% (p) respectively. According Wang et al. [331], addition of various concentrations of Tangerine peel extract controlled the dissolution of J55 steel in sweet environment effectively (s). The active components in the extract adsorbed on the steel surface and prevented further corrosion as indicated in (r) relative to (q). The figures were extracted with permission from the original sources thus: (a–d) were from Singh et al. [322] (Copyright 2014 The Korean Society of Industrial and Engineering Chemistry); (e–h) were from Sun et al. [329] (Copyright Springer Science + Business Media B. V. 2017); (i–l) were from Ibrahim et al. [332] (Copyright 2017 Informa UK limited, trading as Taylor & Francis Group); (m–p) were from Huang et al. [330] (Copyright Emerald Group Publishing Limited); and (q–s) were from Wang et al. [331] (Copyright 2017 The Authors).

robur [345] and pomegranate peels [345] extracts.250 mg/L Quercus robur or 500 mg/L pomegranate peels extracts, according to Buyuksagis et al. [345] could ensure 90% to St37 steel in geothermal fluid. In Cl$_3$CCOOH environment, 200 mg/L Dendrocalamus brandisii leaves extract was required to safeguard cold rolled steel by 97% [346].

Dental amalgam (alloy of mercury, silver, tin and copper) is known for application in dentistry to fill cavities caused by tooth decay [348]. The corrosion and wearing resistance of dental amalgam is a challenge that has drawn serious attention in recent times. The worries are on the toxicity of oral cavity's corrosion inhibitors and amalgam particles products, possible release of Hg in which its adverse effect in terms of antioxidation and neurotoxicology is well known [349,350]. Corrosion scientists have thought it wise to examine plant extracts as nontoxic inhibitor for dental amalgam although such investigation is very scanty. Otaifah et al. [347] studied the effect of Jasminum sambac extract on the deterioration of dental amalgam in saliva medium using chemical, electrochemical, scanning electron microscope, and energy dispersive X-ray spectroscopy techniques. Jasminum sambac extract at concentration of 0.33 g/L was found to provide 100% protection to the alloy in the studied medium. Fig. 10 displays the SEM images and EDAX spectra obtained by Otaifah et al. [237] to demonstrate the effectiveness of Jasminum sambac extract in inhibiting dental amalgam corrosion in artificial saliva.

Modifications on plant extract

The structural and/or compositional modification of a corrosion inhibitor to enhance its inhibition efficiency is a common practice.

Two approaches namely combination with substances that exert synergistic effect [351,352] and compositing [353,354] have been adopted for plant extracts in recent years. For the former approach, combination with halide ions [352] and metals ions [316] is very common. We had pointed out in our previous reviews [249,355] that combination of plant extracts with metals ions like Zn^{2+} [355] and iodide ions [249] improved inhibition efficiency remarkably. We had explained [355] that Zn^{2+} ions apart from being ecofriendly, form complex with inhibitor molecules easily and the Zn^{2+}-inhibitor complex is transported from bulk solution to substrate surface. The complex is converted to substrate-inhibitor complex in the local anodic regions and the latter complex is more stable than the former. The freed Zn^{2+} ions on the substrate surface combine with hydroxyl ions to form Zn(OH)$_2$ precipitates on the cathodic regions. These two products (metal-inhibitor complex and Zn(OH)$_2$ precipitates) are mainly responsible for the enhanced inhibition. For iodide ions, we stated that the ions are predispose to adsorption on metals surfaces than other halide ions [249] because of their higher ionic radius and hydrophobicity as well as their lower electronegativity than the other halides. The ease of chemisorption of iodide ions and the high surface recharging power favour adsorption of more charged inhibitor species. Most recent works on modification of plant extracts are via these approaches (addition of Zn^{2+} or iodide ions). Johnsirani et al. [316] upon addition of addition of 25 ppm Zn^{2+} ions to 6 mL Eclipta alba extract upgraded the inhibition efficiency of the extract from 60% to 92% for carbon steel in sea water environment. Li et al. [351] observed cooperative co-adsorption between bamboo of Dendrocalmus sinicus Chia et J.L. Sun leaf extract (DSCLE), its major compounds (rutin and orientin) with iodide ions on cold rolled
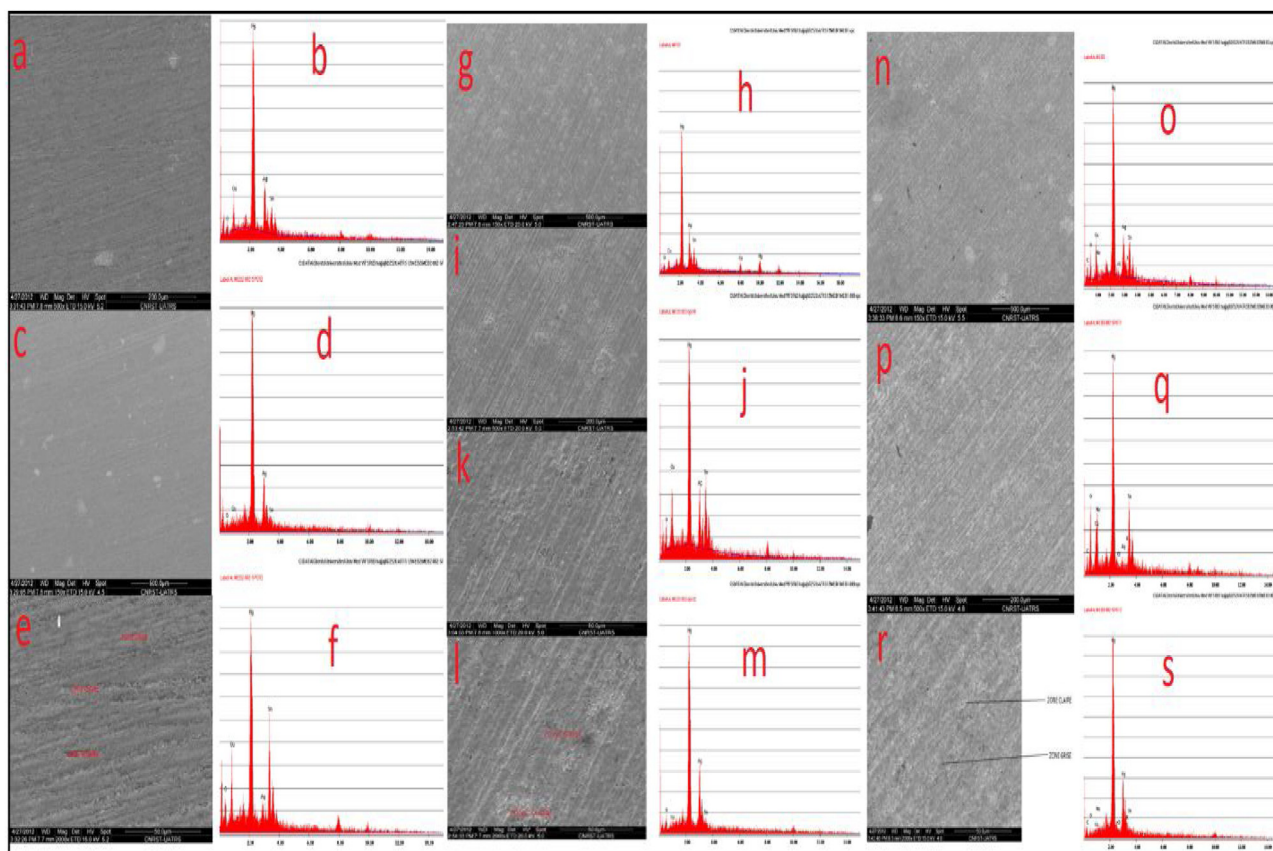

Fig. 10. The SEM and EDAX pictures of amalgam (a–f) before immersion and after immersion in artificial saliva for 7 day (g–m) free of *Jasminum sambac* extract, (n–s) containing *Jasminum sambac* extract at different magnification. It was reported [347] that the presence of 0.33 g/L *Jasminum sambac* extract in the artificial saliva provided 100% protection to the alloy surface (n–s). Adapted with permission from Otaifah et al. [347].

steel in 0.1 M H_3PO_4 solution. By combining 5.0 mL iodide ions with 100 mg/L DSCLE, rutin, and orientin, the mixture synergistically adsorbed on the substrate surface and this led to an enhancement on the inhibition efficiency from 51.2%, 42.5%, 34.0% respectively to 95.7%, 91.0%, and 86.7%. Similar cooperative co-adsorption and upgrade of inhibition efficiency had been reported for Morinda tinctoria leaves extract [356], coconut coir dust extract [352], Mentha pulegium [357], and Artemisia halodendron [358] with iodide ions.

Nanotechnology enrouted synthesis of metal nanoparticles is the center of attention presently. All fields including physics, chemistry, biology, medicine, engineering, material science, etc. [359–362] have developed keen interest on nanotechnology. The reason is not far fetched. Nanoparticles present exceptional properties compared to the conventional macro particles. Their unique and exceptional characteristics are attributed to their large surface to volume ratio [359,360,362]. Plant extract mediated metals nanoparticles have been presented by corrosion scientists as inexpensive, low toxic, and environmentally compatible materials that should be deployed as metals anticorrosive substance. Metals nanoparticles are believed to have the capacity to chemisorb on metal surfaces and retard metals corrosion [361]. Asaad et al. [353] studied the effect of Elaeis guineensis (EG)/silver nanoparticles (AgNPs) on the enhanced corrosion resistance of mild steel in 1 M HCl. The steel surface was found to adsorb the EG/AgNPs and form protective films (Fig. 11) advantageous for suppressing the attack by aggressive ions present in the studied medium. Steel specimen immersed in acid solution containing 10% (v/v) of EG/AgNPs inhibitor was found to be protected by 94.1% (Fig. 11). Similarly, Olive leaves extract/Ti nanoparticles was reported to inhibit mild steel in 1 M HCl solution by 94.3% at 30 °C [354]. In the investigation carried out by Izadi et al. [363], an active nanoreservoir was obtained through inclusion of Nettle extract as a green corrosion inhibitor. The authors deployed a polyelectrolyte layer of polyaniline to load the corrosion inhibitor on Fe_3O_4 nanoparticles (Fe_3O_4-NP). The results obtained indicated that the Fe_3O_4-NP filled with Nettle extract was an efficient active corrosion protective system and the inhibitive performance was attributed to the presence of active agents such as Quercetin, Quinic acid, Hystamine and Serotonin in the Nettle extract.

Mechanism of inhibition by plant extract

Plant extracts, as earlier stated, contained various organic bioactive molecules. The bioactive molecules have heteroatoms and/or π-electrons in them which are believed to serve as the center of interaction with metal surfaces [155,173,174]. The type of adsorption between organic molecule and a metal surface is primarily determined by factors like (i) pH of the corrosive medium, (ii) the true state of the molecule in the corrosive medium, (iii) the anions present in the medium, (iv) the temperature of the medium, and (v) the charge on the substrate surface. The charge on a metal surface could be determined by finding the difference (d) between the corrosion potential (E_{corr}) and the potential of zero charge (PZC) ($E_{q=0}$) [364,365,366]. If d = E_{corr}−$E_{iq=0}$ is negative, the substrate surface is expected to acquire positive charges [366]. If d = E_{corr}−$E_{q=0}$ is positive, the metal surface should be negatively charged [366]. Depending on the pH of a system, an organic molecule may exist as neutral or protonated specie. A positively charged surface in a system that has negative anions would unarguably attract the anions to itself. If inhibitor species exist in the system as protonated species, then the strength of their adsorption will be a function of the recharging power of the adsorbed anions on the metal surface. In such a case, the inhibitor species are electrostatically adsorbed on the metal surface, the so-called physisorption mechanism. Many reports on
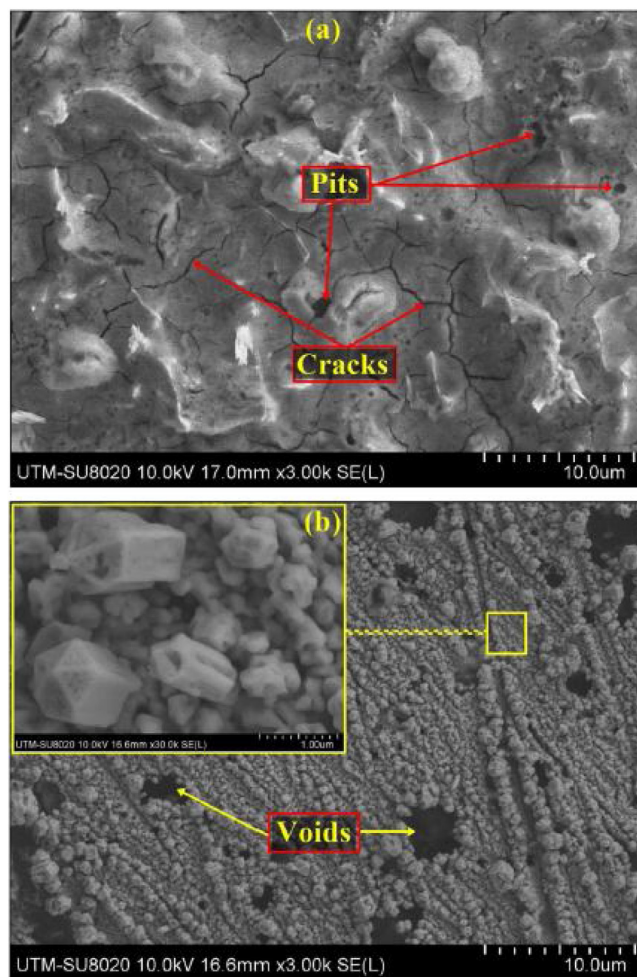

Fig. 11. FESEM images of mild steel (a) uninhibited (1 M HCl), and (b) inhibited with 10% (v/v) of *Elaeis guineensis*/AgNPs after 120 h of immersion. In the absence of any inhibitor, the metal surface present rough microstructure morphology due to acid attack. Deep pits and micro-cracks can be clearly seen in Fig. 11(a). In the presence of EG/AgNPs inhibitor, the metal surface is protected and appears smoother. Experimental results revealed that the protection offered to this surface by *Elaeis guineensis*/AgNPs was up to 94.1%. Extracted with permission from Asaad et al. [353]. Copyright 2018 Published by Elsevier B.V.

plant extracts as metals corrosion inhibitors claimed that their mode of adsorption is physisorption [71,76,302]. In such a case, the extract molecules are held on the metals surfaces by electrostatic force and increase in system temperature usually weaken the adsorption bond, that is, inhibition efficiency decreased with rise in temperature [367]. Neutral inhibitor molecules in a corrosive system can donate their electron pairs to vacant orbital of a metal substrate to form a covalent or co-ordinate type of bond – chemisorption mechanism. Increase in system temperature is beneficial to this type of adsorption, i.e. inhibition efficiency increased with increase in temperature. It should be stated that, protonated inhibitor species can be neutralized on metal surface such that the freed heteroatoms go into covalent or coordinate bonding with the metal surface. Chemisorption mechanism has equally been observed for plant extracts [68]. The two mechanisms – physisorption and chemisorption can as well occur on the same metal surface [69,75,260]. The most appropriate way to ascertain the predominant adsorption mechanism on a metal surface is by calculating the standard enthalpy of adsorption (ΔH^0_{ads}). For an endothermic system, ΔH^0_{ads} value greater than zero reflects chemical adsorption whereas ΔH^0_{ads} value less than zero points

to physisorption mechanism [361]. In the same vein, in exothermic system, ΔH_{ads}^0 value less than 40 kJ/mol signifies physical adsorption and ΔH_{ads}^0 value approaching 100 kJ/mol is indicative of chemisorption [361]. For the adsorption of Phyllostachys nigra Munro leaves' extract on cold rolled steel and zinc in 0.2 M citric acid solution, the calculated ΔH_{ads}^0 values were negative and within 20.35–25.75 kJ/mol [343]. Also, for inulin adsorption, the ΔH_{ads}^0 value was – 27.96 kJ/mol [368] and justify physical adsorption of inulin molecules on 6061Aluminium-15%(v) SiC(P) composite in 0.05 M HCl solution. In the case of Silybum marianum extract adsorption on 304 stainless steel surface in 1.0 M HCl, calculated ΔH_{ads}^0 value was + 26.70 kJ/mol [68] that defined the adsorption process as endothermic and the mechanism as chemisorption.

Drawbacks and future perspective

Based on the numerous research findings reviewed in this article, it can be said that extracts from plants are reliable green and inexpensive metals corrosion inhibitors. However, there are still some unanswered questions regarding the use of plant extracts as metals corrosion inhibitors. Does all the bioactive molecules in plant extract responsible for inhibitive property? Is inhibition a function of the combined influence of bioactive molecules? Are they some components that could diminish the inhibiting ability of others? What is the exact inhibition mechanism by plant extracts? The obvious is that, the mechanism of inhibition by plant extracts may not be mere adsorption mechanism as often claim since extract contains multi-components. Future researches on plant extracts as corrosion inhibitor should therefore be focused on isolation of the specific active molecule that is responsible for the anticorrosive property of extract. Researches should also focus on establishing whether such active molecule exert inhibitory property independently or in synergy with others. Such finding will provide a lead way into understanding the corrosion inhibition mechanism by plant extracts. It is also observed from the reviewed scientific articles that plant extracts cannot withstand very high temperatures and are not effective in highly aggressive environments. We pointed out under the Section "HCl environment" that it appears that plant extracts are not considered for acidization purpose. Acidization process is very severe and requires highly potent inhibitor. If the active biomolecules in extract is isolated and the mechanism of inhibition understood, such a component can be modified to withstand severe corrosive environment. In fact, commercial metals corrosion inhibitors for oil wells acidization are multi-components formulations [369].

Conclusion

The deterioration of metals in an environment of application is inevitable. Metals corrosion has drained the world economy and has caused loss of human lives and valuable properties. Preventive mechanisms are often put in place and the most economical is the use of chemical inhibitors. Organic and inorganic metals corrosion inhibitors are effective but are somewhat expensive and harmful to human and the natural environments. Substances of natural origin like natural polymers and plant parts extracts are the center of current researches. Plant extracts are found in abundant; they are inexpensive, nontoxic, biodegradable, biocompatible, and contain heteroatoms and/or pi electrons that make them suitable as metals corrosion inhibitor. They have been tested in various corrosive environments such as HCl, H_2SO_4, Na_2SO_4, NaCl, NaOH, Na_2CO_3, H_3PO_4, aqueous, CO_2, citric acid, biodiesel, geothermal fluid, trichloroacetic acid, and artificial saliva. In all these media, plant

extracts are adjudged effective. The mechanism of inhibition by plant extracts is basically adsorptive mechanism which could be physisorption or chemisorption or both. The major challenge facing the use of plant extracts as metals corrosion inhibitor is the inability to pinpoint the exact bioactive molecule responsible for corrosion inhibition. Future researches should therefore be focused on this direction. This review article will serve as a masterpiece for corrosion scientists and students of materials sciences and related disciplines.

Acknowledgements

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