Corrosion inhibition of A516 carbon steel in 0.5 M HCl solution using *Arthrospira platensis* extract as green inhibitor

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Abstract

The study on the influence of *Arthrospira platensis* extract as a corrosion inhibitor on A516 carbon steel in 0.5 M HCl solution has been conducted. The extract was obtained by the maceration method and characterized by phytochemical analysis and infrared spectroscopy. The effect of corrosion inhibition of *A. platensis* extract was studied by weight loss method, electrochemical impedance spectroscopic measurements and scanning electron microscopy. The dosage of *A. platensis* extract was varied between 100 to 250 ppm. The results of phytochemical and FTIR analysis show that the extract of *A. platensis* revealed the presence of saponin, steroid and alkaloid compounds, along with hydroxyl, carbonyl, amine, and carboxylic groups, which are responsible for the adsorption on the carbon steel surface. The extract of *A. platensis* showed good performance at a dose of 250 ppm with inhibition efficiency of 66% at 298 K. The charge transfer resistance increases from 82 $\Omega \cdot cm^2$ to 246 $\Omega \cdot cm^2$ in the electrochemical impedance spectrum after the addition of 250 ppm *A. platensis* extract. The adsorption of this extract on the surface of A516 carbon steel obeys the Langmuir isotherm. Thermodynamic parameter of adsorption allows us to suggest the adsorption process was dominated by physical adsorption. SEM analysis supported the inhibitive action of the *A. platensis* extract against the carbon steel corrosion in acid media.

Keywords: *carbon steel, corrosion, green inhibitors, Arthrospira platensis, microalgae.*

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1. Introduction

Carbon steel is the most useful material due to its wide range of engineering and industrial applications. This material is used in many industries due to its high strength and reliability. Carbon steel is used in industries as pipelines for petroleum industries, storage tanks, boiler pipelines, cooling tower and chemical batteries [1]. These various industrial equipments are vulnerable to the formation of scale or rust which can reduce the efficiency of the equipments. Acid solutions are generally used for the removal of undesirable scale and rust in several industrial processes. Hydrochloric acid is widely used in the pickling processes of metals. However, due to their corrosive nature of acids may cause damage to the system components. Use of inhibitors is one of the most practical methods for protection against corrosion especially in acid solutions to prevent metal dissolution and acid [2].

An inhibitor is a substance (or combination of substances) added in very low concentration to protect the surface of a metal that is exposed to a corrosive environment that terminate or reduce the corrosion of a metal [3]. Environmental concerns require corrosion inhibitors to be non-toxic, environment friendly and biocompatible. Organic compounds with functional groups containing nitrogen, sulphur and oxygen atoms are generally used as corrosion inhibitors. However, most of these organic compounds are not only expensive but also harmful to the environment [4]. Current research is increasingly focusing on the use of inhibitors known as green inhibitors. The term "green inhibitor" or "eco-friendly inhibitor" refers to the substances that have biocompatibility in nature. Plants and seaweeds extracts are viewed as an incredibly rich source of naturally synthesize chemical compounds that can be extracted by simple procedures with low cost. The inhibitors like plant extracts presumably possess biocompatibility due to their biological origin [3, 4].

Various plant extracts have been investigated as green inhibitor such as *Zenthoxylum alatum* plant [2], henna [5], *Murraya koenigii* leaves [6], potato peel [7], green tea [8], Fig leaves [9], green *capsicum annuum* fruit [10], *Acalypha torta* leaf [11], *Tagetes erecta* (Marigold flower) [12], *Musa paradisica* peel [13], *Phyllanthus amarus* leaf [14], *Butea monosperma* [15], *Ginkgo* leaf [16], *Citrullus lanatus* fruit [17], galangal rhizome [18], *etc*. Moreover, marine algae are also considered as a natural source for green inhibitor, *inter alia* the extracts of *Hydroclathrus clathratus* [19], *Kappaphycus alvarezii* [20], *Sargasam swartzii* [21], *Spirogyra* [22], *Halopitys Incurvus* [4], *etc*.

Arthrospira platensis belongs to the oxygenic photosynthetic bacteria that cover the groups *Cyanobacteria* and *Prochlorales* which have several therapeutic properties such as hypocholesterolemia, immunology, antiviral, and antimutagenic [23–26]. These microalgae contain amino acids such as methionine, γ-linolenic acid, peptides, protein, carbohydrates and phycocyanin. Therefore, microalgae are widely used in the food industry because of its high protein content [25, 26].

In this study the corrosion inhibitive effect of *A. platensis* extract on A516 carbon steel in a 0.5 M HCl solution has been confirmed by weight loss, electrochemical impedance spectroscopy (EIS) and scanning electron microscopic (SEM) techniques. The inhibitor active components were investigated by phytochemical and infrared spectroscopy (FTIR) analysis.

2. Experimental

2.1 Preparation of A. platensis extract

Microalgae *A. platensis* was obtained from Amorina Padjadjaran University Bandung, Indonesia. Powdered *A. platensis* (100 g) were macerized in 96% ethanol with ratio 1:10 for 5 days, solvent replacement every 1×24 hours. The mixture was filtered off and the filtrate was evaporated. A dark green solid extract was used to prepare the required concentrations (100–250 ppm) of *A. platensis* extract in 0.5 HCl solution.

2.2 Phytochemical analysis

Phytochemical analysis include test for saponin performed by the addition of hot distilled water to extracts, test for terpenoid and steroid carried out by adding a few drops of concentrated HCl and H_2SO_4, test for flavonoid performed by the addition of Mg and a few drops of concentrated HCl, test for tannin carried out by the addition of $FeCl_3$ hydrate, and test for alkaloid performed by the addition a few drops of chloroform and Meyer's reagent [27].

2.3 Characterization by FTIR

FTIR spectra of *A. platensis* extract were obtained using PRESTIGE 21 SHIMADZU FTIR instrument in the frequency range of 4500–400 cm^{-1} and used to investigated the functional groups contained in *A. platensis* extract. FTIR measurement using KBr pellet method.

2.4 Specimen preparation

The A516 carbon steel specimen having composition of C=0.22%, Si=0.43%, P=0.01%, Mn=0.61%, Ni=0.14%, Mo=0.03%, V=0.04% and the remaining Fe. The density of A516 carbon steel is 7.85 g/cm^3. The carbon steel specimens used have rectangular shapes 2 cm×1.5 cm×0.2 cm, whereas specimens of 1 cm in diameter are used for EIS measurements. Prior to each experiment, the specimen was polished with different emery papers. The specimen was washed several times with distilled water, degreased and dried with acetone.

2.5 Weight loss measurements

Weight loss measurements were performed at 298 for 6 h by immersing the A516 carbon steel specimen into acid solution without and with various concentrations (100–250 ppm) of *A. platensis* extract. Afterwards, the specimens were taken out, rinsed off, dried and weighed accurately. The inhibition efficiency (*IE*) and surface coverage (θ) were determined by using the following equation [6, 28]:

$$IE(\%) = \left(1 - \frac{W_1}{W_0}\right) \times 100 \tag{1}$$

$$\theta = \left(1 - \frac{W_1}{W_0}\right) \tag{2}$$

where W_0 and W_1 are the weight loss of the A516 carbon steel in the absence and presence of *A. platensis* extract, respectively. From the weight loss data (ΔW), the corrosion rate (CR) can be calculated by the following equation:

$$CR\,\frac{\text{mm}}{\text{year}} = \Delta W,\quad \text{g}\times\frac{\text{cm}^3}{\rho,\text{g}}\times\frac{1}{A,\text{cm}^2}\times\frac{10\,\text{mm}}{1\,\text{cm}}\times\frac{1}{t,\text{h}}\times\frac{8760\,\text{h}}{1\,\text{year}} \tag{3}$$

Inhibition efficiency could also be calculated from corrosion rate (CR) data according to the following equation [29]:

$$IE(\%) = \left(1-\frac{CR_1}{CR_0}\right)\times 100 \tag{4}$$

where CR_0 and CR_1 are the corrosion rates in the absence and presence of *A. platensis* extract, respectively.

2.6 Electrochemical impedance spectroscopy measurements

EIS experiments were conducted using potensiostat/galvanostat/ZRA GAMRY Reference 3000™ for measurements of impedance parameter such as the charge transfer resistance (R_{CT}) and double layer capacitance (C_{DL}). All experiments were performed with a frequency ranging from 0.2 Hz to 100 kHz. The inhibition efficiency could be determined electrochemically using the following equation [30]:

$$IE(\%) = \left(1-\frac{R_{\text{CT}_0}}{R_{\text{CT}_1}}\right)\times 100 \tag{5}$$

where R_{CT_0} and R_{CT_1} are the charge transfer resistance in the absence and presence of *A. platensis* extract, respectively, obtained from the Nyquist diagrams.

2.7 SEM image

The A516 carbon steel specimens were immersed in 0.5 HCl solution in absence and presence of 250 ppm of *A. platensis* extract for 6 h. The analysis was performed using JEOL JSM-6510 scanning electron microscope with an accelerating voltage of 20 kV.

3. Results and Discussion

3.1 A. platensis maceration and characterization of A. platensis extract

The extract of *A. platensis* was obtained by cold maceration method. This method is simple, efficient, and does not damage the chemical compounds contained in the extract [31]. The obtained *A. platensis* extract form dark green solids with a yield of 16.7%.

Phytochemical analysis shows that *A. platensis* extract contains saponin, steroid and alkaloid compounds. The result of phytochemical analysis of *A. platensis* extract are shown in Table 1 [27].

Table 1. Phytochemical analysis results of *A. platensis* extract.

Phytochemical analysis	Result	Evidence
Test for saponins	+	Formed the stable gas bubbles
Test for terpenoids	–	A grayish color was not observed
Test for steroids	+	Greenish color was observed
Test for flavonoids	–	Pink scarlet color did not appear
Test for tanins	–	A blue-green or black coloration was not observed
Test for alkaloids	+	Turbidity of the resulting precipitate was observed

FTIR spectra of *A. platensis* extract as shown in Figure 1 revealed the presence of hydroxyl, carbonyl, amino, and carboxylic group. The presence of OH group along with carbonyl group confirmed the presence of carboxylic acid groups in *A. platensis* extract. The presence of NH group and OH group along with carbonyl group might be attributed to the presence of amino acid groups [32]. Assignment of bands to functional group for *A. platensis* extract as observed from FTIR spectroscopy is summarized in the Table 2.

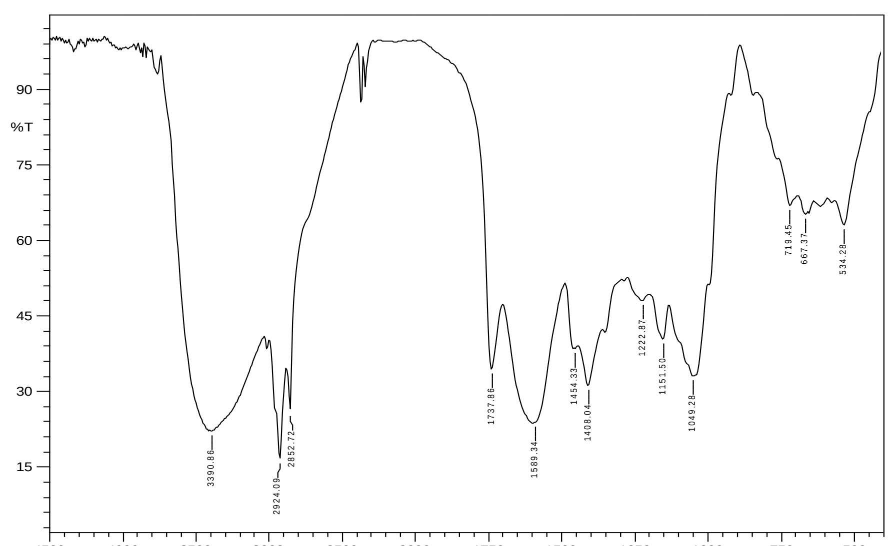

Figure 1. FTIR spectra of the *A. platensis* extract.

Table 2. Assignment of bands to functional group for *A. platensis* extract.

Wave number (cm^{-1})	Functional group
3390.86	O−H and N−H stretching
2924.09	C−H stretching aliphatic
2852.72	C−H stretching asymmetric
1737.86	C=O stretching vibration
1589.34	N−H bending vibration
1454.33	CH_2 bending vibration
1408.04	O−H bending vibration carboxylic acid
1222.87	C−N stretching amine
1151.50	C−O stretching
1049.28	CO−O−CO stretching anhydride

3.2 Weight loss study

Inhibition efficiency (*IE*) and surface coverage (θ) were obtained from measurements of weight loss at various concentrations of green inhibitors (100–250 ppm) at 298 K using Equations (1) and (2), respectively. Whereas, the corrosion rate was obtained from the weight loss data using Equation (3). The values of corrosion rate (*CR*), inhibition efficiency (*IE*) and surface coverage (θ) at different concentrations of green inhibitor at 298 K are given in Table 3.

Table 3. Corrosion parameters obtained from weight loss result of A516 carbon steel in 0.5 M HCl at 298 K for a 6 h immersion period in the presence and absence of different concentrations of *A. platensis* extract.

Concentration of *A. platensis* extract, C_{inh} (ppm)	Corrosion rate, *CR* (mmpy)	Inhibition efficiency, *IE* (%)	θ
blank	43.46	–	–
100	23.50	45.92	0.46
150	23.49	45.95	0.46
200	15.56	64.19	0.64
250	14.82	65.90	0.65

Inhibition efficiency was found to increase with increasing the concentration of inhibitor with maximum *IE* at a concentration of 250 ppm. The increasing in *IE* as a result of the increasing in the amount of constituent molecules of *A. platensis* extract adsorbed on the carbon steel surface at high concentrations, so that the active sites of the carbon steel are protected by the inhibitor molecules [33, 34].

Table 3 also exhibit the corresponding trend of corrosion rate for various concentrations of *A. platensis* extract and as estimated, corrosion rate decrease gradually with increasing green inhibitor concentration in 0.5 M HCl solution.

3.3 Adsorption isotherms and thermodynamic parameters

The type of interactions between *A. platensis* extract molecules and carbon steel surface could be evaluated by different isotherms. The adsorption isotherms of Langmuir, Temkin, Freundlich, and Frumkin were represented by the following equations [17, 35]:

Langmuir
$$\frac{C_{\text{inh}}}{\theta} = \frac{1}{K_{\text{ads}}} + C_{\text{inh}} \tag{6}$$

Temkin
$$\exp(-2\alpha\theta) = b \times C_{\text{inh}} \tag{7}$$

Freundlich
$$\log\theta = \log K_{\text{ads}} + n\log C_{\text{inh}} \tag{8}$$

Frumkin
$$\frac{\theta}{1-\theta}\exp(-2\alpha\theta) = b \times C_{\text{inh}} \tag{9}$$

where C_{inh} is the concentration of the *A. platensis* extract, θ is the surface coverage, and K_{ads} is the adsorption equilibrium constant.

As shown in Figure 2, the Langmuir adsorption isotherm fits the data well in the 0.5 M HCl solution. This revealed that the interactions between constituents molecules of *A. platensis* extract and carbon steel *via* monolayer adsorption over a homogeneous surface of the adsorbent [36]. The values of K_{ads} were calculated from the intercepts of the straight lines on C/θ axis. The values of K_{ads} also facilitated to evaluate the standard free energy of adsorption (ΔG^0_{ads}) using the following equation [4, 22, 37]:

$$\Delta G^0_{\text{ads}} = -RT\ln(C_{\text{H}_2\text{O}} \times K_{\text{ads}}) \tag{10}$$

where $C_{\text{H}_2\text{O}}$ = 10^3 g/L = 55.5 M, R is the universal gas constant, and T is absolute temperature.

The value of K_{ads} at 298 K is 8.22 which reflects the adsorption process is more favorable than the desorption. The negative value of ΔG^0_{ads} indicates the spontaneous adsorption and stable interactions occur between *A. platensis* extract and A516 carbon steel. Generally, ΔG^0_{ads} values of –40 kJ/mol or more negative are related to the chemical adsorption; while those around −20 kJ/mol are related to electrostatic interactions between charged molecules and metal charges (physical adsorption) [4, 39]. In the present work, the value of ΔG^0_{ads} is −15.16 kJ/mol show that *A. platensis* extract physisorbed on the surface of A516 carbon steel.

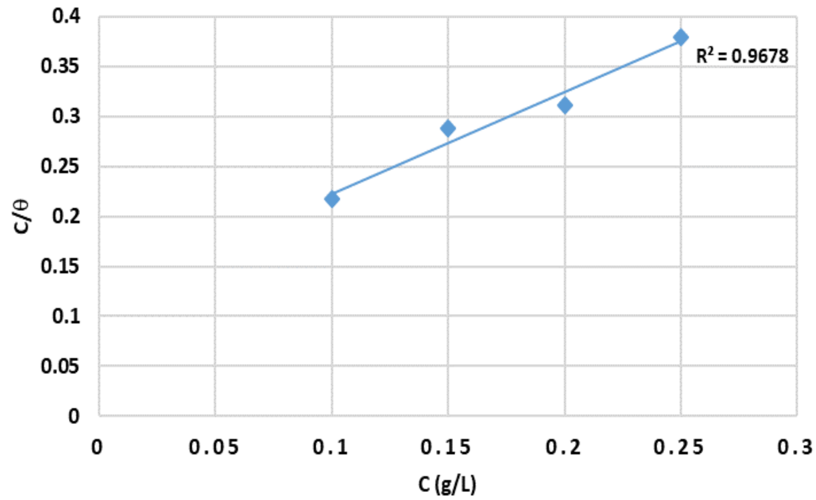

Figure 2. Langmuir adsorption isotherms plot for various concentrations of *A. platensis* extract in a 0.5 M HCl solution at 298 K.

3.4 Electrochemical impedance spectroscopy

Electrochemical impedance spectroscopy (EIS) is an important tool for determining several electrochemical parameters, such as double-layer capacitance (C_{DL}) and resistance to transfer (R_T). The investigation of the change of these parameters as a function of time or with respect to other variables provides us an important information on the kinetics of the corrosion process involved. The Nyquist diagrams for A516 carbon steel in 0.5 M HCl solution in the absence or presence of various concentrations of *A. platensis* extract at 298 K after immersion period of 6 hours are shown in Figure 3. The Nyquist diagrams of A516 carbon steel in acid media are not perfect semi-circles, and this is associated to frequency dispersion as a result of the surface heterogeneity due to the microscopic roughness of the electrode surface and inhibitor adsorption on it [4, 40]. The diameter of the capacitive loop increased with an increase in *A. platensis* extract concentration, indicating increased polarization resistance at the metal–electrolyte interface [38].

The electrochemical impedance parameters, the charge transfer resistance (R_{CT}), the double-layer capacitance (C_{DL}) and inhibition efficiency (*IE*) are summarized in Table 4. The inhibition efficiency (*IE*) was estimated from the charge transfer resistance measured, R_{CT}, using Equation 5. According to Table 4, it is found that the R_{CT} values increase while C_{DL} values reduce with the increasing of *A. platensis* extract concentrations. With a gradual increase of *A. platensis* extract concentration, the degree of surface coverage on the A516 carbon steel was enhanced, and as a consequence, R_{CT} and *IE* values were found to increase [38].

The lowered C_{DL} values comparing with that in the absence of *A. platensis* extract due to a decrease in local dielectric constant and/or an increase in the thickness of the electrical double layer, suggests that the inhibitor molecules function by adsorption at the metal–solution interface [38, 40]. These results denote a decrease in the active surface area in consequence of the adsorption of *A. platensis* extract on the A516 carbon steel surface and this suggests that the corrosion process has been obstructed. Furthermore, the results obtained from the EIS methods are in good agreement with those obtained from the weight loss measurements.

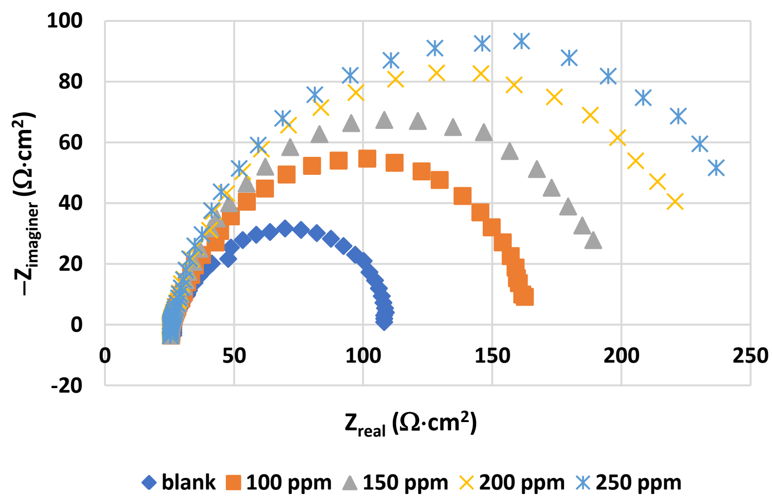

Figure 3. The Nyquist plots of the A516 carbon steel in 0.5 M HCl in the absence and presence of various concentrations of *A. platensis* extract.

Table 4. Electrochemical impedance parameters for the corrosion of A516 carbon steel in 0.5 M HCl in the absence and presence of various concentrations of *A. platensis* extract at 298 K after 6 hours of immersion.

C_{inh} (ppm)	R_{CT} ($\Omega\cdot cm^2$)	C_{DL} ($\mu F/cm^2$)	*IE* (%)
0	82.2	121.12	–
100	141.9	70.14	42.09
150	177.3	56.13	53.65
200	218.7	45.51	62.43
250	245.9	40.47	66.58

3.5 Surface analysis

SEM images of the surface of A516 carbon steel after immersion in 0.5 M HCl for 6 h and after immersion in 0.5 M HCl containing 250 ppm of *A. platensis* extract for 6 h are shown in Figure 4. As it is seen from Figure 4a, the A516 carbon steel surface is strongly damaged in the absence of the inhibitor due to the metal dissolution in a corrosive solution. In the presence of 250 ppm of *A. platensis* extract, the surface is much less rough and more uniform as shown in Figure 4b. This indicates that *A. platensis* extract inhibits corrosion of A516 carbon steel in 0.5 M HCl solution.

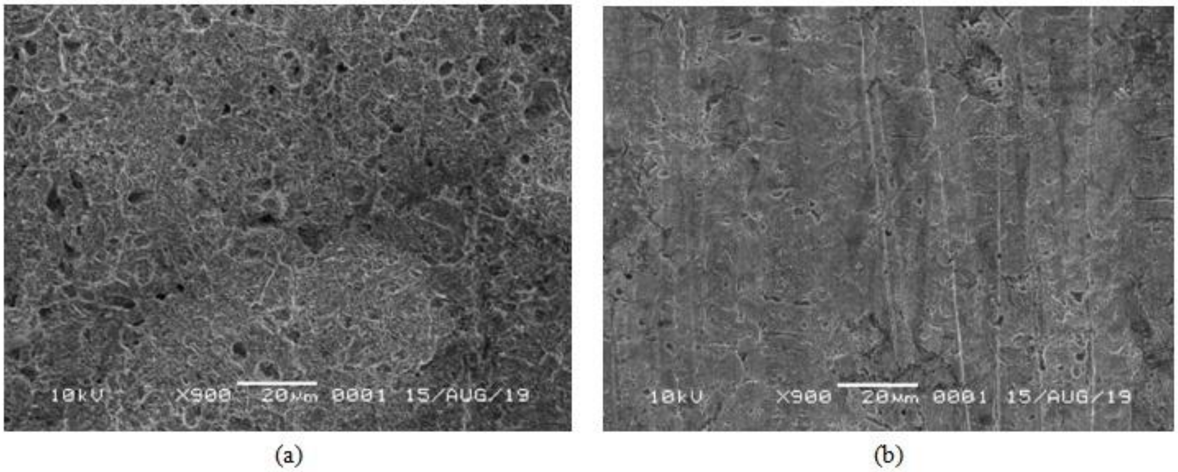

Figure 4. SEM images of the A516 carbon steel surface after immersion in 0.5 M HCl solution (a) in the absence and (b) presence of 250 ppm of *A. platensis* extract for 6 h.

4. Conclusions

The results can be drawn from this study as follows:

- The extract of *A. platensis* contains saponin, steroid, and alkaloid compounds, along with hydroxyl, carbonyl, amine, and carboxylic groups which are responsible for the adsorption on the A516 carbon steel surface.
- *A. platensis* extract acted as green inhibitor for the corrosion of A516 carbon steel in 0.5 M HCl media; the inhibition efficiency increases as the increasing in concentration of *A. platensis* extract with the maximum value of 65.9% at a concentration of 250 ppm at 298 K.
- Adsorption of the constituents molecules of *A. platensis* extract on A516 carbon steel surface is found to obey the Langmuir adsorption isotherm at 298 K.
- According to thermodynamic adsorption parameter, the adsorption mechanism of *A. platensis* extract on A516 carbon steel surface is dominated by physical adsorption.
- The analysis of the surface corroborates the weight loss and the electrochemical results, which shows that *A. platensis* extract is an effective green inhibitor to protect the surface of A516 carbon steel in 0.5 M HCl solution.

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