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Inhibitive impacts extract of *Citrus aurantium* leaves of carbon steel in corrosive media

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ABSTRACT

Relatively inexpensive, stable plant extract, namely *Citrus aurantium* leaves, was employed as highly efficient inhibitor of carbon steel corrosion by corrosive acid. The inhibition efficiency was estimated based on the weight loss method. Inhibition impacts of researched inhibitor increase with the concentration of the plant extract increase. The inhibition efficiency depends on three factors: molecular structure, concentration, and molecular weight of the inhibitor. Inhibition efficiency of 81.2% was achieved with 20% (v/v) of the extract in 1 M hydrochloric acid during 3 h at 25°C. The effect of temperature was also investigated and activation parameters were evaluated. Inhibition adsorption characteristics were approximated by Langmuir adsorption isotherm. Chemical adsorption mechanism was proposed for the studied inhibitor from the trend of inhibition performance and temperature degree in addition to activation energy values and heat of adsorption.

KEYWORDS

Citrus aurantium; carbon steel; hydrochloric acid solution; corrosion inhibition; adsorption



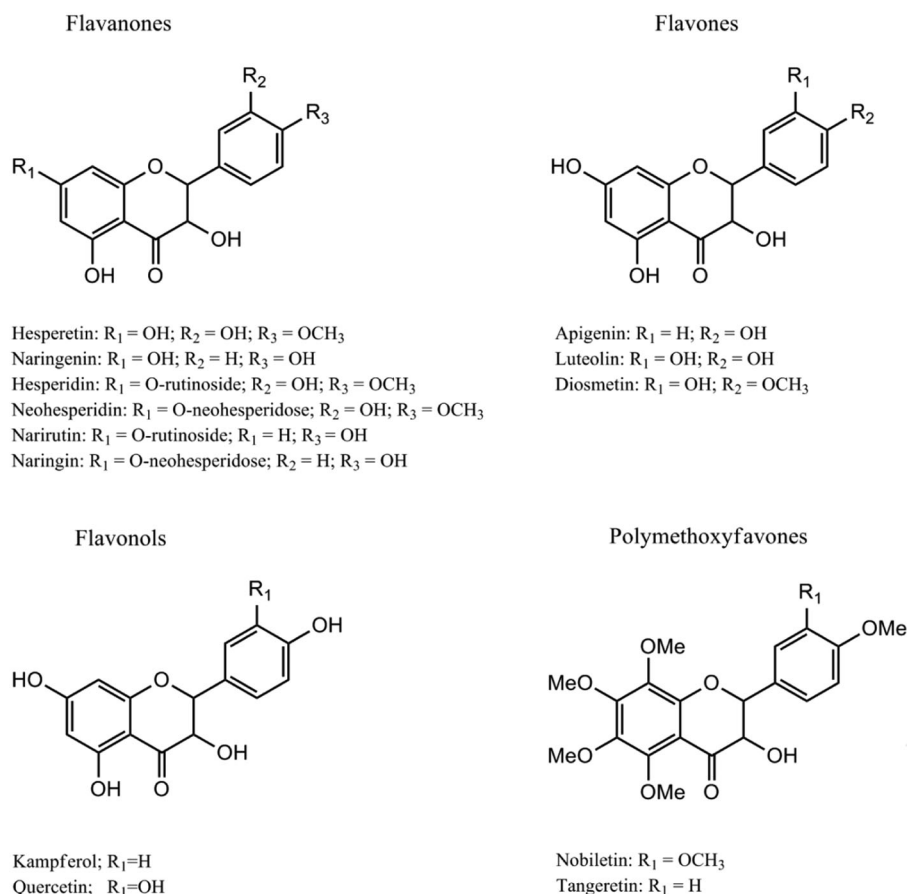
1. Introduction

Corrosion is an event that was considered either chemical or physical in nature, denatures the mineral characteristic, and of alloys make them inefficient for specified function (1–5). The main reason of the corrosion of alloys is their tendency to revert to the steady state. Most of the alloys are inherently unsteady and their natural trend to seek self-demolition through interacting with the environment to reach to lower energy state via producing metal compounds. These were the states that where most of the minerals were found (6). As there are many techniques for controlling the corrosion of alloys, the employ of inhibitor is the superior way for protecting alloys from corrosive solutions (7). Inhibitors were compounds that added in little quantities to corrosive solutions in order to prevent the reaction of surface of metal with the corrosive environment (8). They were added to considerable systems, such as cooling, refinery and gas

production units (9). Anticorrosion coatings are generally employed to inhibit the average of corrosion and increase the longevity of the mild steel. A broad range of organic adsorption inhibitors presently applied in the corrosion domain is expensive (10–16). Inhibitor adsorption on mild steel is affected by the nature of the mild steel, type of electrolyte, and molecular structure of the inhibitor (17–21). Inhibitor molecules are adsorbed on the surface of mild steel forming a barrier and consequently preventing reactions (cathodic or anodic) from processing at the surface of mild steel. These inhibitors could react with the iron atom at the mild steel surface to form inorganic complexes, blocking the surface of mild steel (22–26). Due to the high cost, toxicity, and hazardous of synthesized corrosion inhibitor (27–29), the examination of plant extract as corrosion inhibitors is presently interesting (30–32). Herein we focus on simple, practical, and economical corrosion inhibitors, so in this

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Scheme 1. Chemical structures of *C. aurantium* (35).

study, extract of *Citrus aurantium* leaves was used as green corrosion inhibitor for CS “Carbon Steel” in corrosive media.

2. Experimental

2.1. Specimen preparation

CS “Carbon Steel” of thickness 3×10^{-1} cm was obtained locally and was mechanically cut into coupons with dimensions of 65×10^{-1} cm, 25×10^{-1} cm, 3×10^{-1} cm having a hole with the diameter of 3.0 mm. First, coupons were polished, washed with distilled water, acetone and dried.

2.2. Preparation of plant extract

2.2.1. Collection of plant materials

Fresh *C. aurantium* leaves were obtained from Citrus orchards in Iraq and identified in College of Agriculture of the University of Basra, Basra City. The leaves were air-dried, grinded, and made into a fine powder using laboratory mortar and kept in a sterile air-tight container to avoid contamination.

2.2.2. Preparation of extract

Two hundred grams of dried leaf powder was dissolved in 1000 ml of ethanol for 48 h and centrifuged at 3000 rpm to enable paper diffusion of the active ingredients into the extraction medium. Filtration was later carried out using Whatman’s filter paper and the filtrate was stored at 5°C in a refrigerator.

2.3. Chemistry of *C. aurantium*

The chemical composition of *C. aurantium* includes vitamins, minerals, phenolic compounds, and terpenoids (33). The flavonoids contained in *C. aurantium* can be divided into four groups, including flavones, flavanones, flavonols, and anthocyanins (only in blood oranges) (Scheme 1) (34, 35).

Table 1. Corrosion parameters obtained from weight loss measurements.

Content	Weight loss (mg)	CR (mg cm ⁻² h ⁻¹)	IE (%)
0%	27.9	0.8348	–
5%	7.61	0.2491	70.1507
10%	7.13	0.2289	73.4285
15%	6.74	0.1930	77.5913
20%	5.92	0.1705	80.2070

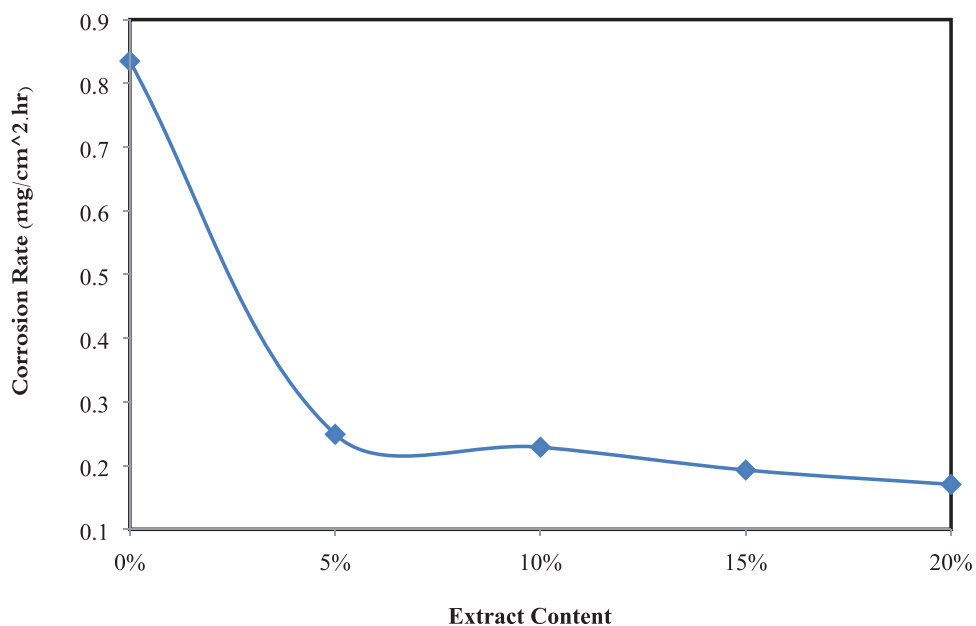


Figure 1. Variation of CR of CS without and with extract in corrosive media at 25°C.

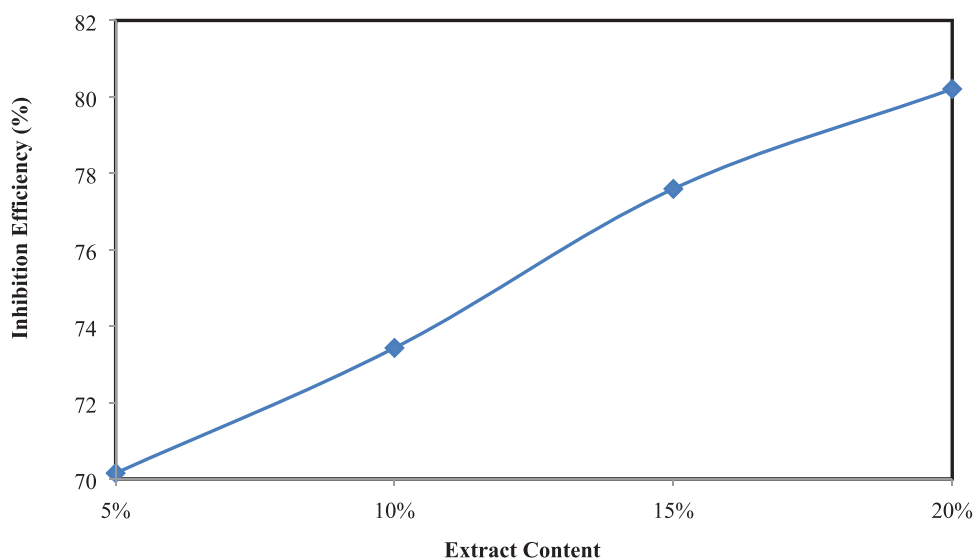


Figure 2. Variation of percentage corrosion IE with the content of the extract in corrosive media at 25°C.

3. Results and discussion

3.1. Impact of inhibitor concentration

CR "Corrosion Rate" of CS in 1 M of hydrochloric acid with different contents of the extract was calculated after immersion of 3.0 h, at 25°C. CRs and IEs "Inhibition Efficiencies" were listed in Table 1. Figures 1 and 2 show clearly that the CR decreases while IE increases with increasing extract content reaching the maximum value of 80.2 at 20% of the inhibitor. This behavior can be imputed to the adsorption of the extract used in this study on the surface of CS thereby inhibiting corrosion (36, 37).

3.2. Impact of temperature

The influence of degrees of temperature on the corrosion parameters namely, WL "Weight Loss", CR, and

Table 2. Corrosion parameters for CS in corrosive media without and with 40% of the extract at different temperatures.

Temperature	Weight loss (mg)	CR (mg cm ⁻² h ⁻¹)	IE (%)
25	27.9	0.8348	–
	5.92	0.1705	80.207
45	51.6	1.3927	–
	8.7	0.2386	82.8618
65	69.3	1.9965	–
	14.1	0.3239	83.7758

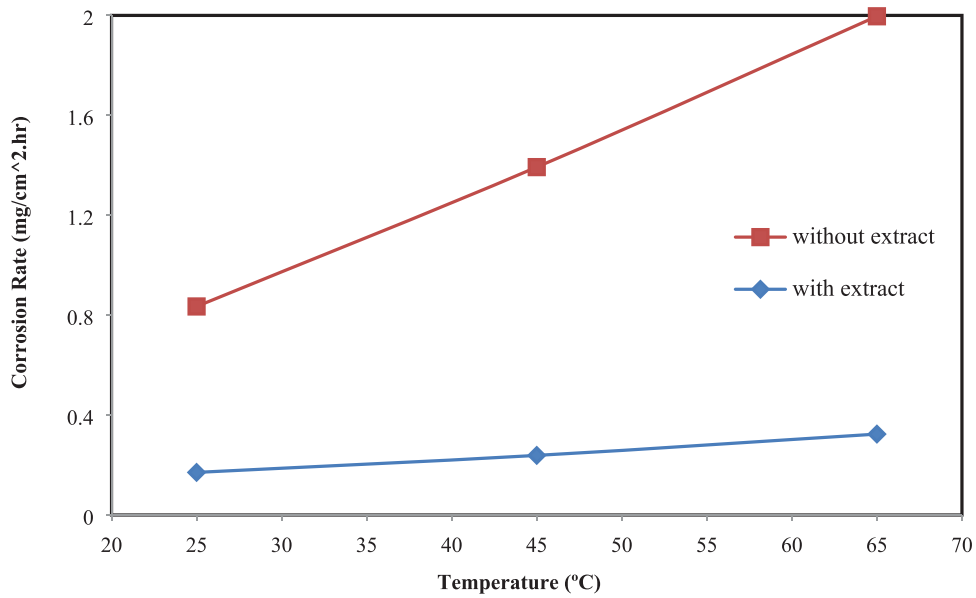


Figure 3. Plot of CR of CS in corrosive media in absence and presence of 40% of the extract versus studied temperature.

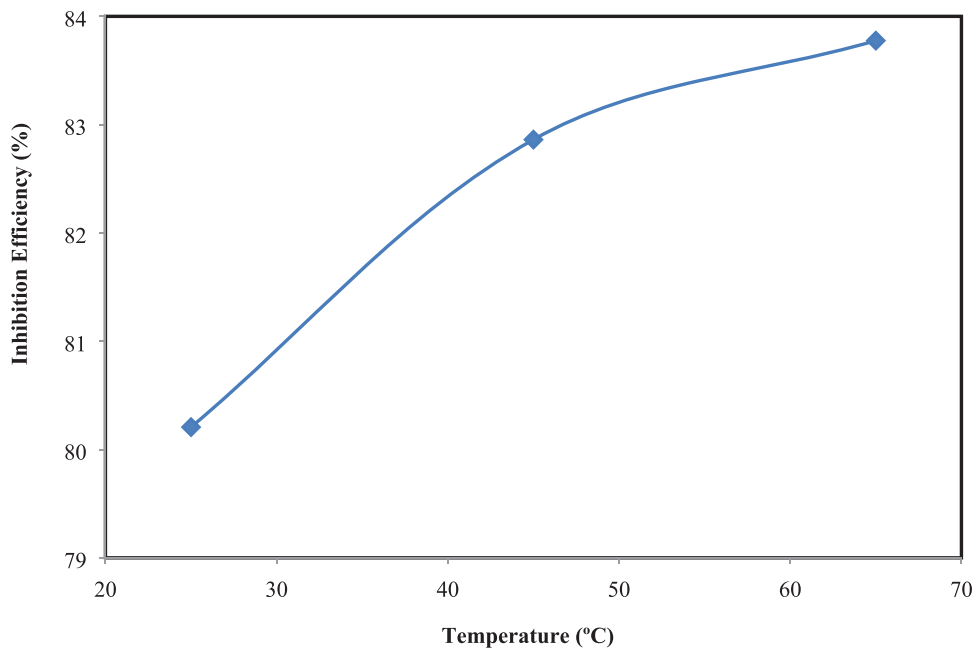


Figure 4. Plot of IE of the extract (40%) in corrosive acid versus temperature.

IE have been studied in corrosive media for 3.0 h, at temperatures 25°C, 45°C, and 65°C. The results (Table 2, Figure 3 and Figure 4) indicated that CR of CS without and with the extract increase with increasing in temperature, although corrosion rate is lowered in the presence of the extracts comparing with a blank solution, but the IE increases with increasing temperature. These results showed that the adsorption of the extract on the CS in corrosive solution is chemisorption (36, 38).

3.3. Activation parameters of the inhibition process

E_a "Activation energy" and enthalpy for the corrosion of CS in absence and presence of 40% (v/v) of the extract in 1 M hydrochloric acid were estimated using Arrhenius equations (1) and (2) (39, 40):

$$\log CR = -\frac{E_a}{2.303RT} + \log A, \quad (1)$$

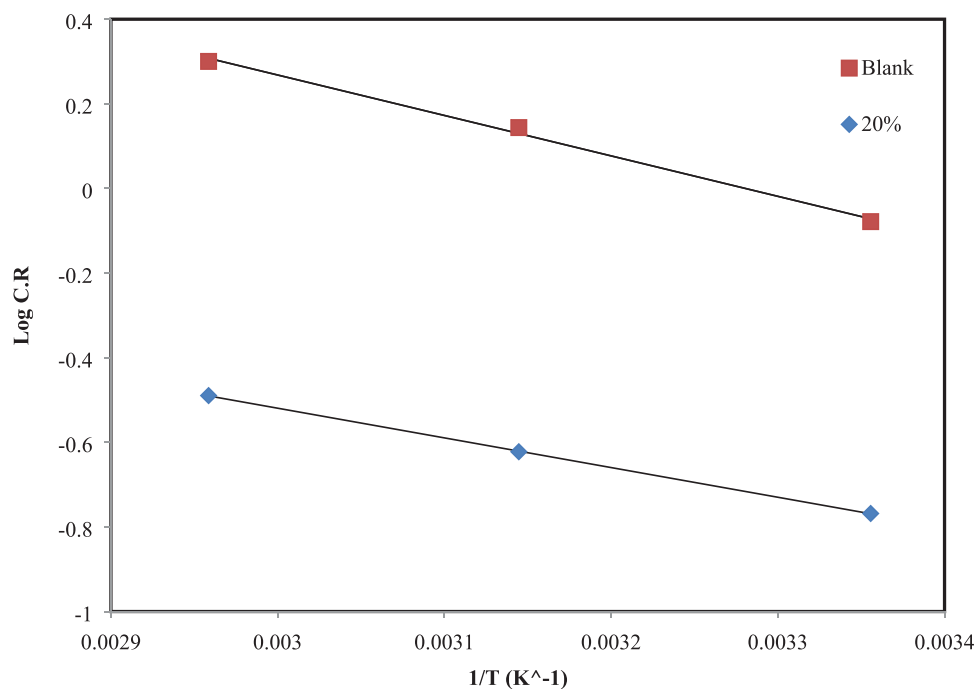


Figure 5. Plot of log CR against 1/T for CS in studied acid without and with 40% of the extract.

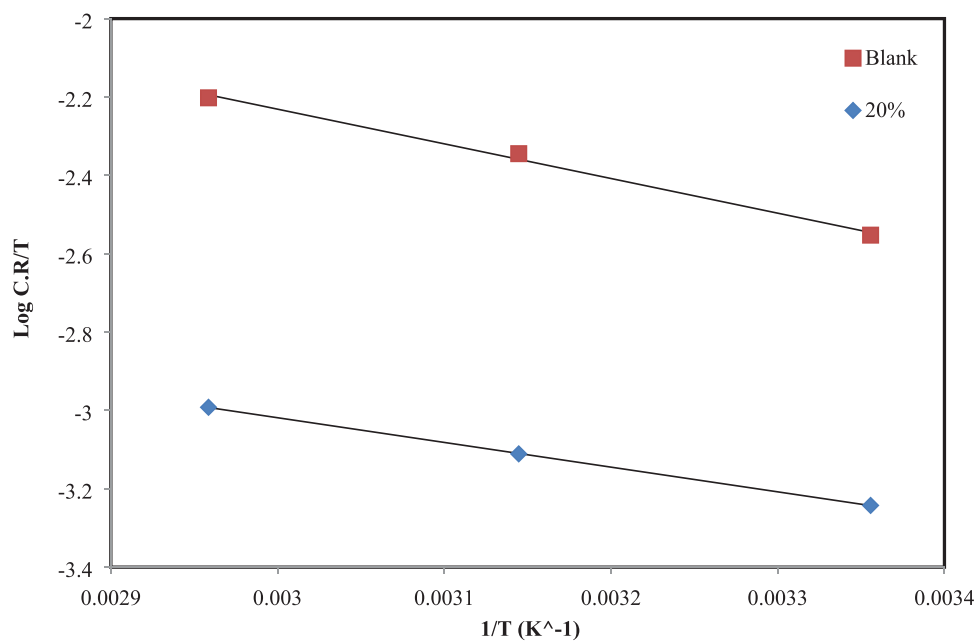


Figure 6. Plot of log CR/T against 1/T for CS in studied acid without and with 40% of the extract.

$$\log \frac{CR}{T} = \left[\log \frac{R}{Nh} + \frac{\Delta S_a}{2.303R} \right] - \frac{\Delta H_a}{2.303RT} \quad (2)$$

As shown in Figure 5, the CR of CS getting from WL measurements versus the temperatures 25°C, 45°C, and 65°C, with a slope of $-\Delta H_a/2.303R$. In addition, Figure 6 has a plot of log CR/T versus 1/T with a slope

of $-\Delta H_a/2.303R$ that the ΔH_a values were calculated. E_a and ΔH_a values are listed in Table 3.

Table 3. Activation parameters for the dissolution of CS in corrosive acid without and with 40% of the extract.

Content	E_a (KJ mol ⁻¹)	ΔH_a (KJ mol ⁻¹)
0.0	18.2855	16.9413
20%	13.4317	12.0722

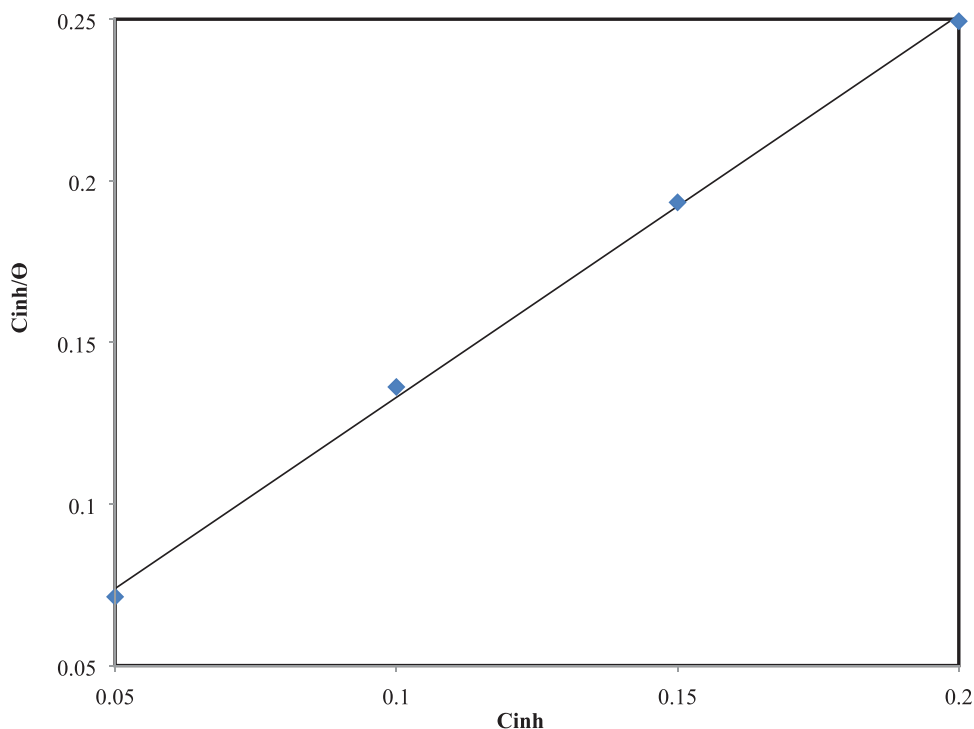


Figure 7. Langmuir isotherm for the adsorption of the inhibitor on the CS surface in corrosive media at 25°C.

3.4. Adsorption isotherm

Increasing of extract molecules that adsorb on the surface of CS and the interaction could have qualified through adsorption isotherm. The data have been tested graphically through appropriate to the Langmuir model. This model is given by (41, 42)

$$\frac{C_{inh}}{\theta} = \frac{1}{K} + C_{inh}. \quad (3)$$

By plotting values of C_{inh}/θ versus C_{inh} (Figure 7), a linear plot was obtained indicating that the adsorption of the studied plant extract as corrosion inhibitor was harmonious with the assumption of Langmuir adsorption isotherm and the slope obtained is close to unity.

4. Conclusion

Citrus aurantium leaves extract acts as efficient corrosion picking inhibitor on CS in 1 M corrosive media. The extract showed maximum IE of 81.2% at the presence of 20% in v/v during 3 h at 25°C. The IE% increased with increasing temperature. E_a decreases in presence of studied plant extract which indicates the chemisorption of the inhibitor molecules on the CS surface. *C. aurantium* leaves extract adsorbs on CS surface appropriate to the Langmuir isotherm.

Symbols

A	constant
CR	corrosion rate ($\text{mg cm}^{-2} \text{h}^{-1}$)
C_{inh}	content of the inhibitor
E_a	apparent activation energy ($\text{kJ mol}^{-1} \text{mol}^{-1}$)
H	Planks constant ($6.626 \times 10^{-34} \text{Js}$)
H_a	enthalpy of activation (kJ mol^{-1})
K	binding constant of the adsorption reaction
N	Avogadro's number ($6.022 \times 10^{23} \text{mol}^{-1}$)
R	gas constant ($8.314 \text{J mol}^{-1} \text{K}^{-1}$)
S_a	entropy of activation ($\text{J mol}^{-1} \text{K}^{-1}$)
T	temperature (K)
Θ	surface coverage

Disclosure statement

No potential conflict of interest was reported by the authors.

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