

Full Length Research Paper

Corrosion and corrosion inhibition of cast Iron in hydrochloric acid (HCl) solution by cantaloupe (*Cucumis melo*) as green inhibitor

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The effect of cantaloupe juice and seed extracts on corrosion of cast iron in 1.0 M hydrochloric acid (HCl) solution using hydrogen evolution measurements (HEM) and mass loss measurements (MLM) were investigated. Cantaloupe extracts inhibited the corrosion of cast iron in 1.0 M HCl solution. The inhibition efficiency increased with concentration of the extracts. The adsorption of the inhibitor molecules on cast iron surface was in accordance to Langmuir adsorption isotherms. In absence of inhibitors, the corrosion rate of cast iron increases with HCl concentration. The fractional reaction order observed in HCl solution indicates the formation of intermediates through the dissolution process or multiple steps mechanism of cast iron dissolution in HCl solution.

Key words: Cantaloupe (*Cucumis melo*), corrosion, cast iron, HCl concentrations, adsorption isotherm.

INTRODUCTION

The use of inhibitors is one of the best options of protecting metals against corrosion, especially green or eco-friendly inhibitors. Now, this field has been promising and effective, and it can be extracted by simple and inexpensive procedures. Comparisons have been made through the years between the toxic inorganic inhibitors such as; chromates, pomegranate, and cyanide, or synthetic organic compounds and the natural inhibitors, it observed that the natural inhibitors could potentially serve as an effective substitute for the corrosion inhibitors

without constituting risk for human health or the environment in which people live in (Shanableh, 2011). Many of these natural inhibitor substances can be extracted from different parts of plants: seed, fruit and leaves. Anyway, the plant extracts are considered as a rich source of environmentally acceptable corrosion inhibitors. For being that it can be extracted by simple procedures, which can keep the environment healthier with low cost and can be applied in various aggressive environments that make it a major importance in research

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Figure 1. Cantaloupe (*Cucumis Melo*).

and studies.

Acidic solutions are widely used in various industries for pickling ferrous alloys and steel. They are also used in oil and gas production to stimulate and increase the oil and gas flow to disqualify encrustations in production wells. Among various acids, the hydrochloric acid is mostly used for this purpose. Due to the extremely aggressive nature of acidic media, localized pitting corrosion starts to occur on the metal surface, over time, produces damage and destruction for products (Gadow and Fouda, 2014). In contrast, the particles of inhibitor are commonly used to reduce acid attack on the substrate metal by blocking active sites against deterioration.

Various natural plants are now used in many industries to protect steel in hydrochloric acid (HCl) solution, example, *Garcinia Mangostana* extract (Kumar et al., 2010), *Black pepper* extract (Damani et al., 2010), *Fenugreek* seed (Bouyanzer et al., 2010), Fennel (*Foeniculum Vulgare*) (Lahhit et al., 2011) and *Grap Pomace* (Rocha et al., 2012).

Cantaloupe (*Cucumis melo*) figure 1, a kind of muskmelon fruit belonging to the family Cucurbitaceae table 1, which is native to India and Africa. The unique aroma of cantaloupe is composed of many volatile compounds, biosynthetically derived from; fatty acids, carotenoids, amino acid and terpens (Nattaporn and Pranee, 2011; Milind and Kulwant, 2011). This article report the effect of cantaloupe juice and seed extracts as corrosion inhibitors of cast iron in 1.0 M HCl solution, using hydrogen evolution measurements (HEM) and mass loss measurements (MLM). In our knowledge, this is the first time that cantaloupe juice and seed extracts have been used as inhibitor of cast iron in HCl solution. Reinforced by the discussion of other study, common adsorption isotherms determine a process and nature of inhibitors adsorption, aim to choose the best adsorption isotherm curves that fit with experimental data (Figure 1 and Table 1).

Table 1. Scientific classification of cantaloupe (*Cucumis Melo*).

Kingdom	Plantae
Subkingdom	Tracheobionta
Super Division	Spermatophyta
Class	Mangoliophyta
Family	Cucurbitaceae
Genus	Cucumis L.
Species	<i>C. Melo</i> var. <i>Cantalupensis</i>

EXPERIMENTAL

Materials and solutions

Test was performed on cast iron specimen with weight percentage compositions in Table 2. The cast iron specimen was manufactured as cylindrical and purchased from ATTAIH Company, KSA. Before all measurements, the specimen was polished with a series of abrasive paper finding a coarse to remove roughness and rust. After that, the sample was washed by double-distilled water and acetone, and finally dried for weighted. The HCl solution was studied for Analar grade reagents. The solution was freshly prepared by double-distilled water in range (0.5 to 2.0 M) concentration by analytical dilution of stock solution (37%).

Cantaloupe extracts preparations

The juice extract of cantaloupe was obtained by putting fresh pulp for five cantaloupes in the blender, then filtered to get homogenous solution. While, the stock solution of seed extract was prepared by boiled weight grams of dried seed in 600 ml from double-distilled water for 90 min. The extract filtered and completed to 500 ml by double-distilled water. Both extracts kept freshly in refrigerator.

Gravimetric and volumetric measurements

The measurements were carried out by tow method; hydrogen evolution (HEM), and mass loss (MLM). Evolved H₂ was collected in a calibrated tube by downward displacement of water over time. The temperature was adjusted at room temperature 27°C by thermostat. The rates of HEM (R , ml/cm².min) and MLM (R_s , g/cm².min) were calculated as related in Equations (1) and (2), respectively (Mathur and Vasudaven, 1982; El-Etre, 2003):

$$R = \frac{\Delta V/A}{t} \quad (1)$$

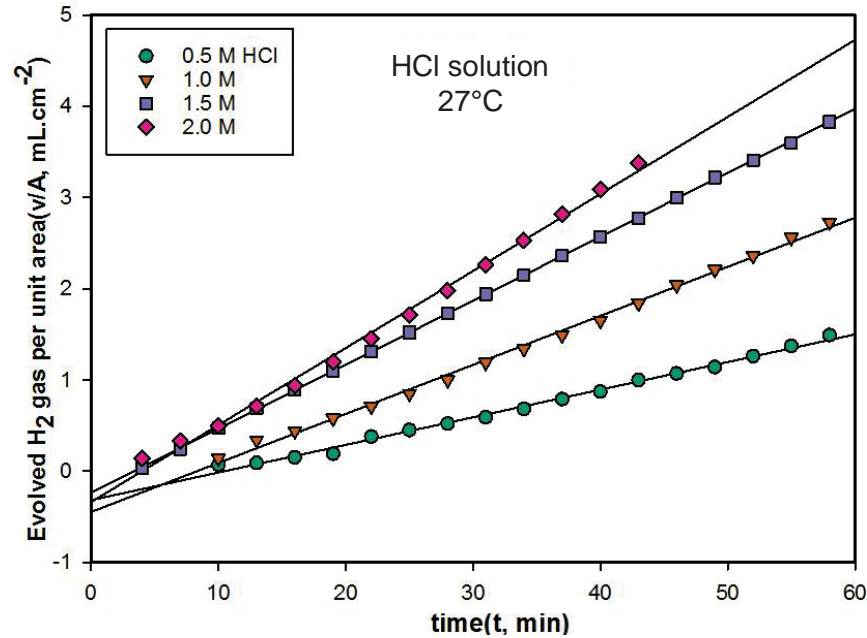
$$R_s = \frac{W_1 - W_2}{A t_f} \quad (2)$$

Where ΔV is the displacement of evolved gas, t is the time for evolved gas in minute, W_1 and W_2 are the mass of cast iron specimen before and after immersion in tested solution, respectively, t_f is the final time of experiment and A is the surface area of cylindrical specimen in cm².

The specimen was immersed in 1.0 M HCl solution in presence inhibitors at 27°C. The inhibition efficiency (%I_E) and degree of

Table 2. Chemical compositions of cast iron specimen with weight percentage (%W).

%C	%Si	%Mn	%P	%S	Remain
3.45 - 3.65	2.40 - 2.70	0.60 - 0.70	0.17 - 0.26	0.04 - 0.06	Fe

**Figure 2.** Volume of evolved H₂ per unit area versus exposure time for cast iron corrosion in various concentrations of HCl solutions at 27°C.

surface coverage (θ) were calculated from both HEM and MLM by Equations (3), (4), (5) and (6) respectively (Oguzie, 2007).

$$\%IE_{HEM} = \frac{R - R'}{R} \times 100 \quad (3)$$

$$\theta = \frac{R - R'}{R} \quad (4)$$

$$\%IE_{MLM} = \frac{R_s - R'_s}{R_s} \times 100 \quad (5)$$

$$\theta = \frac{R_s - R'_s}{R_s} \quad (6)$$

Where R and R_s are the HEM and MLM in the absence inhibitor, respectively. While R' and R'_s are HEM and MLM in presence inhibitor, respectively.

The corrosion rate for MLM (C.R, mmy) of the cast iron was also calculated by using the following (Equation 7) (Quraishi et al., 2009):

$$C.R. (mmy) = \frac{87.6W}{AtD} \quad (7)$$

Where W is the weight loss of the metal (mg), A is the surface area of the metal specimen (cm²), t is the exposure time (h) and D is the density of the metal (g/cm³).

RESULTS AND DISCUSSION

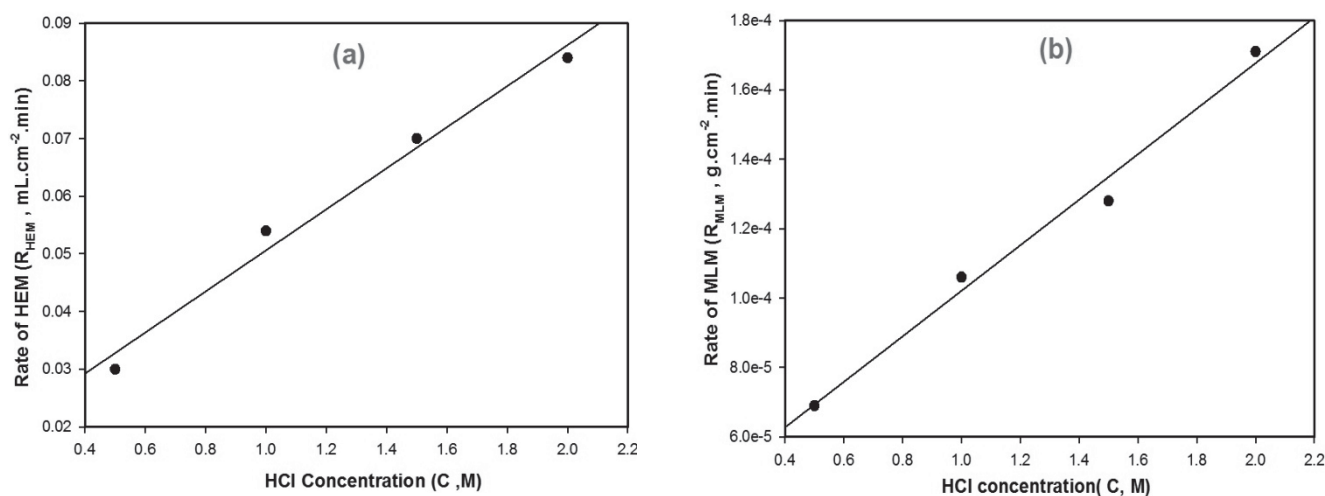
Behavior of cast iron (CI) corrosion in various concentrations of HCl solutions at 27°C

The data plotted for volume of evolved H₂ per unit area against time in minutes for 0.5 to 2.0 M of HCl concentrations at 27°C, is presented in Figure 2. The slopes of such lines were estimated in Table 3, taken as rates of cast iron corrosion reacted with HCl solution as corrosive environment using HEM. It is clear after inspection through duration experiment; the volume of evolved H₂ gas per unit area (V/A) increase upon increasing concentration.

The rates of corrosion of cast iron in various

Table 3. The corrosion rates of HEM & MLM for corrosion of cast iron in various concentrations of HCl solutions at 27°C.

HCl Concentration(M)	0.5	1.0	1.5	2.0
R _{HEM} (mL.cm ⁻² .min)	0.030	0.054	0.070	0.084
R _{MLM} (g.cm ⁻² .min)	6.90x10 ⁻⁵	1.06x10 ⁻⁴	1.28x10 ⁻⁴	1.71 x10 ⁻⁴
C.R. (mmy)	49.59	73.53	88.77	119.34

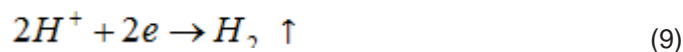
**Figure 3.** Rate (a) HEM (b) MLM of cast iron corrosion against various concentrations of HCl solutions at 27°C.

concentrations of HCl solutions resulted from HEM, constructed that by MLM after weight specimen; which were characterized by rapid effervescence, this influence is shown in Figure 3. The C.R. (mmy) of cast iron increase with increasing acid concentration, this indicates that cast iron corrosion in HCl is concentration dependent. It can also be observed from the Table 3 and Figure 3 a very good agreement between values of corrosion rates obtained from the three methods.

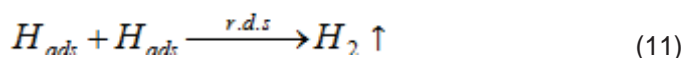
This result was expected, because with increased acidic concentration; both acidity and Cl⁻ anions concentration are increased too. This observation agrees with the fact that the rate of chemical reaction, diffusion, and ionization activates with increased concentration (Al-Turkustani et al., 2010).

The straight line shown in Figure 2 when a metal reacted with aggressive solution caused rapid reaction between acid, and air indicates a soluble passive layer (oxide film) formed on the surface of cast iron. As well, the presence of "induction period" at the beginning of the interaction (this obvious at 0.5 M) means dissolution of the formed oxide layer. It leads to none protection occurring on the surface and prevented solution from coming to the surface. This layer starts to fade rapidly with increase concentration of aggressive solution especially up to 2.0 M HCl solution. This concentration gives very good identity in linearity, the attack on oxide

film by Cl⁻ anions was instantaneous, forming local thinning passive layer on metal surface, over time create pitting localized corrosion (Al-Turkustani et al., 2010). Generally, the corrosion of iron in HCl solution revealed that it (Popova et al., 2005) takes place with hydrogen depolarization. The spontaneous dissolution of iron can be described by anodic dissolution reaction (Equation 8), accompanied by the corresponding cathodic reaction (Equation 9):



The corrosion of metals in acidic solution is cathodically controlled by the hydrogen evolution reaction which occurs in two steps (Equations 10 and 11) according to (Mathur and Vasudaven, 1982):



The rate determining step for the hydrogen evolution

Table 4. Kinetics parameters of the Mathur and Vasudevan model and conventional model.

Methods	Mathur and Vasudevan model			Conventional model		
	ln k	B	R2	lnk	n	R ²
HEM	-3.52	0.57	0.99	-2.88	0.60	0.99
MLM	-9.82	0.58	0.98	-9.15	0.63	0.98

reaction is the recombination of adsorbed hydrogen evolution reaction which the recombination of adsorbed hydrogen atoms form hydrogen molecules (Equation 11). Corrosion rate data as a function of acid concentration can be used to show the rate dependence of hydrochloric acid concentration by Mathur and Vasudevan model (Equation 12) and based on the kinetic equation (Equation 13) (Mathur and Vasudaven, 1982):

$$\ln R = \ln k + BC \quad (12)$$

$$\ln R = \ln k + n \ln C \quad (13)$$

Where R is the rate of metal dissolution, k is corrosion rate constant, B and n is the reaction order and C molar concentration of HCl solution.

The values of k , B and n obtained using HEM and MLM data and listed in Table 4. The fractional order observed in HCl solution may indicate the formation of intermediates through the dissolution process (Zaafarany, 2012), or multiple steps mechanism of cast iron dissolution in HCl solution.

Inhibition action of cantaloupe extracts as green inhibitor in cast iron corrosion

The corrosion rates for cast iron in 1.0 M HCl in absence and presence of cantaloupe extracts were determined by using HEM and MLM. Figure 4 shows the variations of evolved H_2 with time during the corrosion of cast iron in 1.0M HCl for various concentrations of cantaloupe extracts (a) juice and (b) seed at 27°C. The corresponding values of corrosion rates were given in Table 5 from slope of each line.

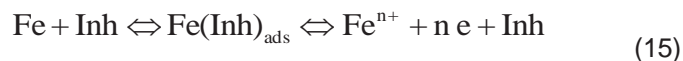
In comparison, blank solution (absence inhibitors) with added various concentrations of inhibitors (%v/v) of juice and seeds extracts; were noted with straight lines with much lower decreased rather than blank solution. The decline become even more with increased concentration of juice or seeds extracts. This indicates to oppositely occur when studied behavior of cast iron corrosion in HCl solution and the passive layer (adsorption film) formed presence inhibitor become insoluble, that inhibitors were first adsorbed onto the surface after impede corrosion process.

The adsorption of an organic adsorbate between

metal/solution interface can be represented as a substitutional adsorption process between the organic molecules in the aqueous solution $Org_{(soln)}$ and the water molecules on the metallic surface $H_2O_{(ads)}$ (Equation 14) (Bockris and Swinkels, 1964).



where $Org_{(ads)}$ are the organic molecules adsorbed on the metallic surface, $H_2O_{(sol)}$ is the water molecules in the aqueous solution and x is the size ratio representing the number of water molecules replaced by one molecule of organic adsorbate. According to (Bockris and Drazic, 1962) the inhibition mechanism could be explained by the $Fe(inh)_{ads}$ reaction as intermediates:



With further clarification, the $Fe(Inh)_{ads}$ did not have enough covered metal surface at low concentration of inhibitor, maybe because the added low concentration of inhibitors, or the rate adsorption is slow. So, the metal dissolution takes place on sites more than the formed $Fe(Inh)_{ads}$. Otherwise, at high concentration of inhibitor on the cast iron surface forms compact and coherent inhibitor over layer, then reducing chemical attack for metal (Branzoi et al., 2000; Singh and Quraishi, 2012).

The inhibitor efficiency of cantaloupe juice and seed extracts may be due to presence of many organic substance that acts as a good corrosion inhibitors, branched-chain and aromatic amino acid (Gonda et al., 2010; Nattaporn and Pranee, 2011; Milind and Kulwant, 2011). These compounds usually contain polar functions with heteroatoms such as nitrogen, oxygen, sulphur and phosphorus, and have triple or double bonds or aromatic rings. These groups are more a donor of electron and it offers itself the possibility to be a center of adsorption. The adsorption of these organic compounds by deferent centers of adsorption on the electrode surface makes a barrier for mass and charge transfers. This situation leads to a reduction in the double layer and a protection of the metal surface from the attack of the aggressive anions of the aggressive solution (Barouni et al., 2008; Emran et al., 2014).

The plot rates of corrosion HEM and MLM versus concentrations of juice or seeds extracts by (%v/v) at

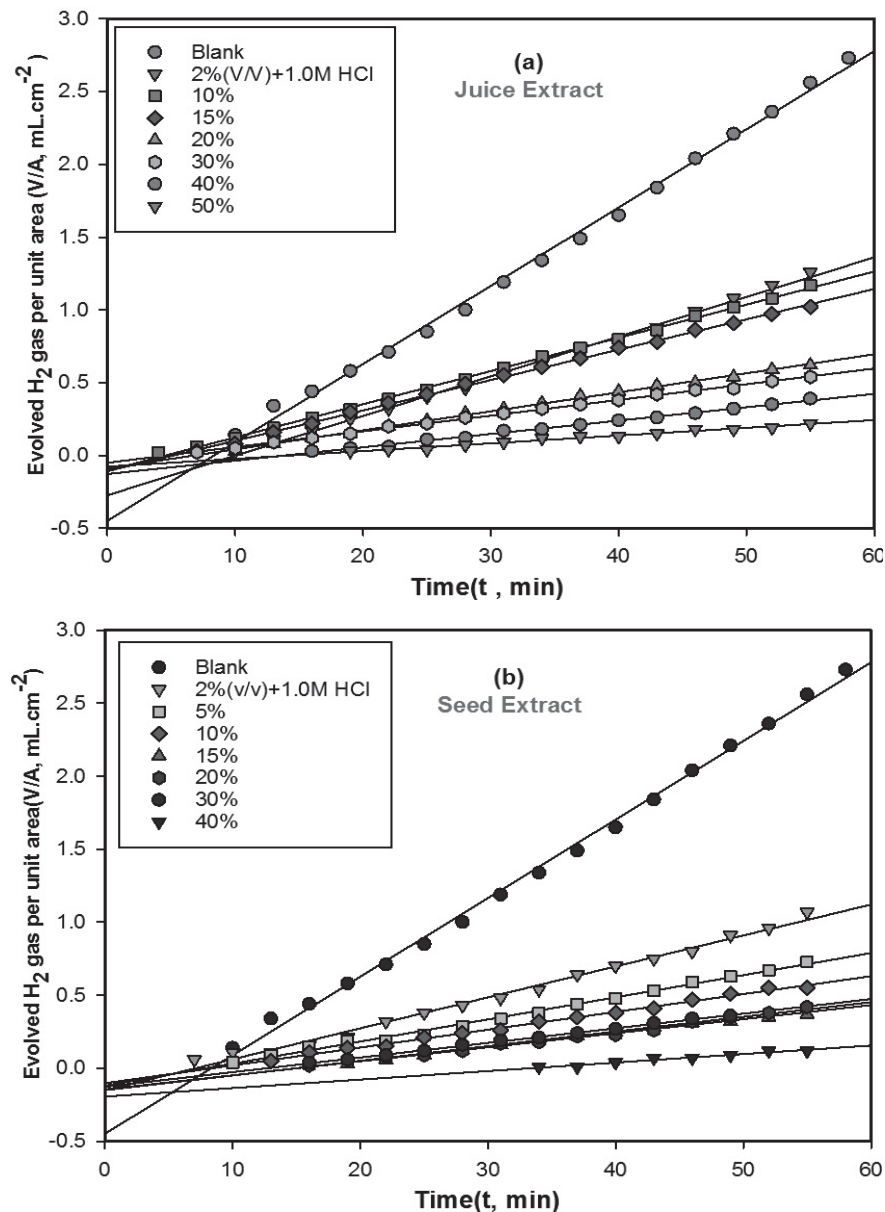


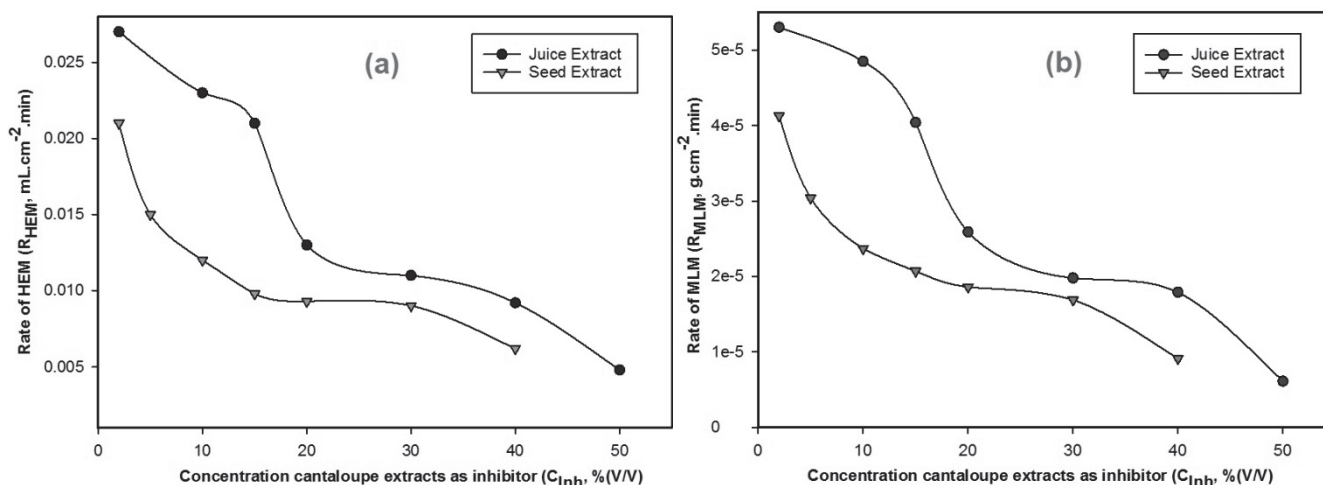
Figure 4. Variations of evolved H_2 for (a) juice (b) seed extracts of cantaloupe in 1.0 M HCl solution at 27°C.

27°C were presented in Figure 5. It is obvious that the added extracts into 1.0 M HCl solution caused noticeable reduction in amount rates obtained on cast iron surface. Seeds extract of cantaloupe has a great decrease in rate of HEM followed by MLM at different concentrations than juice extract. This is precisely what was interpreted in Table 5. 2 ml (%v/v) of juice and seeds extracts, significantly reduced the mass loss of cast iron with a factor 2 and 2.6 times respectively, and arched to 5.87 and 8.71 times at 40 (%v/v) compared with blank respectively. The values of inhibition efficiency of % IE_{HEM} , % IE_{MLM} degree of surface coverage (θ_{HEM}) and

θ_{MLM} were listed in Table 5. The surface coverage and inhibition efficiency values increase with increasing extract concentration (Figure 6). The maximum inhibition efficiency %IE value of 91.11 and 91.16% for juice extract at 50%(v/v), whilst in seeds extract were 91.30% and 88.5% at 40%(v/v) by using MLM and HEM at 27°C, respectively. This is due to the blocking active sites on metal surface and decreasing the effective area of corrosion attack by adsorption of effect compounds present in cantaloupe, like; Vitamin, Phenolic compounds, Terpenoids etc. (Nattaporn and Pranee, 2011; Milind and Kulwant, 2011).

Table 5. Effect of inhibitors on the corrosion of cast iron in 1M HCl solution by using HEM and MLM at 27°C.

Inhibitor concentration %(v/v)	HEM			MLM			
	R_{HEM} ($ml.cm^{-2}.min$)	$\%IE_{HEM}$	θ_{HEM}	R_{MLM} ($g.cm^{-2}.min$)	$\%IE_{MLM}$	θ_{MLM}	
Blank (1.0 M HCl)	0.054	-	-	1.06×10^{-4}	-	-	
Juice extract	2%	0.027	50	5.30×10^{-5}	50	0.50	
	10%	0.023	57.41	4.85×10^{-5}	54.25	0.54	
	15%	0.021	61.11	4.04×10^{-5}	61.89	0.62	
	20%	0.013	75.93	2.59×10^{-5}	76.04	0.76	
	30%	0.011	79.63	1.98×10^{-5}	81.32	0.81	
	40%	9.20×10^{-3}	82.96	0.83	1.79×10^{-5}	83.11	0.83
	50%	4.80×10^{-3}	91.11	0.91	6.13×10^{-6}	94.22	0.94
Seeds extract	2%	0.021	61.11	4.13×10^{-5}	61.04	0.61	
	5%	0.015	72.22	3.04×10^{-5}	71.32	0.71	
	10%	0.012	77.78	2.37×10^{-5}	77.64	0.78	
	15%	9.80×10^{-3}	81.85	0.82	2.07×10^{-5}	80.47	0.80
	20%	9.30×10^{-3}	82.78	0.83	1.86×10^{-5}	83.11	0.83
	30%	9.02×10^{-3}	83.30	0.83	1.69×10^{-5}	84.06	0.84
	40%	6.20×10^{-3}	88.50	0.89	9.12×10^{-6}	91.39	0.91

**Figure 5.** Plots (a) R_{HEM} (b) R_{MLM} via concentrations (%v/v) of cantaloupe extracts in 1.0 M HCl solution at 27°C.

Adsorption isotherm and adsorption parameters

Adsorption isotherms are usually used to describe the adsorption process. The most frequently used isotherms include: Langmuir, Temkin and Flory-Huggins. The adsorption isotherm provides important clues regarding the nature of the metal inhibitor interaction, and inhibitor molecules adsorb on the metal surface if the interaction between molecule and metal surface is higher than that of the H_2O molecule and the metal surface (Shukla and Ebenso, 2011). Langmuir isotherm was tested for its fit to the experimental data according to Equation (16)

(Langmuir, 1917; Christov and Popova, 2004).

$$\log \frac{C}{\theta} = -\log K + \log C \quad (16)$$

Where C is the concentration of inhibitor, K is the adsorptive equilibrium constant, θ is the surface coverage.

Langmuir isotherm given band is represented in Figure 7, and listed in Table 6. Where plots of $\log C/\theta$ versus $\log C_{inh}$, for juice and seed extracts were found straight lines with a good square correlation coefficient (R^2)

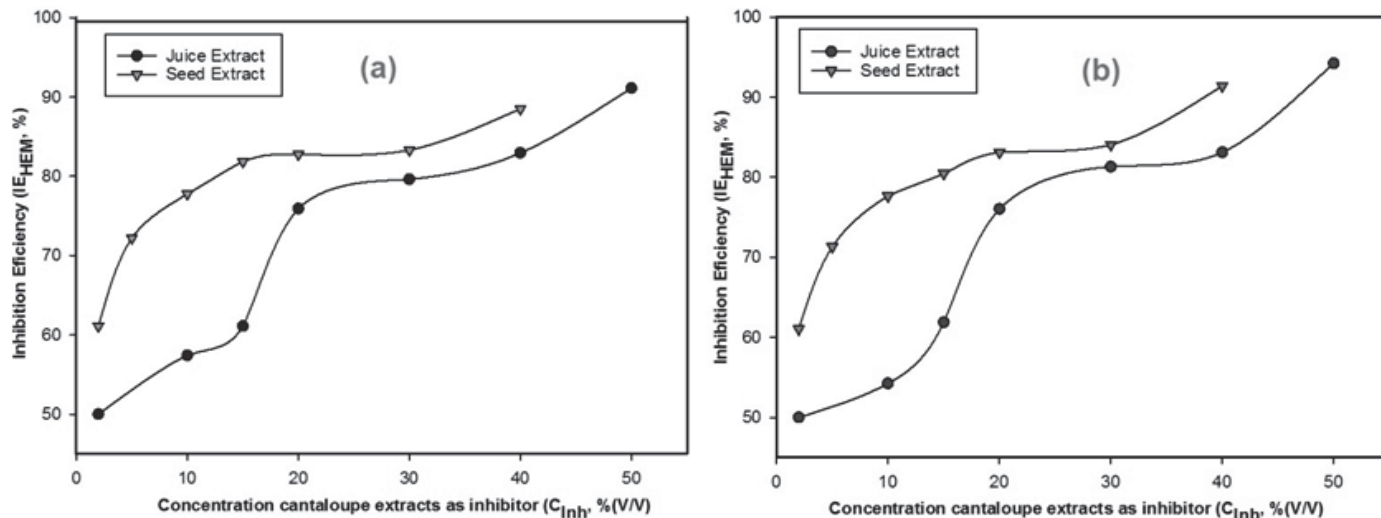


Figure 6. Variations of the inhibition efficiency for (a) HEM (b) MLM with concentrations of inhibitors % (v/v) in 1.0 M HCl solution at 27°C.

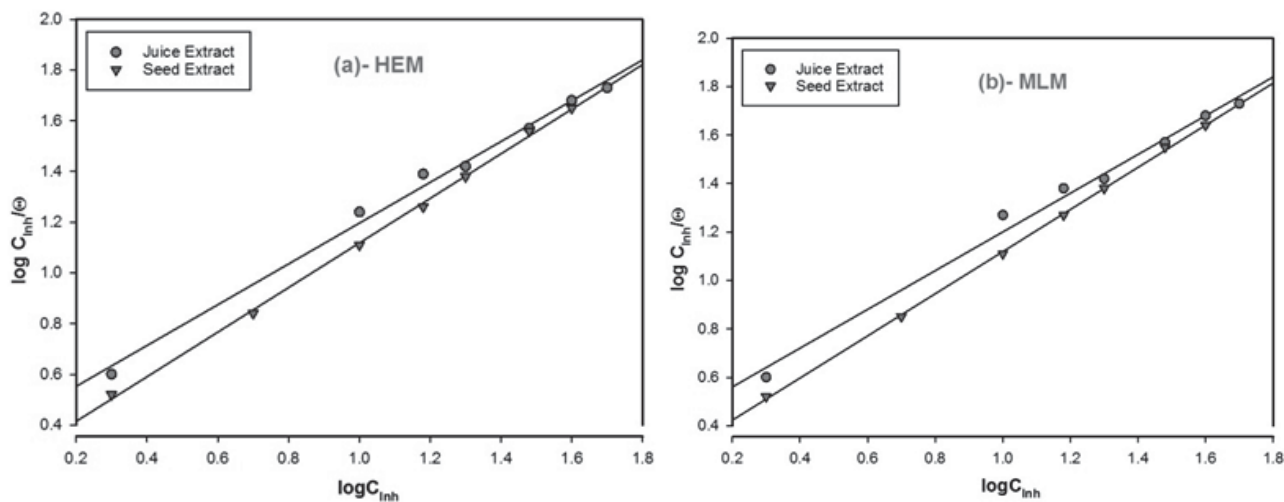


Figure 7. Langmuir adsorption isotherm for cast iron in 1.0M HCl solution of cantaloupe extracts as inhibitors by using (a) HEM (b) MLM at 27°C.

Table 6. Adsorption parameters for adsorption of cantaloupe extracts on cast iron surface under effect 1M HCl solution by using HEM MLM at 27°C.

Isotherm and extracts		R ²	HEM		R ²	MLM	
			K	slope		K	slope
Langmuir	Juice	0.992	0.41	slope= 0.80	0.989	0.40	Slope= 0.80
	Seed	0.999	0.58	slope= 0.87	0.999	0.56	slope= 0.87
Temkin	Juice	0.845	15.48	a= -3.90	0.816	11.48	a= -3.69
	Seed	0.961	741.31	a= -5.80	0.976	416.87	a= -5.45
Flory-Huggins	Juice	0.664	4.57	x= 1.18	0.576	0.15	x= 0.90
	Seed	0.938	2.04	x= 2.17	0.895	1.29	x= 1.85

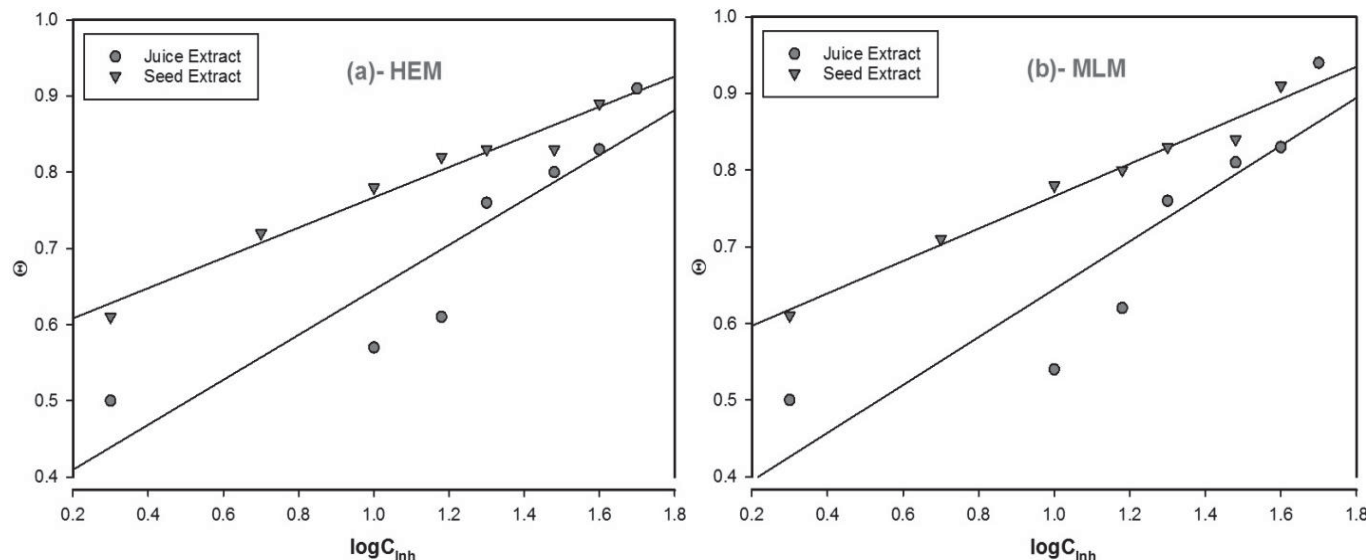


Figure 8. Temkin adsorption isotherm for cast iron in 1.0 M HCl solution of cantaloupe extracts as inhibitors by using (a) HEM (b) MLM at 27°C.

0.992, 0.999 for HEM, and 0.989, 0.999 for MLM, respectively. The slopes lines for each method and extracts are arched unity, it is assumed that the inhibition of cast iron corrosion in 1.0 M HCl by cantaloupe extract occurs by monolayer adsorption at appropriate sites on the metal, the metal surface contains a fixed number of adsorption sites and each site holds one adsorbate, and no interaction between adsorbate molecules. From the intercepts of the straight lines $\log C/\theta$ axis for juice and seeds extracts, K value calculated were 0.41 and 0.58 Lmol^{-1} for HEM, and 0.40 & 0.56 Lmol^{-1} for MLM, respectively. For Temkin adsorption isotherm, the degree of surface by using HEM and MLM is related to logarithmic inhibitor concentration (C) according to (Equ. 17) (Christov and Popova, 2004):

$$\theta = -\frac{2.303}{2a} \log K - \frac{2.303}{2a} \log C \quad (17)$$

where K is the adsorption equilibrium constant and (a) is the attractive parameter. Plots of θ against $\log C$, as presented in Figure 8 gave linear relationship, which shows that adsorption data fitted Temkin adsorption isotherm at seed extract ($R^2 = 0.961$ and 0.976) for HEM and MLM more than juice ($R^2 = 0.845$ and 0.816) for HEM and MLM, respectively. Adsorption parameters obtained were recorded in Table 6. The values of interaction parameter (a) was negative in all cases, which indicate that repulsion exists in the adsorption layer. Flory-Huggins adsorption isotherm can be expressed according to (Equation 18) (Christov and Popova, 2004):

$$\log \left(\frac{\theta}{C} \right) = \log K + x \log (1 - \theta) \quad (18)$$

Where x is the number of inhibitor molecules occupying one site, or the number of water molecules replaced by one molecule of the inhibitor. The value x substituted by a given inhibitor molecule adsorbed surface by plots of $\log (\theta/C)$ against $\log (1 - \theta)$ by using HEM and MLM, were shown in Figure 9. The values of the size parameter x are positive 1.18 and 2.17 of HEM, 0.90 and 1.85 of MLM for juice and seed cantaloupe extracts, respectively as shown in Table 6. The values of $x \approx 1$ for juice extract, implied that one inhibitor molecule replaces one water molecule, while, the values of $x > 1$ for seeds extract, means that one inhibitor molecule replaces more than one water molecule. The x obtained for the seeds extract were higher than those obtained for the juice extract, suggesting that the adsorption behavior of the seeds extract is better than that of the juice extract in Flory-Huggins isotherm. According to the fit experimental data (R^2), the better adsorption isotherm of juice and seeds extracts of cantaloupes is Langmuir isotherm.

Conclusion

The results obtained from HEM and MLM of corrosion and corrosion inhibition of cast iron in various concentrations HCl solutions (0.5-2.0 M), under 27°C; can be deduced:

- (1). The corrosion rate of cast iron increase with increase concentration of HCl solution.
- (2). Juice and seed extracts of cantaloupe acts as good natural inhibitor for corrosion of cast iron in 1.0 M HCl solution.

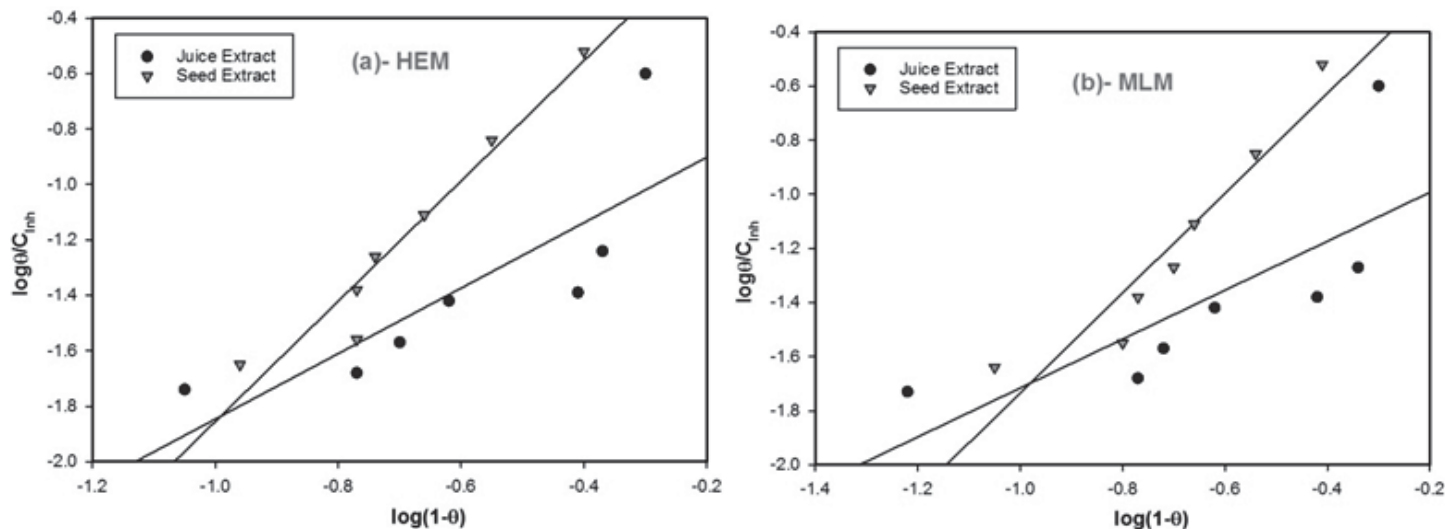


Figure 9. Flory-Huggins adsorption isotherm for cast iron in 1.0M HCl of cantaloupe extracts as inhibitors by using (a) HEM (b) MLM at 27°C.

(3). The inhibition efficiency increase with increase concentration of inhibitors % (v/v), with maximum value obtained in juice extract 91.11 and 94.22 at 50% (v/v), while in seed extract 88.50 and 91.39 at 40% (v/v) for HEM and MLM, respectively.

(4). Seed extract good natural inhibitor than juice extract of cantaloupe.

(5). The square correlation coefficient (R^2) was used to choose the adsorption isotherm that fits experimental data. The adsorption of cantaloupe juice and seed extracts molecules on cast iron surface in 1.0M HCl follows Langmuir adsorption isotherm.

Conflict of Interest

The authors have not declared any conflict of interest.

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