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Cantaloupe Extracts as Eco Friendly Corrosion Inhibitors for Aluminum in acidic and alkaline solutions

Khadijah M. Emran[a]*, Najla M. Ahmed[b], Bayan A. Torjoman[b], Arwa A. Al-Ahmadi[b], Sara N. Sheekh[b]
[a*] *Chemistry Department, Faculty of Science, Taibah University, Saudi Arabia (kabdalsamad@taibahu.edu.sa).*
[b] *Chemistry Department, Applied Chemistry, Faculty of Science, Taibah University, Saudi Arabia*

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Corresponding : Dr Emran K.M.: k_and_e154@hotmail.com

Abstract
The corrosion rate of aluminum alloy at deferent concentrations of HCl and NaOH was deduced by chemical methods. It was found that dissolution rate of aluminum alloy depends on the concentration of corrosive media. The inhibition efficiency of *Cantaloupe juice and seed extracts* in 1.0M HCl was higher than that in 1.0M NaOH solutions. In both acidic and basic media, the increase in Cantaloupe extracts resulted in a increase both of the inhibition efficiency and the degree of surface coverage. Good corrosion inhibitor properties of cantaloupe extracts are due to presence of many organic substances. The adsorption of these organic compounds on the aluminum surface makes a barrier for mass and charge transfers. The adsorption of *Cantaloupe juice and seed extract* molecules on aluminium metal surface in 10.M HCl obey Langmuir's adsorption isotherm. In case of base media, the adsorption *Cantaloupe juice* obey Langmuir's isotherm and Temkin isotherm was fit to *Cantaloupe seeds*.

Keywords: Corrosion, Aluminium Acid concentration Alkaline concentration, cantaloupe (Cucumis Melo), chemical methods

1. Introduction
Aluminium and Aluminium alloys represent an important category of materials and find a number of applications due to their desirable characteristics, high technological value and these usages expose them to environments that could be acidic or alkaline. Many reports have been published concerning the corrosion of unprotected aluminium and its alloys in varied environments and mineral acids.

Acid and alkaline solutions are generally used for the remove of undesirable scale and rust in several industrial process. During this process metal loss occurs due to the dissolution of the metals in acids and alkaline solutions. In order to avoid metal loss and for reducing solutions consumption many organic compounds are used as corrosion inhibitors. Thus, organic and inorganic compounds are usually added to alkaline and Acid media as corrosion inhibitors in industrial processes such as alkaline cleaning, pickling and etching, and to improve efficiency in devices such as aluminum alkaline battery [1,2]; but some of these inhibitors are toxic, expensive and non-biodegradable.

There is a growing demand for corrosion inhibitors that are less toxic and biodegradable compared to the current formulations. Green inhibitors displaying substantially improved environmental properties will be the inhibitors most widely used in the future. Many studies the past two decades foxes on of green corrosion inhibitors for Aluminium and Aluminium alloys corrosion inhibition as cheap and effective molecules at low or zero environmental impact in acid as *mucilage extracted* [3], *exudate gum* [4], *root of ginseng* [5], *henna (Lawsonia inermis)* [6], *Aloe extract* [7] and *Melia azedarach* [8]. Also in alkaline media *damsissa (Ambrosia maritime, L.) [9], Phyllanthus amarus extract [10], Euphorbia hirta and Dialum guineense leave extracts [11], Brahmi (Bacopa monnieri) [12], L. varius l,[13], plumbago europaea [14], Vitex negundo* [15] are used as eco-friendly inhibitors.

In present study, the effect of *cantaloupe(Cucumis Melo) juice and seeds extracts* on both the corrosion and kinetics of corrosion process of aluminum in both 1.0M NaOH solution and 1.0M HCl has been investigated using weight loss and hydrogen evaluation measurements. To our knowledge this is the first time that cantaloupe juice and seeds extracts has been used as inhibitor for aluminum metal in aggressive media.

2. Materials and methods

2.1 Specimen

Aluminum alloy (32177) was cut cylindrical. This is to allow for easy polishing to remove roughness and rust. The chemical analysis of aluminum alloy composed of different elements (wt%) given in **Table-1**. The specimen bars were manually polished by a series of abrasive paper (emery paper) until mirror finish. Than the aluminum alloy sample degreased with acetone (A.R), washed in double distilled water and dried in warm air and weighted.

Table 1: Composition of the Aluminum sample(W%).

Mg	Si	Mn	Fe	Cu	Cr	Ti	Zn	rest
0.75	1.06	0.62	0.33	0.070	0.007	0.055	0.020	Al

2.2 Test solutions

Different concentrations of HCl acid (0.5, 1.0 and 1.5 M) and NaOH (0.5, 1, 1.5, 2 M) were used. All test solutions were prepared with analytical grade reagents (A.R.) and double distilled water. Inhibition efficiency of cantaloupe extracts (juice in range 2-50% v/v and the seed in range 2-40 % v/v) was performed in 1.0 M HCl and 1.0 M NaOH. All experiments were conducted at 27°C.

2.4 Preparation of extract

The Cantaloupe juice extract was obtained directly from put of the fresh pulp of five Cantaloupes in a blender, then filtered to get homogenous solution. Stock solutions of the seed extract were prepared by boiling weighed grams of dried Cantaloupe seeds in 600 ml of double distilled water for 90 min. The extract is filtered and completed to 500 ml with double distilled water in measuring flask. The freshly prepared extract kept in refrigerator.

2.5 Chemical measurements

The chemical measurements were carried out by two methods; hydrogen evolution method (HEM) and mass loss method (MLM) using Mylius cell. The pre-weighed aluminum specimen was immersed in test solutions without and with different concentrations (% v/v) of the extracts at 27°C. The corrosion rates (R_{MLM}) were calculated from Eq.(1):

$$R_{MLM} = \frac{W_1 - W_2}{At_f} \qquad (1)$$

W_1 is the weight before metal corrosion, W_2 is the weight after metal corrosion, A is the specimen surface area, A(cm^2) and time, t(min) the immersion period in mint from the corrosion rate. The surface coverage (θ) as a result of adsorption of inhibitor molecules, and inhibition efficiencies (EI%) of the extract were determined using Eqs. (2&3), respectively.

$$\theta = \frac{R_{MLM} - R_{MLM(inh.)}}{R_{MLM}} \qquad (2)$$

$$\%EI = \frac{R_{MLM} - R_{MLM(inh.)}}{R_{MLM}} x100 \qquad (3)$$

where R_{MLM} and $R_{MLM(inh.)}$ are the corrosion rate in the absence and presence of the inhibiting molecules, respectively.

Gasometric technique is based on the principle that corrosion reactions in aqueous media is characterized by the evolution of gas resulting from the cathodic reaction of the corrosion process, which is proportional to the rate of corrosion [16]. The rate of evolution of the hydrogen gas (R_{HEM}) is determined from the slope of the graph of volume of gas evolved (V) per surface area (A) versus time (t), according to Eq.(4):

$$\mathrm{RVH} = \frac{\Delta V}{\Delta t} \qquad (4)$$

and the inhibitor surface coverage (θ) and efficiencies (%EI) determined using Eq.(5&6), respectively.

$$\theta = \frac{R_{HEM} - R_{HEM(inh.)}}{R_{HEM}} \qquad (5)$$

$$\%EI = \frac{R_{HEM} - R_{HEM\,(inh.)}}{R_{HEM}} x100 \quad (6)$$

where R_{HEM} and $R_{HEM(inh.)}$ are the rate of hydrogen evolution in the absence and presence of the extract, respectively. The data presented here represents the average of two to three measurements from the weight loss and gasometric techniques.

3. Results and discussion

3.1 The effect of HCl and NaOH concentration on aluminum corrosion rate

The influence of HCl (0.5, 1.0 and 1.5 M) and NaOH (0.5, 1.0, 1.5, 2.0 M) concentrations on aluminum corrosion at 27 °C is shown in **Fig-1,a,b.**

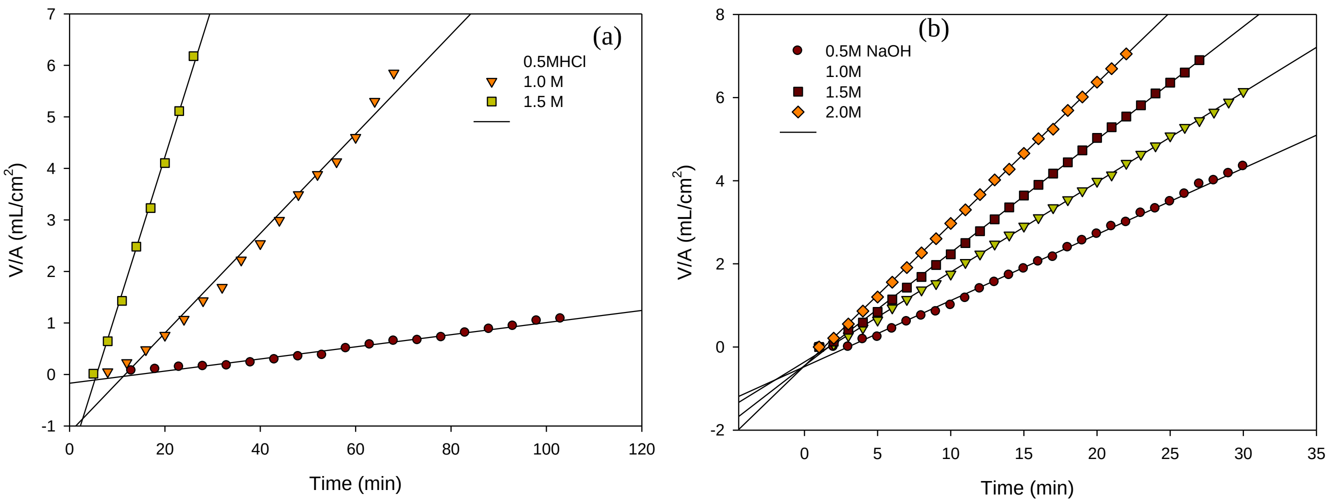

Figure 1: Volume of hydrogen evolution / time curves for the corrosion of aluminum in different concentrations of (a) HCl and (b) NaOH at 27°C.

As the concentration of the HCl or NaOH solutions increases, the slop of the straight lines also increases, indicating an increase in the dissolution of aluminum, e.g. increases of corrosion rate. An induction period was observed especially at low concentrations of HCl due to the slow reaction between the acid and the air formed oxide film on aluminum surface. The straight lines in **Fig.(1)** is due to the absence of any insoluble formed layer on the surface of the metal through the process of corrosion and indicates that aluminum corrosion in HCl or NaOH is concentration dependent. The aggressivity of NaOH on aluminum alloy is clearer by comparison of corrosion rate in HCl. The corrosion rate, R_{HEM}, which is obtained from the slope of the linear part of the gasometry plots for the test solutions and corrosion rates obtained from the weight loss technique, R_{MLM}, are shown in **Table-2**.

Table 2: Corrosion rates for aluminum in different concentrations of HCl and NaOH at 27 °C.

Corrosion Rate	HCl (M)			NaOH (M)			
	0.5	1.0	1.5	0.5	1.0	1.5	2.0
R_{HEM} (ml.cm^{-2}min)	0.012	0.0996	0.295	0.159	0.216	0.271	0.340
R_{MLM} (g.cm^{-2}min)	6.72×10^{-6}	7.37×10^{-5}	1.68×10^{-4}	1.18×10^{-4}	1.48×10^{-4}	1.81×10^{-4}	2.24×10^{-4}

Ali [8] reveled that corrosion of aluminum in aqueous solution depends on the concentration of anions in solution. A general mechanism for the dissolution of aluminum as follows:

$$Al_{(s)} + H_2O \rightarrow AlOH_{ads} + H^+ + e \quad (7)$$

$$AlOH_{ads} + 5H_2O + H + \rightarrow Al^{3+} + 6H_2O + 2e \quad (8)$$

$$Al^{3+} + H_2O \rightarrow [AlOH]^{2+} + H^+ \quad (9)$$

$$[AlOH]^{2+} + X^- \rightarrow [AlOHX]^+ \quad (10)$$

The controlling step in the metal dissolution is the complexation reaction between the hydrated cation and the anion, Eq. (10). In the presence of chloride ions the reaction will correspond to:

$$[AlOH]^{2+} + Cl^- [AlOH]^{2+} + Cl^- \rightarrow [AlOHCl]^+ \quad (11)$$

Thus soluble complex ion formed leads to the dissolution of the metal.

The corrosion rate of aluminium in alkaline solution is under anodic control [17,18] which is:

$$Al + 4OH^- \rightarrow Al(OH)_4^- + 3e \quad (12)$$

The relation between corrosion rate and acid concentration can be illustrated by the kinetic Eq.(13)

$$\log R = \log A + n \log C \quad (13)$$

where A represents corrosion rate constant which represents the rate of metal dissolution (corrosion), n reaction order and C molar concentration of HCl or NaOH. The relationship log R_{MLM} and/or log R_{HEM} vs. log C gave a straight lines as in **Fig-2,a,b**. The slopes of these lines represent the reaction order (n) and the intercept is log A. It was found that the values of n are equal about 3 and 0.5 for HCl and NaOH respectively. This indicating that corrosion of aluminum in HCl solution is third order reaction which is agreement with privios study of Al-Turkustani et al.[7]. In NaOH solution is the 0.5 order reaction. The fractional order observed in NaOH solution may indicate the formation of intermediates through the dissolution process [19].

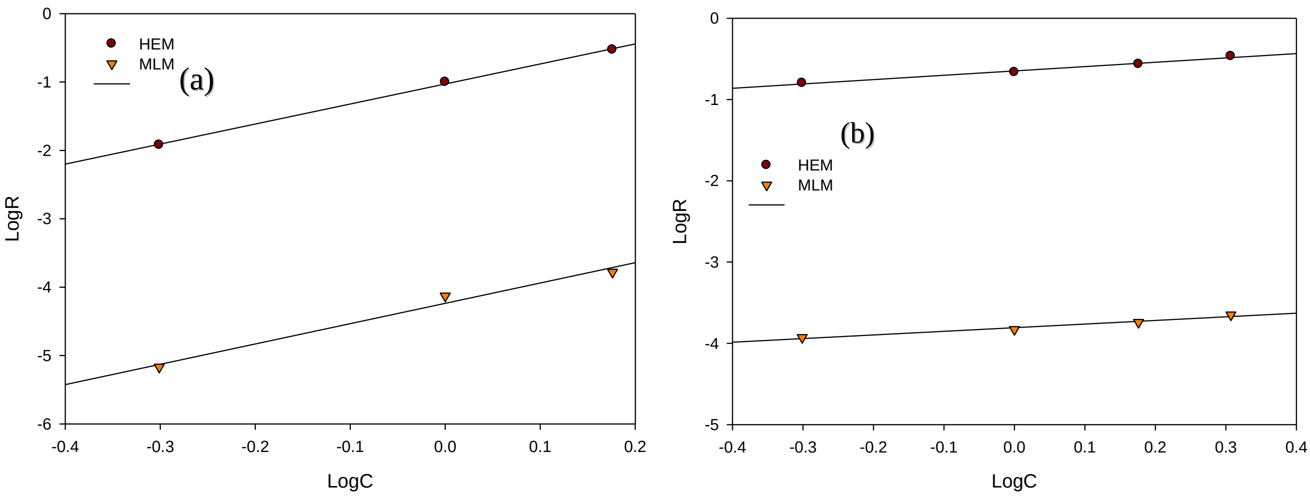

Figure 2: The relation between log R_{HEM} and/or log R_{MLM} for aluminum in (a)HCl and (b) NaOH solutions.

3.2 Inhibition characterization of Cantaloupe juice on corrosion rate

The inhibitory effect of the *cantaloupe juice* in absence and presence deferent concentration (%v/v) on the corrosion rate of aluminum in 1.0M HCl and 1.0M NaOH was measured by gasometry and mass loss methods at 27°C. As shown in **Table-3,** there is a good correlation between the *cantaloupe juice* concentrations and the hydrogen gas concentrations measured and effect of aluminum corrosion by gasometry in both media. This is illustrated in **Fig- 3,a,b.**

It is clear that as the concentration of *cantaloupe juice* increases, the rate of hydrogen evolution is decreased and surface coverage (θ) increase i.e., the rate of steel corrosion decreases, which indicates inhibition of the corrosion process. This implies that cantaloupe juice retard aluminum corrosion in the studied solutions. The corrosion rates (R_{MLM} and R_{HEM}) for aluminum sample in absence and presence of different concentrations of cantaloupe juice 27°C are given in **Table-3**.

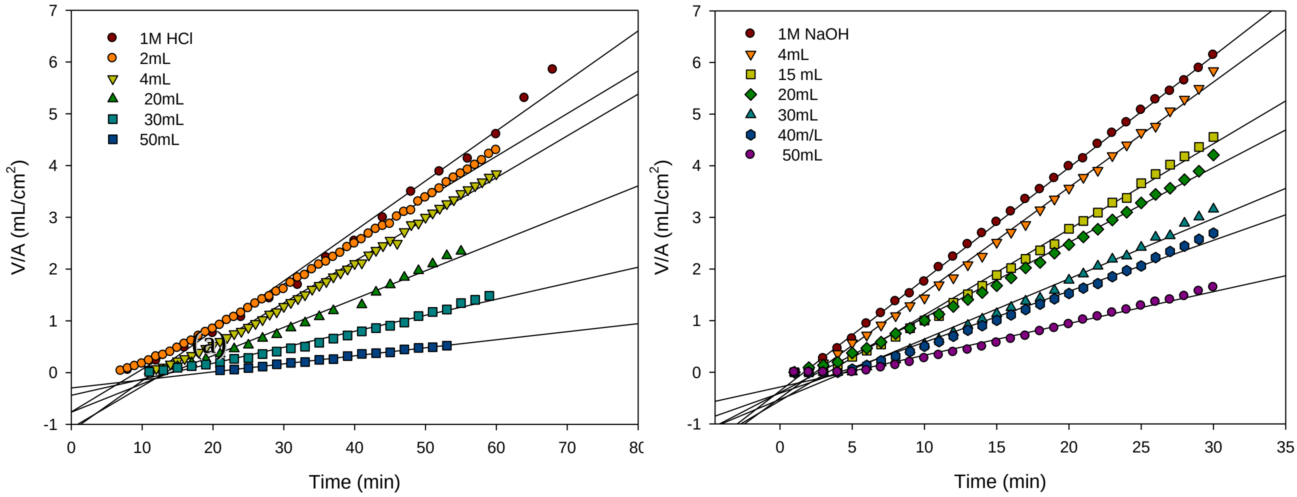

Figure 3: Relation between the volume of hydrogen evolved and exposure time in the absence and presence of different concentrations of cantaloupe juice (%v/v) in (a)HCl and (b) NaOH at 27°C.

Table 3: Corrosion rates , inhibition efficiencies and surface coverage in 1.0 M HCl and 1.0 M NaOH in absence and presence of different concentrations of cantaloupe juice at 27 °C.

Media		*Corrosion rate*		*Inhibition Efficiency*			
	$C_{inh.}$(%v/v)	R_{MLM} g.cm^{-2}.min	R_{HEM} mL.cm^{-2}.min	IE_{MLM} %	IE_{HEM} %	θ_{MLM}	θ_{HEM}
HCl	0	7.37×10^{-5}	0.099	-	-	-	-
	2.0	4.56×10^{-5}	0.082	38.11	15.01	0.38	0.15
	4.0	4.52×10^{-5}	0.081	38.62	16.15	0.38	0.16
	20.0	2.71×10^{-5}	0.055	63.13	43.48	0.63	0.43
	30.0	1.46×10^{-5}	0.031	80.08	67.91	0.80	0.68
	50.0	5.34×10^{-6}	0.015	92.75	84.16	0.93	0.84
NaOH	0	1.48×10^{-4}	0.216	-	-	-	-
	4.0	1.24×10^{-4}	0.204	15.61	5.690	0.15	0.056
	15.0	1.02×10^{-4}	0.166	30.68	23.31	0.31	0.233
	20.0	9.37×10^{-5}	0.146	36.64	32.81	0.37	0.328
	30.0	7.95×10^{-5}	0.117	46.28	45.99	0.46	0.459
	40.0	6.49×10^{-5}	0.106	56.11	51.14	0.56	0.511
	50.0	4.44×10^{-5}	0.067	69.95	68.61	0.69	0.686

As shown in **Table-3**, increase the concentrations of *cantaloupe juice* increase the inhibitor efficiency (EI%). The good efficiency of inhibition was found at concentration of 50 mL(%v/v) (84.16% & 92.75%) in HCl solution. This mean the *juice* can act as excellent eco-friendly corrosion inhibitor at high concentration up to 50:50 (inhibitor:acid) compared to it's effect in NaOH (68.61% & 69.95%). Also data in **Table-3** reveal good agreement between mass loss and hydrogen evaluation measurments.

Relation between the volume of hydrogen evolved and exposure time in the absence and presence of different concentrations of *cantaloupe seed* extract (%v/v) in 1.0 M HCl and 1.0 M NaOH at 27°C is represent in **Fig-4.** Data in **Table-4** indicate the inhibitory effect of the *seeds extract* for aluminum corrosion is a concentration-dependant manner.

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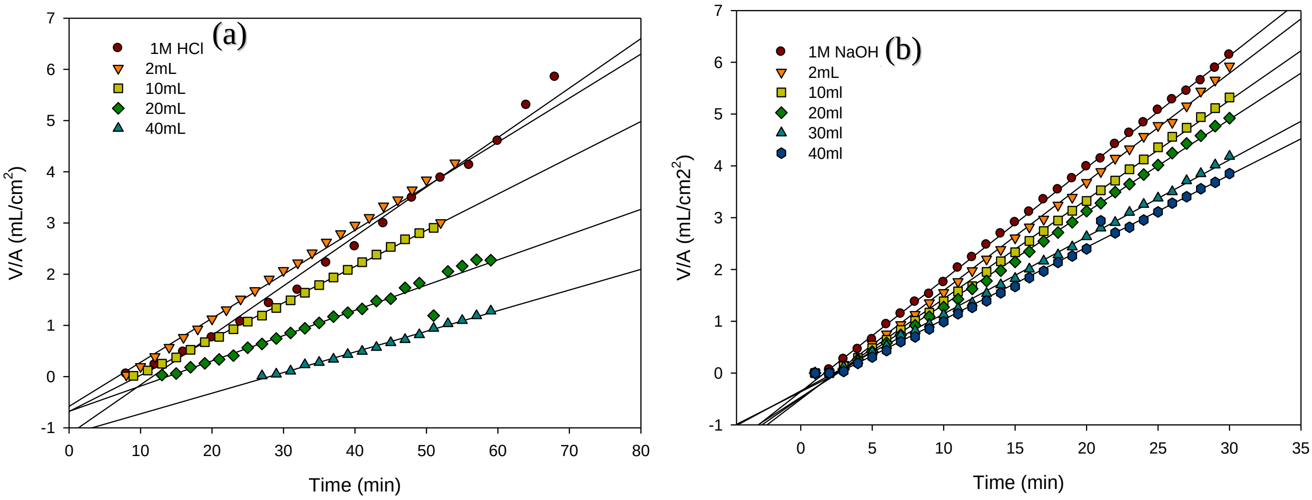

Figure 4: Relation between the volume of hydrogen evolved and exposure time in the absence and presence of different concentrations of cantaloupe seed extract (%v/v) in (a)HCl and (b) NaOH at 27°C.

Table 4: Corrosion rates, inhibition efficiencies and surface coverage in 1.0 M HCl and 1.0 M NaOH in absence and presence of different concentrations of cantaloupe seed at 27 °C.

Media		***Corrosion rate***		***Inhibition Efficiency***			
	$C_{inh.}$(%v/v)	R_{MLM} g.cm^{-2}.min	R_{HEM} mL.cm^{-2}.min	IE_{MLM} %	IE_{HEM}%	θ_{MLM}	θ_{HEM}
HCl	0	7.37×10^{-5}	0.099	-	-	-	-
	2.0	5.00×10^{-5}	0.086	32.14	10.97	0.32	0.11
	10.0	4.15×10^{-5}	0.071	43.70	26.50	0.44	0.27
	20.0	3.00×10^{-5}	0.049	59.24	48.69	0.59	0.49
	40.0	2.09×10^{-5}	0.040	71.60	58.28	0.72	0.58
NaOH	0	1.48×10^{-4}	0.216	-	-	-	-
	2.0	1.44×10^{-4}	0.209	2.818	2.98	0.03	0.03
	10.0	1.29×10^{-4}	0.191	12.54	11.45	0.13	0.11
	20.0	1.22×10^{-4}	0.178	17.66	17.33	0.17	0.17
	30.0	9.81×10^{-5}	0.148	33.68	31.25	0.34	0.31

Table-4 also shows that the corrosion rate according the weight loss values of aluminum in 1.0 M HCl and 1.0 M NaOH solutions containing *cantaloupe seed extract* decreased as the concentration of the inhibitor increased, i.e., mass and charge transfers rate decrease with increase of seed extract concentration at 40 mL. This trend, it may result from the fact that adsorption amount and the coverage of surfactants (θ) on the aluminum surface increase with the increase of the concentration, thus the aluminum surface is efficiently separated from the aggressive medium. The inhibitor efficiency of *cantaloupe juice and seed extract*s may due to presence of many organic substance that acts a good corrosion inhibitors and various vitamins [20]. These organic compounds usually contain polar functions with oxygen atoms and have triple or double bonds or aromatic rings. The adsorption of these organic compounds 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.

As conclusion, **Fig-5** represent the inhibition efficiency (IE%) using MLM and HEM respectively, for aluminum corrosion in 1.0 M HCl and 1.0 M NaOH by cantaloupe extracts. It is clear that *juice extract* is more protective for aluminum in all studded concentrations used in this research than *seeds extract* in both media:

juice extract > seed extract

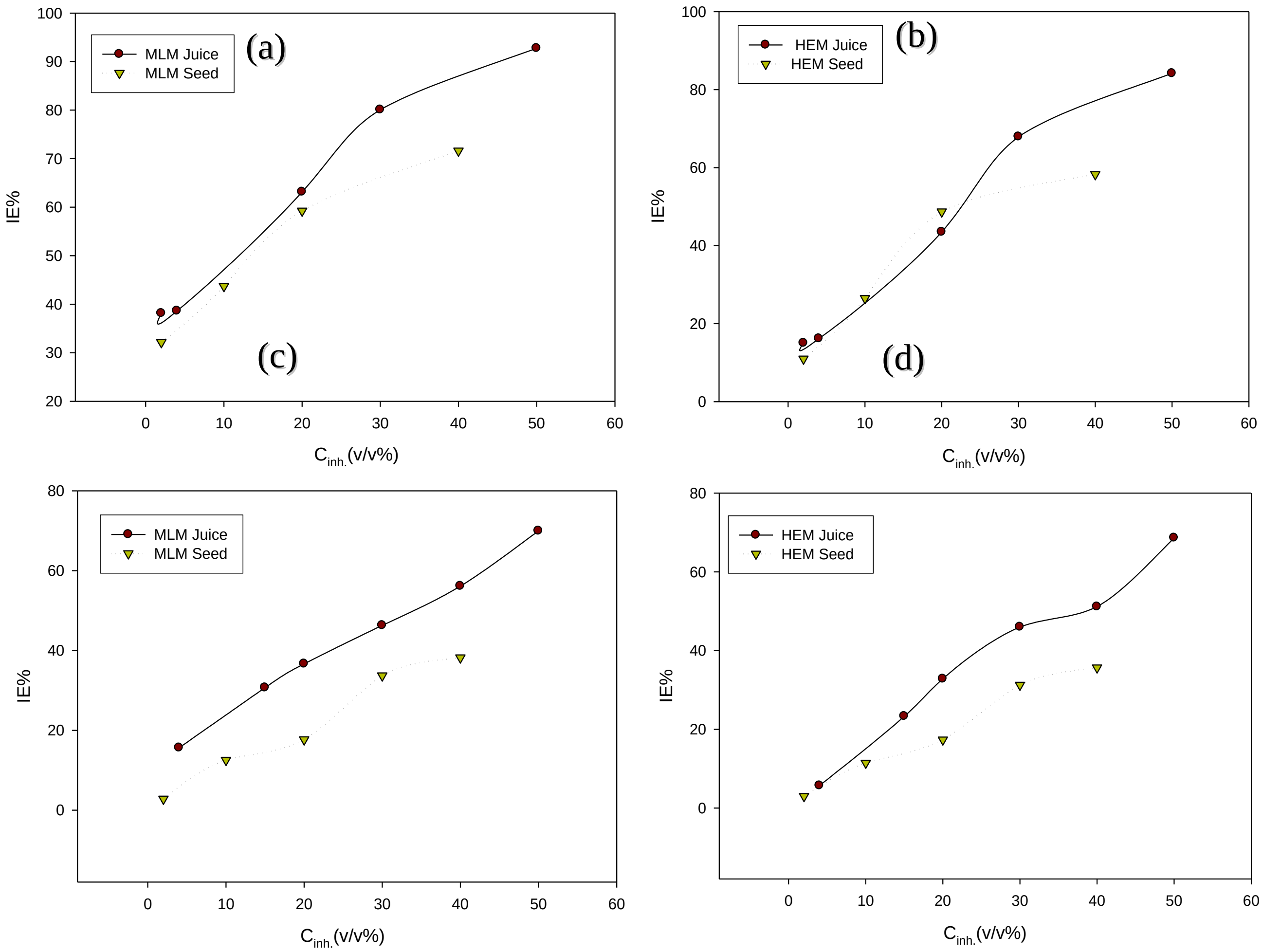

Figure 5: Relationship between inhibition corrosion inhibition efficiencies and the cantaloupe extracts (a,c) HCl and (b,d) NaOH.

3.4 Thermodynamic/Adsorption Parameters

Assuming the corrosion inhibition was caused by the adsorption of Cantaloupe juice and seeds extracts on the aluminum in 1.0M HCl, the degree of surface coverage, θ, **Table-4** of the metal surface was calculated using Eqs. (2 & 5). The adsorption of an organic adsorbate at a metal solution interface can be represented as a substitutional adsorption process between the inhibitor molecules, Eq. (14), in the aqueous solution $Inh._{(sol)}$ and the water molecules on the metallic ,M, surface $H_2O_{(ads)}$ [21].

$$M_{(H_2O_{ads})x} + Inh._{(sol)} \rightarrow M_{(Inh.)} + xH_2O \qquad (14)$$

From above equation the inhibition efficiency of *Cantaloupe juice and seed* extracts measured by determined how molecules from water, (x), was replacement by inhibitor case block for the active site in aluminum and decrease the corrosion rate. The inhibition mechanism of aluminum could be explained by the Al-$(inh)_{ads}$ reaction intermediates, Eq. (15).

$$Al + Inh. \rightleftarrows Al(Inh.)_{ads} \rightleftarrows Al^{n+} + n\,e + Inh. \qquad (15)$$

At first, where there is not enough $Al_{(Inh)ads}$ to cover the metal surface, because the inhibitor concentration is low or because the adsorption rate is slow, metal dissolution takes place on sites of the aluminum surface free of $Al_{(Inh)ads}$. With high inhibitor concentration, a compact and coherent inhibitor over layer forms on the aluminum surface, reducing chemical attack of the metal [22].

Basic information on the interaction between the inhibitor and the aluminum surface can be provided using the adsorption isotherm. Data obtained for the degree of surface coverage were used for the evaluation of different adsorption isotherms (Langmuir and Temkin).

3.4.1 Langmuir Isotherm

Langmuir adsorption isotherm is expressed according to Eq. (16)

$$\frac{C}{\theta} = \frac{1}{K} + C \qquad (16)$$

where C is the concentration of the inhibitor, K is the adsorption equilibrium constant and θ is degree of surface coverage of the inhibitor. Plotting $\frac{C}{\theta}$ against C (% v/v) of *juice* and *seed*, **Fig-6**, using MLM and HEM in 1.0 M HCl gave a linear relationship.

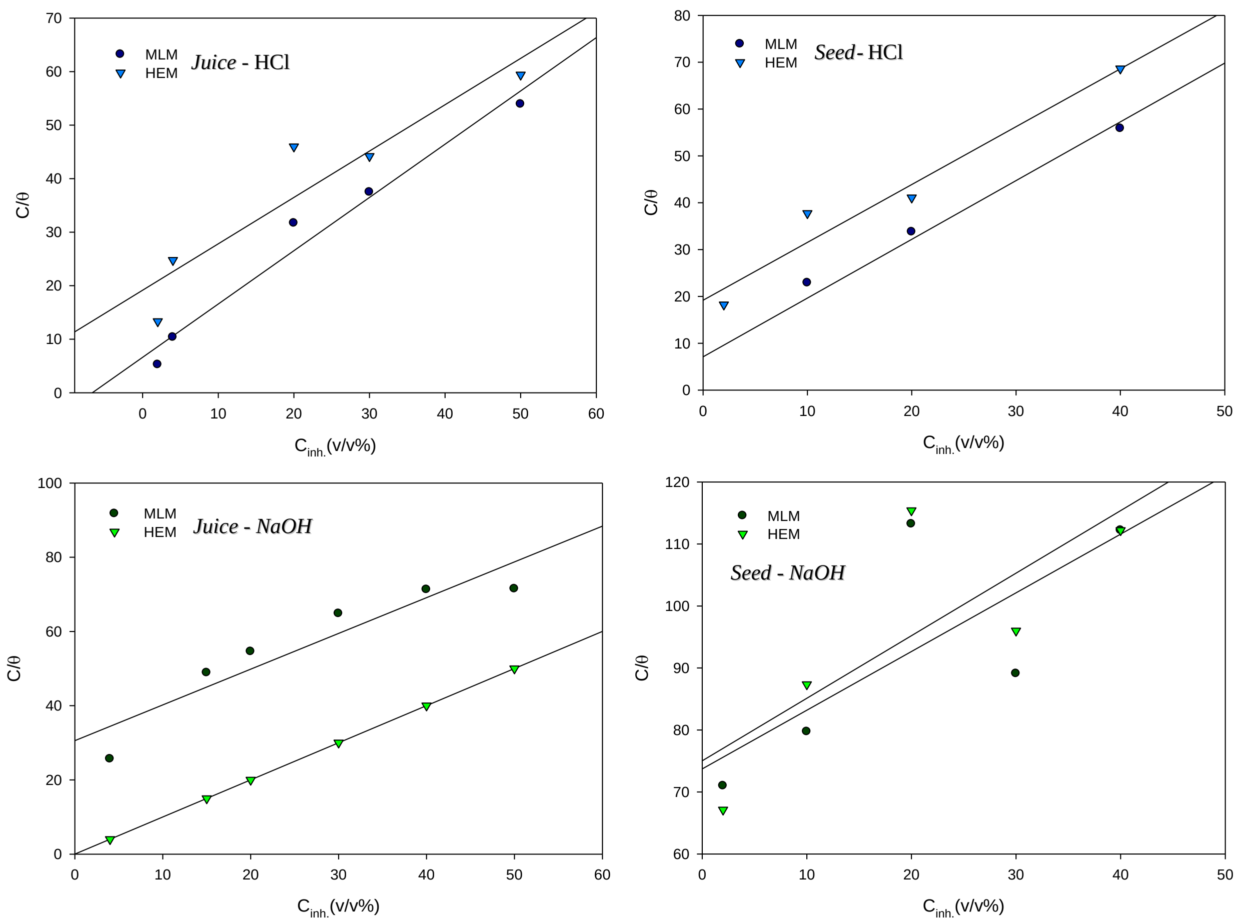

Figure 6: Langmuir isotherm for adsorption of Cantaloupe juice and seed extract on aluminum surface.

The linear correlation coefficient values, r^2, of fitted data is very close to 1 in 1.0 M HCl solution as shown in **Table-5** for MLM and HEM in presence of *juice and seed extract*s. This indicates strong adherence to Langmuir adsorption isotherm. Otherwise, the adsorption of only *cantaloupe juice* on aluminum surface in 1.0 M NaOH obeys the Langmuir adsorption isotherm. The application of Langmuir isotherm on surface of aluminum indicated that there is no interaction between the adsorbate molecules and they only occupy one site. The equilibrium constant K for the adsorption /desorption process of the inhibitor molecules on the metal surface calculated and listed in **Table-5**.

3.4.2. Temkin Isotherm

The degree of surface coverage (θ) is related to inhibitor concentrations (C) according to Eq.(17) :

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

where K is the adsorption equilibrium constant and a is the attractive parameter. Plots of θ against logC (%v/v) of *juice and seed extract*s as presented in **Fig-7** gave linear relationship with r^2>0.90 in HCl solution, which shows that adsorption data fitted Temkin adsorption isotherm. Adsorption parameters obtained from Temkin adsorption isotherms are recorded in **Table-5**. The values of attractive parameter (a) are negative in all cases, indicating that repulsion exists in the adsorption layer. The adsorption constant K calculated and also represents in **Table-5**.

As the natural extract contains infinite components at various contents, we may introduce that inhibitory effect is conducted by the intermolecular synergistic effect of several molecules [23-25].

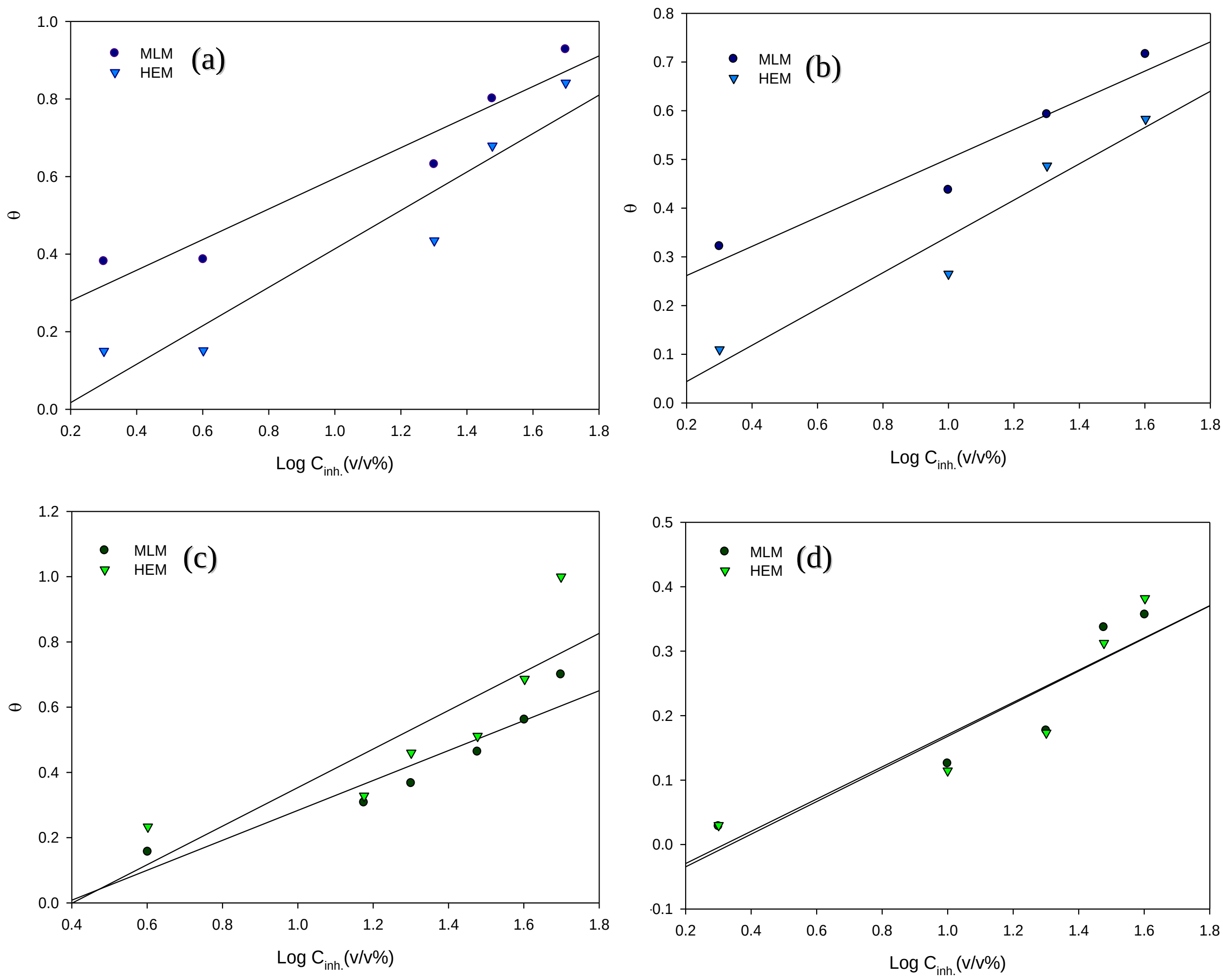

Figure 7: Temkin isotherm for adsorption of Cantaloupe (a,c) juice and (b,d) seed.

Table 5: Adsorption parameters for adsorption of aluminum surface in mass loss and hydrogen evaluation measurements.

			Isotherm	r^2	Log K	
HCl	MLM	juice	Langmuir	0.972	-8.21	Slope= 0.99
		seed		0.979	-0.851	Slope=1.25
		juice	Temkin	0.929	0.509	a= -2.92
		seed		0.931	0.672	a= -3.85
	HEM	juice	Langmuir	0.879	-1.284	Slope= 0.87
		seed		0.955	-1.284	Slope= 1.24
		juice	Temkin	0.911	-0.154	a= -2.34
		seed		0.941	-0.083	a= -3.09
NaOH	MLM	juice	Langmuir	0.868	-1.49	Slope=0.96
		seed		0.568	-1.87	Slope=0.95
		juice	Temkin	0.891	-0.08	a = -0.53
		seed		0.856	-0.02	a = -0.29
	HEM	juice	Langmuir	0.999	2.41	Slope=1.00
		seed		0.607	-1.88	Slope=1.00
		juice	Temkin	0.721	-0.14	a = -0.68
		seed		0.833	-0.02	a = -0.29

Conclusion

The results obtained from HEM and MLM of corrosion and corrosion inhibition of aluminum in various concentrations HCl **(0.5-1.5M)** and NaOH **(0.5-2M)** solutions, under 27°C; can be deduced:

1- Dissolution of aluminum in HCl and NaOH solutions is concentration depend.
2- Base media is more aggressive for aluminum surface than the acid media.
3- *Cantaloupe juice and seeds extracts* are good inhibitors for aluminum in both 1.0M HCl and 1.0M NaOH, and inhibition efficiency increases with the increase in the concentration of *extracts*.
4- *Cantaloupe juice* shows good inhibitive properties for aluminum in both 1.0M HCl and 1.0M NaOH than *seed extracts*.
5- The correlation coefficient (r^2) was used to choose the isotherm that best fits the experimental data. The adsorption of *Cantaloupe juice and seed* extract molecules on Aluminium metal surface in 1.0M HCl follows Langmuir adsorption isotherm.
6- The best fit to the experimental data was obtained when the Langmuir isotherm was used for *Cantaloupe juice* and Temkin isotherm for *Cantaloupe Seeds* in 1.0M NaOH.

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