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Comparative data on the protection performance of celery, pomegranate and green tea distillates on mild steel in weak H_2SO_4 solution

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ABSTRACT

The non-toxic properties of plant distillate for application as corrosion inhibitors is an effective alternative to the toxic conventional derivatives currently applied in industries. Data on the corrosion inhibition effect of celery, pomegranate and green tea distillates on mild steel corrosion in 0.5 M H_2SO_4 from coupon analysis is presented. Results showed the distillates performed poorly at low concentrations from 15% to 45% distillate concentration. At 60% distillate concentration, their inhibition performance was average, but below the values for effective inhibition performance with final inhibition efficiency values of 54.55%, 48.65% and 32.39% for celery, pomegranate and green tea distillates. However, at 75% concentration, the inhibition efficiency values significantly increased to 85.74%, 82.85% and 62.70%. At 90% distillate concentration, the inhibition efficiency outputs of the distillates increased further to 95.83%, 87.08% and 92.37% signifying effective inhibition performance of the distillates. Statistical analysis through ANOVA shows distillate concentration strongly influence the corrosion inhibition performance of the distillates with values above 90% compared to exposure time with values below 7%, which is relatively negligible but statistically relevant. Generally, the results show the distillates are effective at high concentration with respect to distillate concentration.

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1. Introduction

Corrosion is the gradual or accelerated depreciation of the mechanical, physical and aesthetic attributes of metallic alloys resulting from the molecular interaction of the alloy with their environment [1–4]. According to NACE, the total cost of corrosion damage worldwide amounts to $ 2.5 trillion [5]. Industrial operations such as petrochemical refining, chemical processing, fertilizer production, acid descaling, oil and gas pipelines, heat exchangers, automobile radiators, desalination and energy generation extensively use carbon steels for construction of structures, equipment and components due to low cost of production and retail value, reusability and ease of fabrication [6–8]. Carbon steel is the largest steel produced worldwide constituting about 80% of the total tonnage produced. The steel exhibits low resistance to corrosion by contrast to stainless steels because of the absence of inert protective oxide which forms on stainless steel surface. Corrosion of carbon steels is a prevailing problem resulting in extensive damage to metallic parts, plant shut downs, toxic leakages, and industrial accidents etc. These are responsible for the expensive maintenance and rectifications, and huge financial loss [9]. Sulphate anions exist in aqueous electrolyte in petrochemical distillation unit, oil well maintenance, rust removal, desalination and pickling processes, chemical production etc. where they cause significant damage to the steels in service [10]. Their effect can be mitigated through the utilization of fluid derivatives tagged as corrosion inhibitors. Corrosion inhibitors are one of the most economical techniques of stifling corrosion attack [11–15]. Research has identified corrosion inhibitors with heteroatoms such as O, N, and S to be highly effective and strongly adsorb unto the ferrous alloys. This phenomenon is also a product of the physical and chemical characteristics of the inhibitor, reactive groups, electron configuration of the contributor atom, p-orbital property, and the electronic configuration of inhibitor molecules [16–22]. However, conventional corrosion inhibiting compounds are toxic, carcinogenic and severely contaminate the environment. Application of sustainable and biodegradable compounds has been proven to be effective in suppressing the oxidation and reduction of half-cell reactions [23–31]. The data in this article compares the protection efficiency of celery,

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pomegranate and green tea distillates on mild steel in 0.5 M H_2SO_4 solution (See Fig. 1).

2. Experimental methods

Mild steel (MS) rod (6 mm thickness and 12 mm diameter) with ostensible weight constituent (wt. %) of 0.8% Mn, 0.04% P, 0.05% S, 0.16% C and 98.95% Fe was divided with hacksaw into 3 distinctive categories of 7 samples each. The circular surface end of the steel samples was grinded and leveled with emery papers of different grits, and brightened with 6 μm diamond fluid. The samples were subsequently washed with deionized H_2O and CH_3COCH_3. Celery (CL) and pomegranate (PO) were individually chopped to tiny dimensions and grinded. The grinded marsh was squeezed to release the concentrate solution. Green tea (GT) was extricated with a rotary evaporator in 3 L of deionized H_2O. CL, PO and GT were separately formulated in volumetric concentrations of 10%, 15%, 30%, 45%, 60%, 75%, and 90% in 200 mL of 0.5 M H_2SO_4 solution. MS specimens were independently submerged in 200 mL of 0.5 M H_2SO_4 solution with the plant distillates at specific concentration and weighed every 24 h for a total of 240 h with Oarhus weighing balance in the physical metallurgy laboratory. Corrosion rate, C_R (mm/y) was enumerated from Eq. (1) [32];

$$C_R = \left[\frac{87.6\omega}{DAt}\right] \tag{1}$$

ω illustrates weight loss (mg), D illustrates density (g/cm^3), A illustrates total exposed surface area of MS specimen (cm^2), 87.6 is a corrosion rate constant and t illustrates is the time (h). Inhibition efficiency (η) was determined from Eq. (2) [33];

$$\eta = \left[\frac{\omega_1 - \omega_2}{\omega_1}\right] \tag{2}$$

ω_1 illustrates weight loss of MS from the acid solution without plant distillates while ω_2 illustrates weight loss of MS at determined CL, PO and GT concentrations. Dual-factor mono level experimental ANOVA test (F – test) was employed to estimate the statistical influence of the plant distillate concentrations and exposure time on their inhibition performance. The evaluation was done at confidence level of 95% i.e. a significance level of α = 0.05 with respect to the following equations. The addition of squares along the columns (exposure time) was enumerated from Eq. (3) while the addition of squares along rows (distillate concentration) was enumerated from Eq. (4). The total addition of squares was enumerated from Eq. (5).

$$SS_c = \frac{\sum T_c{}^2}{nr} - \frac{T^2}{N} \tag{3}$$

Addition of squares along the rows (plant distillate concentration)

$$SS_r = \frac{\sum T_r{}^2}{nc} - \frac{T^2}{N} \tag{4}$$

Total addition of squares

$$SS_{Total} = \sum x^2 - \frac{T^2}{N} \tag{5}$$

3. Results and discussion

3.1. Coupon measurement

Tables 1–3 presents the corrosion rate outputs of MS at 0%-90% concentration of CL, PO and GT distillates for 216 h. Tables 4–6 shows the corresponding inhibition efficiency values obtained for CL, PO and GT distillates. Observation of Table 1 depict the significant variation between the corrosion rate of MS without CL distillates and the values obtained in the presence of specific CL distillate concentration. The corrosion rate of MS without CL distillate varied from 137.92 mm/y at 24 h to 50.54 mm/y at 216 h. This figure is significantly high and signifies accelerated degradation of MS in the acid solution due to oxidation of MS by SO_4^{2-} anions. The redox reaction mechanism causes Fe^{2+} ions to pass into the acid solution causing degradation of the steel [34]. The visible reduction in corrosion rate with time is due to the gradual weakening of the electrolyte [35]. At 15% Cl distillate concentration the corrosion rate has decreased to values between 53.09 mm/y at 24 h to 52.51 mm/y at 216 h. The value is relatively high and shows lack of effective inhibitor protection of MS. The trend continued but no effective inhibition protection was observed till 60% CL distillate concentration where the corrosion rate at 216 h has reduced to 21.35 mm/y. Beyond 60% CL distillate concentration, the corrosion rate of MS decreased significantly with final values of 7.11 mm/y and 2.14 mm/y at 75% and 90% CL distillate concentration signifying effective inhibition performance. The same observation occurred for MS in the presence of PO and GT distillates. MS corrosion rate at 15% PO and GT distillate concentration at 24 h of exposure time are 39.29 mm/y and 71.66 mm/y whereas at 216 h of exposure time, corrosion rate values of 47.49 mm/y and 51.77 mm/y were observed. At 75% PO and GT distillate concentration, MS corrosion rate values are 8.27 mm/y and 17.08 mm/y at

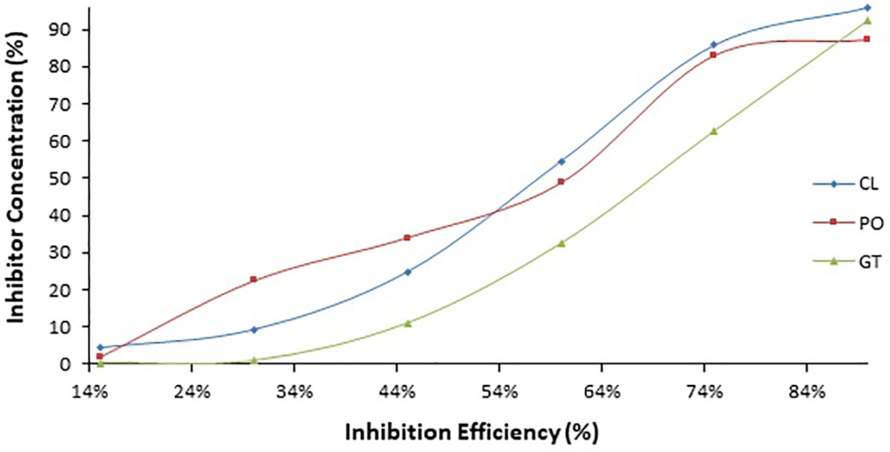

Fig 1. Comparative plot of plant distillate inhibition efficiency versus distillate concentration at 216 h.

Table 1
Corrosion rate result (mm/y) of MS at specific concentrations of CL (n = 1).

	CL Conc. (%)						
Exp. Time (h)	0% CL	15% CL	30% CL	45% CL	60% CL	75% CL	90% CL
24	137.92	53.09	50.19	15.47	5.22	1.24	1.49
48	137.92	53.09	50.19	15.47	5.22	1.24	1.49
72	108.56	59.84	48.82	16.22	6.63	1.65	0.90
96	84.46	65.81	57.41	26.58	11.93	3.38	1.24
120	69.93	63.00	54.38	27.47	12.08	3.59	1.54
144	75.82	64.99	59.19	33.46	17.02	5.40	1.91
168	64.98	63.14	57.86	35.04	18.15	5.66	1.88
192	56.86	57.16	53.35	36.59	19.95	6.47	2.03
216	50.54	52.51	49.84	37.80	21.35	7.11	2.14

Table 2
Corrosion rate result (mm/y) of MS at specific concentrations of PO (n = 1).

	PO Conc. (%)						
Exp. Time (h)	0% PO	15% PO	30% PO	45% PO	60% PO	75% PO	90% PO
24	113.22	39.29	27.44	20.35	11.5	4.94	1.02
48	113.22	39.29	27.44	21.8	11.5	4.94	2.51
72	82.9	44.63	31.80	24.7	12.87	4.76	2.30
96	68.97	50.54	39.85	31.1	18.11	7.54	2.56
120	65.05	48.33	37.96	30.77	18.93	7.43	2.73
144	63.46	50.69	40.28	32.64	21.01	8.04	5.53
168	65.02	50.70	39.39	31.99	22.04	7.95	5.78
192	56.89	48.90	38.29	28.58	23.05	8.13	6.59
216	50.57	47.49	37.44	31.84	23.84	8.27	6.2

Table 3
Corrosion rate result (mm/y) of MS at specific concentrations of GT (n = 1).

	GT Conc. (%)						
Exp. Time (h)	0% GT	15% GT	30% GT	45% GT	60% GT	75% GT	90% GT
24	138.47	71.66	57.42	24.54	3.18	8.58	1.3
48	138.47	71.66	47.28	24.54	16.64	8.58	1.3
72	116.08	75.52	51.96	31.21	21.15	10.11	2.11
96	116.79	81.36	58.93	38.34	26.08	13.36	3.07
120	93.43	76.59	55.94	38.26	25.9	12.82	2.97
144	77.86	75.44	56.58	40.77	28.33	14.09	3.36
168	66.74	66.56	54.61	40.61	28.13	14.51	3.58
192	58.40	58.24	52.83	42.33	30.54	15.96	3.69
216	51.91	51.77	51.45	43.68	32.42	17.08	3.78

Table 4
Inhibition efficiency result (%) of CL at specific concentrations (n = 1).

	CL Conc. (%)					
Exp. Time (h)	15% CL	30% CL	45% CL	60% CL	75% CL	90% CL
24	70.15	71.79	91.3	97.07	99.3	99.16
48	66.38	68.22	90.2	96.7	99.21	99.06
72	50.9	59.94	86.69	94.56	98.65	99.26
96	30.32	39.21	71.85	87.37	96.42	98.69
120	19.14	30.21	64.75	84.5	95.39	98.02
144	21.2	28.23	59.43	79.36	93.45	97.69
168	10.68	18.15	50.43	74.33	92	97.35
192	7.59	13.75	40.84	67.75	89.54	96.73
216	4.49	9.35	31.25	61.17	87.08	96.1
240	4.49	9.35	24.87	54.55	85.74	95.83

216 h while at 90% concentration the corrosion rate values are 6.2 mm/y and 3.78 mm/y at 216 h. Observation of the inhibition efficiency values indicated in Tables 4–6 gives more detail to the performance of the plant distillates. CL, PO and GT distillates performed poorly from 15% to 60% distillate concentration. At 60% distillate concentration, inhibition efficiency values of 54.55%, 48.65% and 32.39% were obtained for CL, PO and GT distillates at 216 h which shows poor inhibition performance. However, at 75% and 90% distillate concentration, the inhibition performance of the distillates significantly improved as shown in the Tables. Final values of 85.74%, 82.85% and 62.70% were obtained at 75% distillate concentration at 216 h. At 90% distillate concentration and 216 h of exposure time, final values of 95.83%, 87.08% and 92.37% were obtained for CL, PO and GT distillates. Distillates of plant materials contain many active components. The distillates consist of phytochemical compounds and heteroatoms atoms such as S, N, O, P

Table 5
Inhibition efficiency result (%) of PO at specific concentrations (n = 1).

	PO Conc. (%)					
Exp. Time (h)	15% PO	30% PO	45% PO	60% PO	75% PO	90% PO
24	65.29	75.76	82.03	89.84	95.64	99.10
48	65.29	75.76	80.74	89.84	95.64	97.78
72	46.16	61.64	70.20	84.47	94.26	97.22
96	26.72	42.22	54.90	73.74	89.06	96.29
120	25.69	41.64	52.69	70.90	88.58	95.81
144	20.13	36.53	48.58	66.90	87.34	91.28
168	22.02	39.42	50.80	66.10	87.78	91.12
192	14.05	32.69	49.77	59.48	85.71	88.42
216	6.09	25.96	37.04	52.85	83.64	87.75
240	1.89	22.37	33.96	48.65	82.85	87.08

Table 6
Inhibition efficiency result (%) of GT at specific concentrations (n = 1).

	GT Conc. (%)					
Exp. Time (h)	15% GT	30% GT	45% GT	60% GT	75% GT	90% GT
24	48.25	58.53	82.28	97.70	93.81	99.06
48	48.25	65.86	82.28	87.98	93.81	99.06
72	34.95	55.24	73.11	81.78	91.29	98.19
96	30.34	49.54	67.17	77.67	88.56	97.37
120	18.03	40.13	59.05	72.27	86.28	96.82
144	3.11	27.33	47.63	63.62	81.90	95.68
168	0.26	18.17	39.16	57.85	78.26	94.64
192	0.26	9.53	27.50	47.69	72.67	93.68
216	0.26	0.89	15.85	37.54	67.09	92.72
240	0.26	0.89	10.97	32.39	62.70	92.37

etc. which are capable of reacting with the steel surface through physiochemical mechanism and stifling the reduction–oxidation reactions causing corrosion. This reaction processes are strongly dependent on the distillates concentration; hence the increase in inhibition efficiency analogous to distillate concentration [4,36]. Comparative plots of the inhibition efficiency CL, PO and GT plant distillates with respect to concentration at 216 h show a progressive appreciation in inhibition efficiency values. The plot shows the performance of the plant distillates is strongly concentration dependent i.e. accumulation of inhibitor species on the steel exterior to counteract the corrosive effect of the acid species dominates the corrosion inhibition process of the distillates [37].

3.2. Statistical evaluation

Results from statistical analysis depicting the mean, standard deviation and margin of error for the inhibition efficiency of CL, PO and GT distillates are shown in Table 7. The standard deviation values show the extent of disparity between the inhibition efficiency results for the distillates. Observation of Table 7 shows the standard deviation values decreases with increase in inhibitor concentration. At 90% inhibitor concentration, the lowest standard deviation values for CL, PO and GT distillates (i.e. 1.27, 4.55 and 2.52) were obtained. This indicates that the obtained values at this concentration are closer to the mean value for the results due to the stability of the protonated inhibitor species on the steel exterior. The high standard deviation for the data at lower concentration of the distillates shows that the values vary over a wider range as a result of the instability of the protective covering by the distillates over the steel surface. CL distillate exhibited the lowest standard deviation value at 90% CL concentration, and the highest standard deviation value at 15% CL concentration. The value obtained for margin of error at 95% confidence level shows that 55%, 50% and 43% of the inhibition efficiency data of CL, PO and GT plant distillates are above 70% +6.4%.

Statistical analysis (ANOVA) was done to evaluate the influence of exposure time and distillate concentration (independent variables) on the inhibition performance of the distillates [38]. The

Table 7
Statistical data for standard deviation and margin of error for the distillate inhibition performance.

CL Distillates						
Distillate Conc.	15%	30%	45%	60%	75%	90%
SD	25.22	24.08	24.21	15.06	4.99	1.27
Mean	28.53	34.82	61.16	79.74	93.68	97.79
Margin of Error	6.4%					
PO Distillates						
Distillate Conc.	15%	30%	45%	60%	75%	90%
SD	22.49	19.19	16.56	14.51	4.69	4.55
Mean	29.33	45.40	56.07	70.28	89.05	93.19
Margin of Error	6.5%					
GT Distillates						
Distillate Conc.	15%	30%	45%	60%	75%	90%
SD	20.41	24.53	26.48	21.75	11.16	2.52
Mean	18.40	32.61	50.50	65.65	81.64	95.96
Margin of Error	6.4%					

Table 8
Statistical analysis (ANOVA) for CL, PO and GT plant distillates inhibition performance on MS at 95% confidence level.

CL						
Source of Variation	Addition of Squares	Degree of Freedom	Mean Square	Mean Square Ratio (F)	Theoretical Significance Factor	Statistical Relevance (%)
CL Conc.	278534.96	5	55706.99	457.39	2.44	93.77
Exp. Time	13011.95	9	1445.77	11.87	2.10	4.38
Residual	5480.73	45	121.79			
Total	297027.64	59				
PO						
Source of Variation	Addition of Squares	Degree of Freedom	Mean Square	Mean Square Ratio (F)	Theoretical Significance Factor	Statistical Relevance (%)
PO Conc.	251688.15	5	50337.63	830.25	2.44	95.23
Exp. Time	9891.13	9	1099.01	18.13	2.10	3.74
Residual	2728.33	45	60.63			
Total	264307.61	59				
GT						
Source of Variation	Sum of Squares	Degree of Freedom	Mean Square	Mean Square Ratio (F)	Theoretical Significance Factor	Statistical Relevance (%)
GT Conc.	221537.87	5	44307.57	461.94	2.44	91.38
Exp. Time	16594.41	9	1843.82	19.22	2.10	6.84
Residual	4316.27	45	95.92			
Total	242448.55	59				

influence is given in the form of statistical relevance shown in Table 8. It was done to assess the statistical relevance of exposure time and CGT concentration (sources of variation) on the inhibition efficiency values of CGT compound. The results in Table 8 shows that distillate concentration overwhelmingly influence the resulting inhibition efficiency values at statistical relevance factor of 93.77%, 95.23% and 91.38%. The corresponding values for exposure time were relatively and significantly low. However, they are still statistically relevant because the corresponding value for mean square ration is higher than the theoretical significance factor. The theoretical significance factor is the threshold minimum for which a value is statistically relevant. The statistical relevance values obtained showed that the performance of the plant distillates is strongly dependent on its concentration and to a lesser degree acts with respect to variation.

4. Conclusion

Gravimetric analysis of the corrosion protection efficiency of celery, pomegranate and green tea distillates on mild steel corrosion in 0.5 M H_2SO_4 solution showed the distillates underperformed at low concentrations. Effective inhibition performance was observed at relatively higher distillate concentration with values generally above 90% at optimal distillate concentration. Statistical analysis through ANOVA shows distillate concentration strongly influence the corrosion inhibition performance of the distillates with values above 90% compared to exposure time with values below 7%, which is relatively negligible but statistically relevant. Statistical data of the mean, standard deviation and margin of error for the inhibition efficiency of the distillates show the extent of disparity between the values due to the stability of the protonated inhibitor molecules on the steel surface. Analysis of variance evaluated showed distillate concentration strongly influenced the inhibition performance of the extracts.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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