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Research Article

SOME NATURAL PRODUCT EXTRACTS AS ECO-FRIENDLY INHIBITORS ON THE CORROSION BEHAVIOR OF MILD STEEL EXPOSED TO 1.0M SULPHURIC ACID

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ABSTRACT

The effect of some natural products (Punica Granatum Peel PG, Solenostemma Argel SA, Olibanum O, Datura Seeds DS and Datura Leaves DL)on the corrosion behavior of mild steel exposed to 1.0M sulfuric acid was studied by electrochemical techniques (open circuit potential, linear and Tafel polarizations).The present study revealed that as the concentration of inhibitor increased the inhibition efficiency increased. The polarization studies showed that all used inhibitors act as mixed type inhibitors. The adsorption of the inhibitor molecules on mild steel surface obeys Langmuir adsorption isotherm. The surface analysis via SEM techniques indicates that the active molecules from all inhibitors absolute retard the corrosion on the specimen surfaces.

Keywords: Corrosion inhibitor; Natural products; Mild steel; Acidic media.

1. INTRODUCTION

Mild steel (MS) is important metal which used in different human activities and it is susceptible to different mechanisms of corrosion due to its exposure to different corrosive acidic media.H_2SO_4 is usually used as industrial acid cleaning and pickling acids, but it affects negatively on the metal surface due to containing aggressive ion (SO_4^{--}).Addition of inhibitors one of the methods which reduce or retard the corrosion process for mild steel. There are many researches which study the suitable compounds at exposed to acidic media used as inhibitor corrosion, compounds have heterocyclic atoms act as inhibitors, but it has extensive, hazard for human, it may be difficult in synthesized[1]. To avoid obvious reasons in inhibitor uses almost researchers directed their studies for using natural products as inhibitors[2-6].From these studies different years[7]such as fenugreek, henna, olive, jojoba, black pepper, Occimum viridis, Andrographis paniculata, Phyllanthus amarus, onion, garlic, Eugenia jambolans, Pongamia glabra, opuntia and eugenol, Zenthoxylum alatum, Nypa fruticans Wurmb, Oxandra asbeckii, Ferula assa-foetida and Dorema ammoniacum, Lavandula angustifolia, Justicia gendarussa, Gissipium hirsutum, Lupinous albus, Aloe vera, etc[8-31].The constituent and characterization of the natural products used in this study were presented in prior research[32]

The aim of this study was to investigate the inhibitory effect of some natural product extracts as a cheap, easily preparation and non-toxic corrosion inhibitor on MS corrosion in 1.0 M H_2SO_4 acid. The electrochemical measurements were used to evaluate the inhibition efficiencies.

EXPERIMENTAL APPROACH

2.1. Preparation of Natural Products Extracts and Corrosives Media:

An aqueous extract solution was prepared by reflux 10 g of (Punica Granatum PG, Solenostemma Argel SA, Olibanum O, Datura Seeds DS and Datura Leaves DL) in 100 ml of double distilled water for about 1 hr, then, filtering the refluxed solution to remove any contaminations, and evaporate 1 ml of filtered extract solution and weighted the residue[33] and preparation of 1.0M HCl solution as corrosive media.

2.1.1 Characterization of Natural Products (PG, SA, O, DS and DL)

The solid plant extracts of PG, SA, O, DS and DL characterized by Fourier transform infrared (FTIR) spectroscopy. FTIR spectrum recorded in Jasco-FTIR-480 plus (Japan) which extended from 4000 to 400 cm^{-1}, using the KBr disk technique. The active substances and medical uses of the natural products, which used in this work represented by Mahross et al [32].

2.2. Preparation of test specimens:

The MS specimens tested in this present paper, are in the sheet form, supplied by the AL - EZZ Company in Alexandria. The designation and analysis of the material by weight % is (C 0.175, Si 0.130, Mn 0.534, P 0.008, S 0.0094, Cr 0.0117, Mo 0.002, Ni 0.0188, Al 0.005, Co 0.0075, Cu 0.0047, Nb 0.003, Ti 0.002, V 0.005, W 0.172 and Fe 98.9). Prior immersion the MS electrodes in the tested solutions, it was prepared by polishing with SiC paper up to 1200 grade, washing with distilled water and then degreased with alcohol about 5 minutes, washed again with distilled water then dried it by used filter papers. H_2SO_4 (1.0M) prepared by analytical method (from Aldrich, its purity of acid 98%)

2.3. Electrochemical Cell (Corrosion Cell):

The corrosion cell involving three electrodes, MS as working electrode, Saturated Calomel Electrode (SCE) as reference electrode and a platinum wire used as counter electrode. The exposed area of working electrode to solutions was $1Cm^2$.

2.4. Adopted Techniques:

The major electrochemical techniques used in this study are open circuit potential (OCP) and Potentiodynamic polarizations (linear polarization resistance (LPR) and Tafel plots polarizations (TP)) which carried out with model 352/252 corrosion measurement system, which consists of EG & G potentiostat/galvanostat model 273A driven by software from IBM computer. Measurements of polarization curves were started after immersing the working electrode in the used solutions for close to 30 minutes, where the values of E_{corr} became stable. The LPR was measured from +20 mV up to -20 mV vs E_{corr} and the TP was measured from -250 mV up to +250 mV vs E_{corr}. The scan rate for LPR and TP measurements were 0.166 mV/S and 0.3mV/S, respectively.

3. RESULTS AND DISCUSSION

3.1. Open Circuit Potential Measurements (OCP):

OCP describes the relation between potential (mV) versus time (sec) at zero current. OCP represents the behavior of anodic and cathodic reactions takes place on the surface of metal electrode. As the potential electrode shift into a positive direction (noble direction) that indicating to anodic inhibitor, that means blocking of inhibitor molecules of the active anodic sites by its adsorption on the metal surface electrode, and vice versa for the potential electrode that the shift to more negative potentials (increasing in negatively charge) that indicating to cathodic inhibitor, that means retarding or decreasing the cathodic reaction. The potential-time curves for (PG, SA, O, DS and DL) at 1.0M H_2SO_4 for MS electrode at exposure time 4 days(where its best values) were presented in Fig. (1).It is clear from curves in all concentrations of each inhibitors used in this study, the steady state potential ($E_{s.s}$) of electrode shift into significant positive potential than blank solution, and the values of these $E_{s.s}$ listed in Table 1, according the values there is formation of layer of particles of inhibitor which adsorbed on the active anodic sites on the surface of MS electrode.

3.2. Potentiodynamic Polarization Measurements (linear and Tafel polarizations):

These techniques can provide significant useful information regarding the corrosion rate. Polarization methods involve changing the potential of the working electrode and measuring the current which is produced as a function of potential. Potentiodynamic polarization is a technique which describes the relation between applied potential at MS electrodes at 0.4mV/Sec and current as a result from this applied potential, and the rate of this potential is long reach to 1000 mV. The values of current give us an indication about corrosion rate, we can calculate the inhibition efficiencies of testing media from corrosion rate data, this relation given by namely Tafel plot polarization. On the other hand, as a portion of potentiodynamic polarization (linear polarization resistance LPR) which describe the linear relation between applied potential and current results from this potential and the rate of potential is short nearly 50mV, we can calculate resistance polarization from LPR which means the ability of a solution to retard or decrease the corrosion of metal in corrosive media by using inhibitors. Fig.(2) Showed Tafel polarization curves for MS in 1 M H_2SO_4 with the addition of various concentrations of testing inhibitors at exposure time 4 days. The different corrosion parameters derived from these curves are presented in Tables (2 and 3). From the

Table 3, it is clear that the corrosion current density (I_{corr}) value decreased from 12078µA/cm^{2}at a blank solution to 90.59µA/cm^{2}to give to give IE%, nearly to 99% at the presence of 5%(v/v)% concentration of SAE in 1 M H_2SO_4. In blank solution, both βa and βc values changed significantly and E_{corr} shift to anodic region, hence, SAE acted as a mixed type inhibitor.

IE% values of MS in 1.0 M H_2SO_4solution containing different concentrations of inhibitor extracts were calculated from the Tafel plots polarization by I_{corr} data as using equation 1:

$$IE\% = [\frac{(I_{corr(uninhib)}) - I_{corr(inhib)}}{I_{corr(uninhib)}}] \times 100............ (1)$$

Where $I_{corr\ (uninh.)}$ and $I_{corr\ (inh.)}$ are the corrosion current density without and with inhibitor presence, respectively.

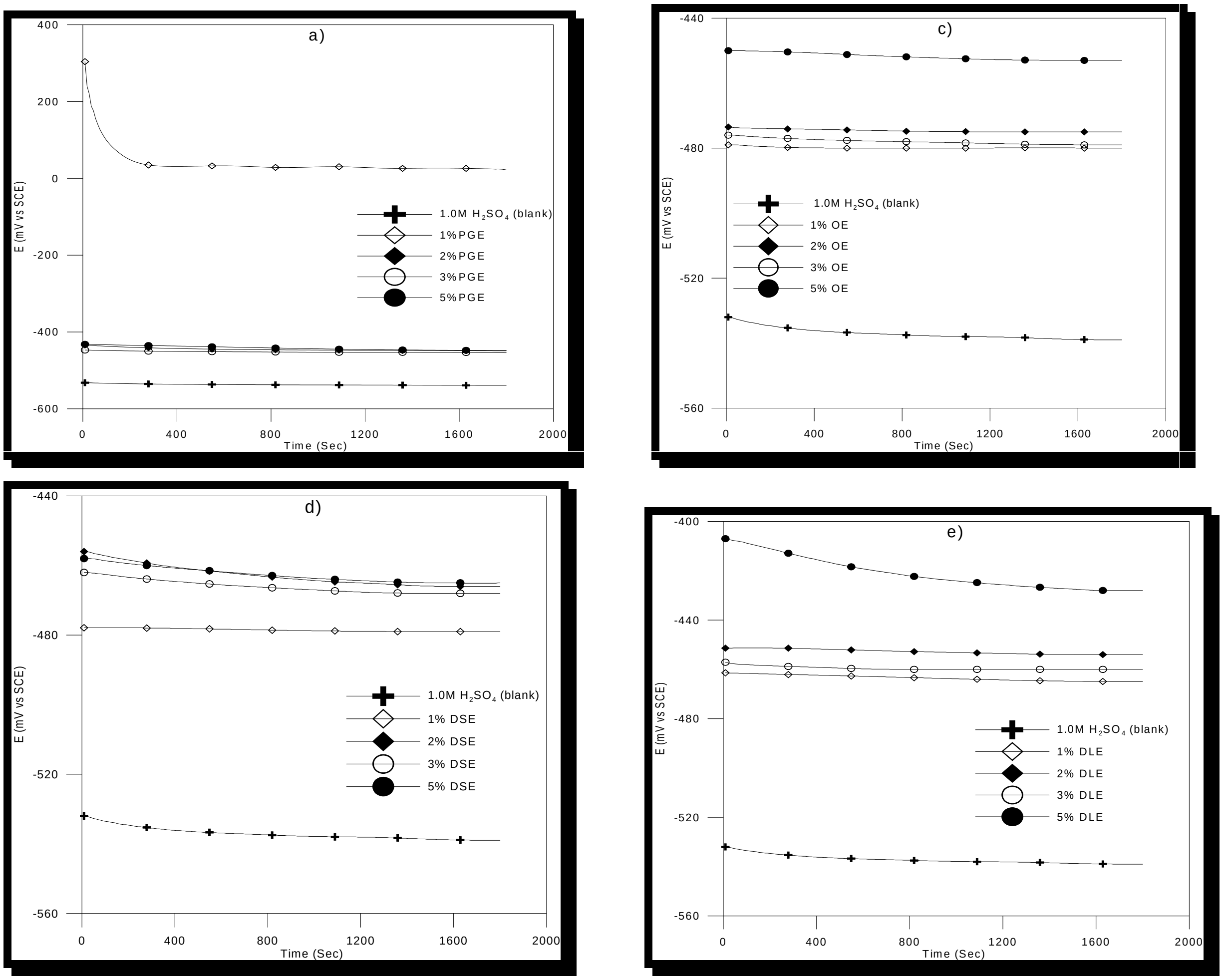

Figure 1

Potential-time curves for mild steel in 1.0M H_2SO_4 at different concentrations of natural products after 4days.

We can concluded that the sequence of inhibitor extracts in the presence of 1.0M H_2SO_4 at 2 hrs, 1 day, 2 days and 4 days was arranged as O (48.27%–88.61%) > SA (43.64%–87.26%) > PG (28.45%–79.99%) > DS (25.13%–57.82%) > DL (29.75%–57.31%), SA (89.44%–98.44%) > O (90.70%–98.20%)> PG (93.06%–96.88%) > DL (87.45%–92.99%) > DS(86.56%–92.31%), SA (91.65%–98.85%) > O (92.77 %–98.68%) > PG (94.47% – 98.30%) > DL (90.40%–94.77%) > DS (90.16% – 94.14%) and SA (92.26%–99.03%) > O (93.85%–98.99%)> DS (92.01%–96.89%) > PG (94.89%–98.71%)> DL (91.91%–96.68%), respectively.

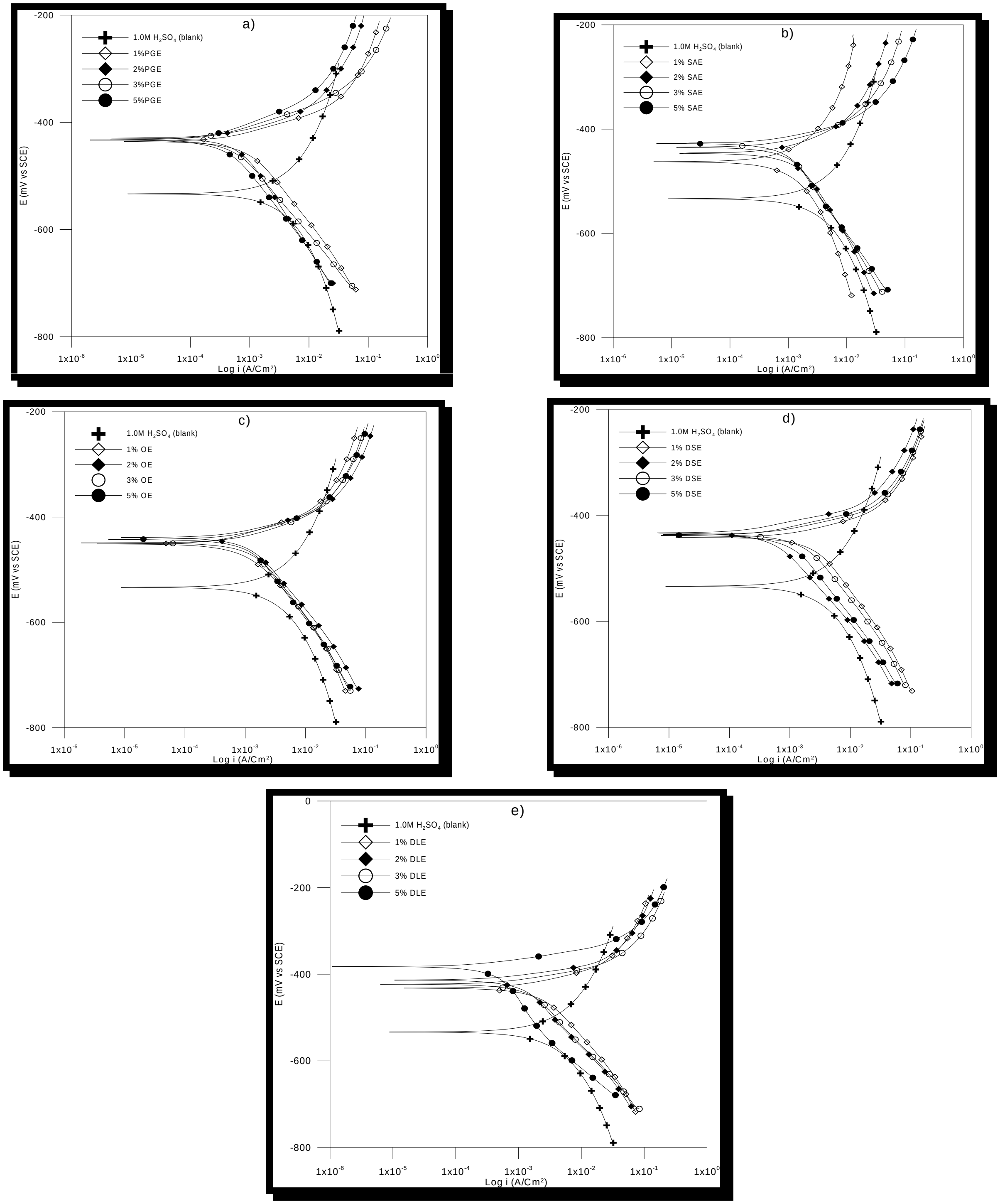

Figure 2

Tafel plots polarization curves of mild steel in 1.0M H_2SO_4at different concentrations of natural products after 4days.

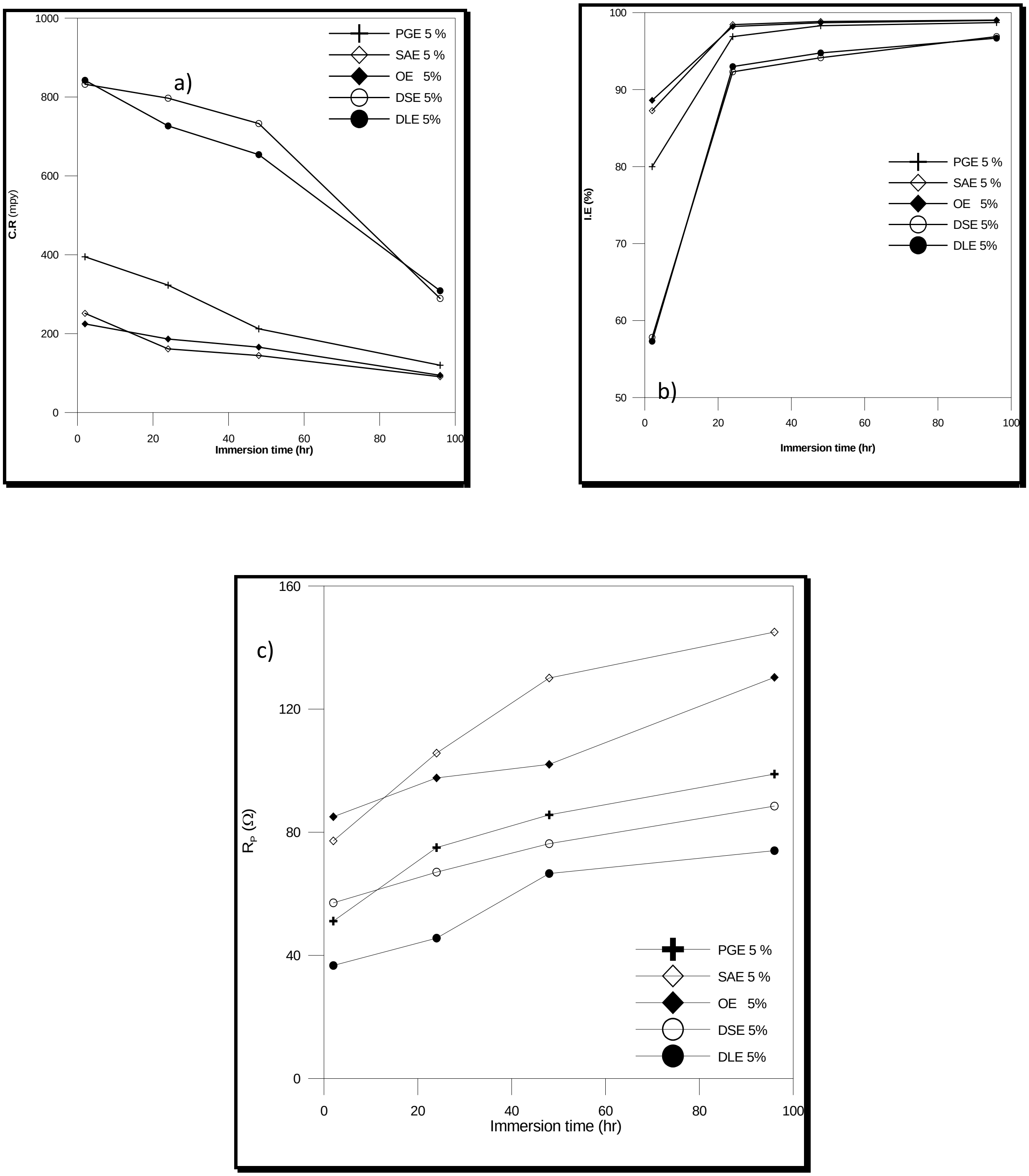

Figure 3

Time dependence of a) corrosion rate, b) inhibition efficiency and c) resistance polarization for mild steel exposed to natural product extracts at 1.0M H_2SO_4.

Fig. (3) Shows time dependence of immersion times against corrosion rate, inhibition efficiency IE% and resistance polarization R_p, it reveals as the immersion time increase from 2 hrs to 96 hrs the R_p for corrosion metal in the tested solutions of natural product extracts increases that follows the CR of metal decreases and IE% increases to reach 99.03% for SAE at immersion time 4 days.

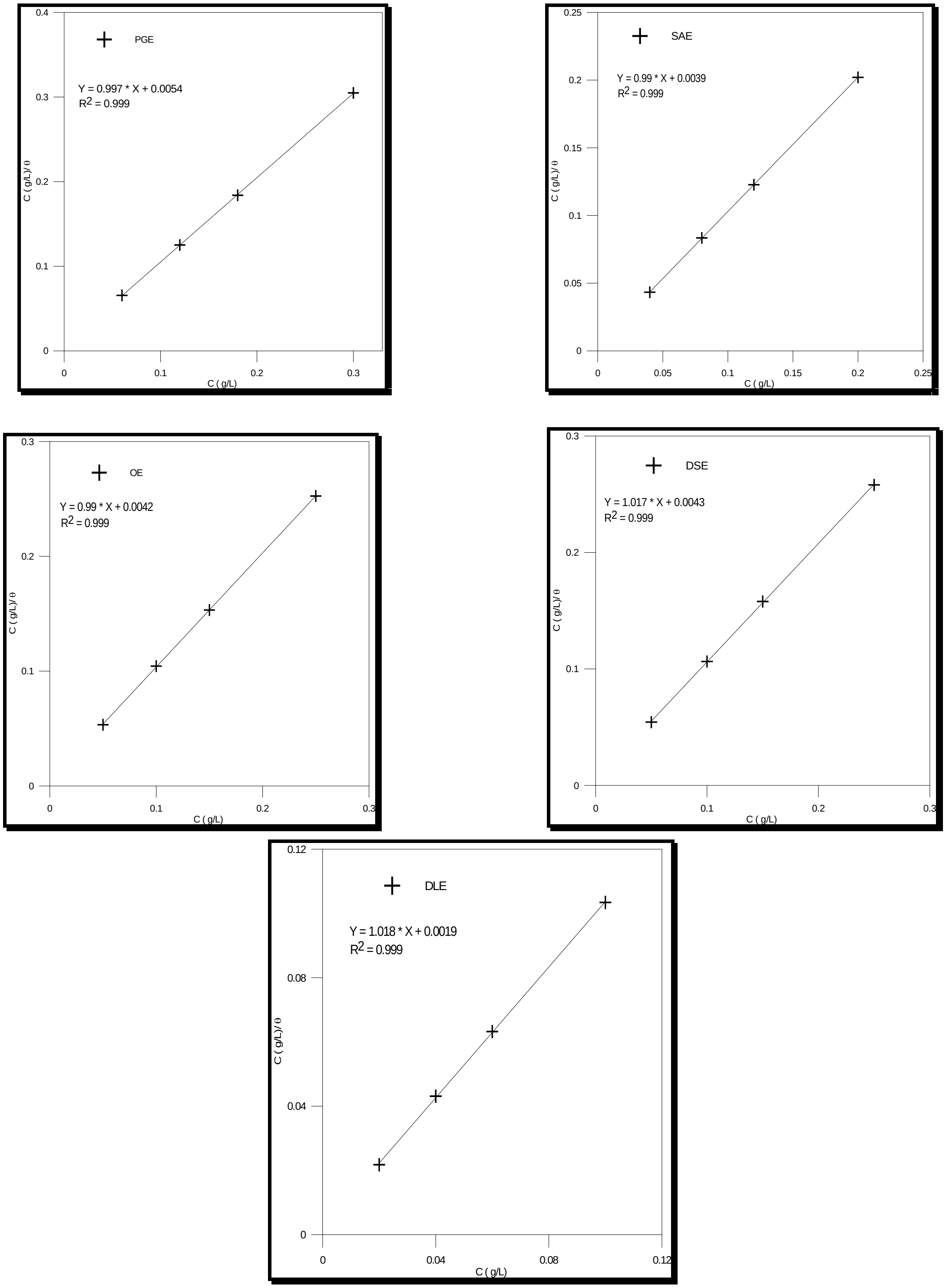

Figure 4
Langmuir adsorption isotherm for adsorption of natural product extracts on the surface of mild steel in 1.0 M H_2SO_4 after4 days.

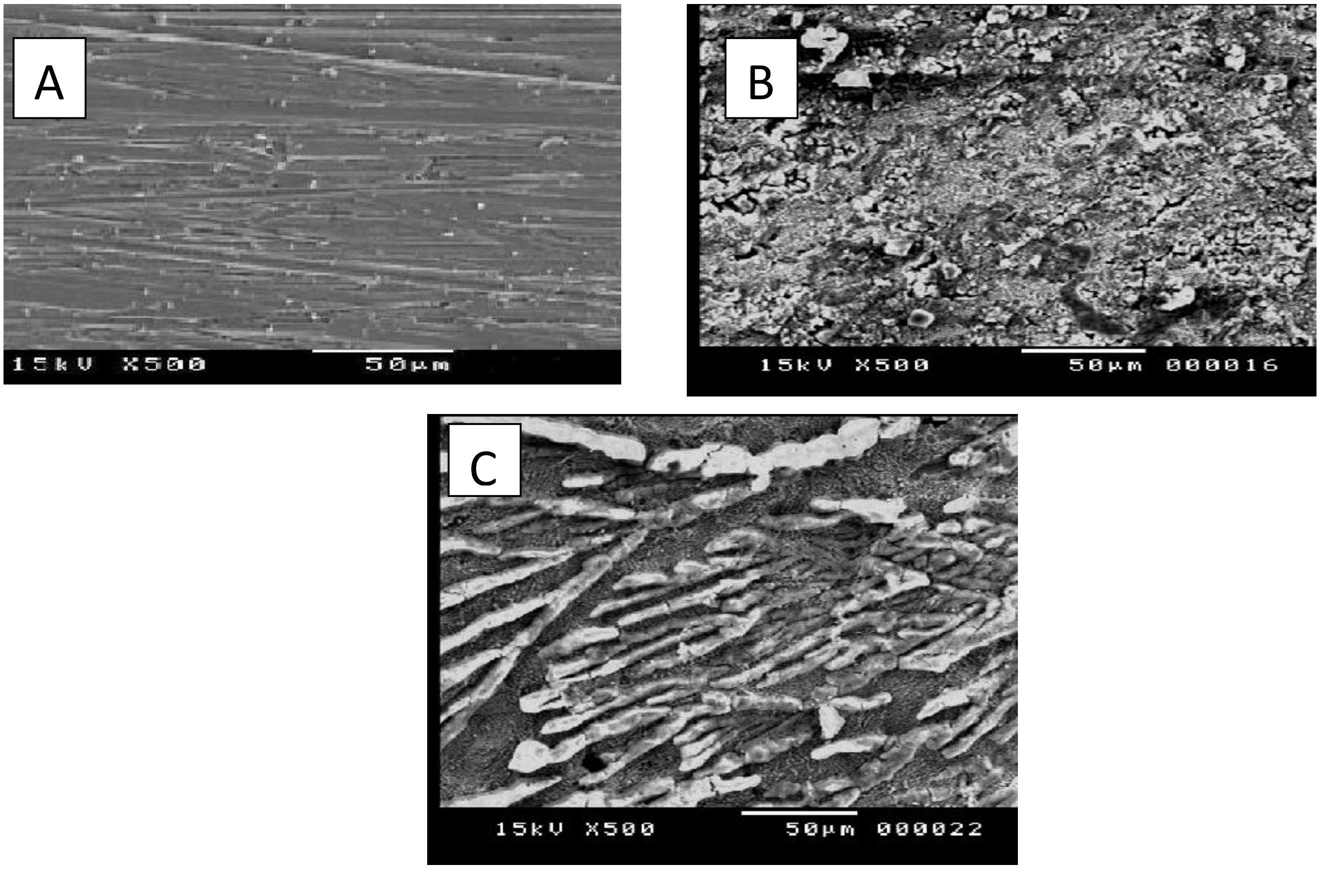

Figure 5
SEM micrographs of the morphologies of (A) Polished MS, (B) MS+1.0M H_2SO_4 and (C) MS+1.0M H_2SO_4 + 5% SAE.

Table 1
Values of E_{imm} and $E_{s.s}$ (mV) for mild steel in 1.0M H_2SO_4 and different concentrations of natural products at different exposure times

Test Solution	C V/V%	2hr		1day		2days		4days	
		$-E_{in}$	$-E_{s.s}$	$-E_{in}$	$-E_{s.s}$	$-E_{in}$	$-E_{s.s}$	$-E_{in}$	$-E_{s.s}$
H_2SO_4	1.0M	489	492	500	500	501	503	532	540
PG	1	464	464	454	466	474	477	474	477
	2	437	440	448	449	444	450	434	449
	3	444	449	392	433	451	454	447	453
	5	449	450	428	444	428	444	432	448
SA	1	470	472	472	473	471	472	468	469
	2	476	479	436	437	468	469	459	463
	3	434	447	455	466	452	457	455	461
	5	444	452	421	443	454	459	447	454
O	1	472	472	470	475	472	480	479	480
	2	459	460	458	455	453	459	474	457
	3	465	466	456	458	465	468	476	479
	5	455	459	450	453	444	453	470	471
DS	1	460	466	454	457	451	451	478	479
	2	441	449	460	464	462	458	456	466
	3	449	456	458	465	466	470	462	468
	5	447	452	452	455	449	455	458	465
DL	1	464	466	461	465	429	438	478	479
	2	439	443	463	465	461	462	451	454
	3	451	461	435	440	453	459	457	460
	5	439	448	424	428	423	431	407	428

Table 2
Polarization parameters for mild steel in 1.0M H_2SO_4 at different concentrations of natural products after 2hr and 1day.

Tested Solution	C (V/V)%	R_P (Ω)	ß$_a$ mV\decade	ß$_c$ mV\decade	-E_{corr} (mV)	I_{corr} (μ A/Cm^2	C.R (may)	I.E (%)	Surface coverage (θ=IE/100)
	2hrs								
$H_2SO_4$1.0M	----	8.992	59.68	120.1	461	1973	1819	------	------
PG	1	13.86	101.8	179.6	438	1531	1411.64	28.45	0.285
	2	15.21	97.25	152.08	422	1250	1152.76	41.57	0.416
	3	31.44	75.23	132.5	426	1050	968.14	50.93	0.509
	5	51.14	40.58	119.58	428	428.2	394.82	79.99	0.800
SA	1	20.15	72.83	153.2	448	1206	1111.98	43.64	0.436
	2	23.32	44.65	107.5	445	972.6	896.77	54.55	0.545
	3	30.25	57.76	140	412	524.1	483.24	75.51	0.755
	5	77.20	31.72	56.91	432	272.7	251.44	87.26	0.873
O	1	14.84	48.14	112.5	441	1107	1020.69	48.27	0.483
	2	25.91	45.30	150.9	437	740.2	682.49	65.41	0.654
	3	36.54	24.44	42.79	438	436	402.01	79.62	0.796
	5	85.04	21.90	43.57	423	243.7	224.70	88.61	0.886
DS	1	12.79	93.49	168.3	439	1602	1477.10	25.13	0.251
	2	15.98	88.25	133.58	428	1405	1295.46	34.34	0.343
	3	25.25	72.58	129.47	432	1212.3	1117.78	43.35	0.433
	5	57.07	66.58	114.78	424	902.65	832.28	57.82	0.578
DL	1	12.42	102.03	133.09	437	1503.2	1386.00	29.75	0.298
	2	25.84	95.24	118.54	409	1389.2	1280.89	35.08	0.351
	3	30.16	87.45	110.22	447	1010.5	931.72	52.78	0.528
	5	36.71	75.88	101.87	414	913.56	842.34	57.31	0.573
	1day								
$H_2SO_4$1.0M	------	7.549	176	260	465	10360	9552	------	------
PG	1	18.20	87.99	145.89	444	780.05	719.23	93.06	0.931
	2	42.43	75.18	173.1	423	589.4	543.45	94.75	0.948
	3	55.18	99.90	195.88	413	512.2	472.27	95.44	0.954
	5	75.02	63.40	160.13	422	350.12	322.82	96.88	0.969
SA	1	24.58	188.0	127.12	455	1186.6	1094.09	89.44	0.894
	2	27.83	102.5	144.00	402	556.9	513.48	95.04	0.950
	3	39.03	75.12	125.17	453	402.12	370.77	96.42	0.964
	5	105.7	43.68	99.9	420	174.9	161.26	98.44	0.984
O	1	15.78	182.6	203.5	439	1045	963.53	90.70	0.907
	2	42.99	179.5	197.8	439	720.05	663.91	93.59	0.936
	3	73.03	113.0	97.5	421	409.9	377.94	96.35	0.964
	5	97.64	77.2	54.87	443	202.1	186.34	98.20	0.982
DS	1	14.79	174.8	190.5	438	1510.2	1392.46	86.56	0.866
	2	25.29	123.2	141.5	451	1400.5	1291.31	87.54	0.875
	3	39.64	110.5	138.5	446	1179.5	1087.54	89.50	0.895
	5	67.05	104.5	128.25	434	864.36	796.97	92.31	0.923
DL	1	14.80	189.8	217.7	459	1410.25	1300.30	87.45	0.874
	2	26.62	177.5	203.8	428	1221.9	1126.64	89.13	0.891
	3	39.21	156.2	190.87	415	916.3	844.86	91.84	0.918
	5	45.61	145.2	174.5	413	788.05	726.61	92.99	0.930

Table 3
Polarization parameters for mild steel in 1.0M H_2SO_4 at different concentrations of natural products after 2 days and 4 days.

Tested Solution	C (V/V)%	R_P (Ω)	$ß_a$ mV\decade	$ß_c$ mV\decade	$-E_{corr}$ (mV)	I_{corr} (µA/Cm^2)	C.R (may)	I.E (%)	Surface coverage (θ=IE/100)
	2 days								
H_2SO_4 1.0 M	------	5.478	288.21	330.28	489	12510	11534.67	------	------
PGE	1	20.14	123.4	159.9	443	750.25	691.76	94.47	0.945
	2	55.03	115.28	144.4	436	530.04	488.72	96.09	0.961
	3	65.21	102.58	128.8	430	495.6	456.96	96.35	0.963
	5	85.64	90.47	114.25	429	230.12	212.18	98.30	0.983
SAE	1	55.32	110.52	130.54	475	1133	1044.67	91.65	0.916
	2	59.23	29.19	64.01	443	625.7	576.92	95.39	0.954
	3	63.02	20.83	30.28	423	386.1	356.00	97.15	0.972
	5	130.11	14.58	27.87	436	156.7	144.48	98.85	0.988
OE	1	20.64	196.8	284.9	468	980.6	904.15	92.77	0.928
	2	55.84	130.8	230.2	438	690.02	636.22	94.91	0.949
	3	75.60	49.18	176.2	443	389.2	358.86	97.13	0.971
	5	102.04	41.91	166.9	431	179.6	165.60	98.68	0.987
DSE	1	23.60	113.4	226.4	432	1334.6	1230.55	90.16	0.902
	2	38.23	89.68	186.9	428	1312.5	1210.17	90.33	0.903
	3	59.39	52.62	81.96	445	1111.9	1025.21	91.80	0.918
	5	76.30	31.37	48.69	433	794.58	732.63	94.14	0.941
DLE	1	24.03	91.48	163.6	412	1302.3	1200.77	90.40	0.904
	2	37.69	53.71	104.5	435	1044.5	963.07	92.30	0.923
	3	58.26	51.38	90.17	439	820.8	756.81	93.95	0.940
	5	66.59	43.44	123.5	414	709.1	653.82	94.77	0.948
	4 days								
H_2SO_4 1.0 M	-----	3.187	402.54	444.2	533	13100	12078.67	------	------
PGE	1	29.37	157.24	197.58	436	515.65	475.45	94.89	0.949
	2	58.60	122.55	187.25	429	295.02	272.02	97.07	0.971
	3	72.13	103.25	134.25	432	210.2	193.81	97.92	0.979
	5	98.90	72.25	119.47	433	130.02	119.88	98.71	0.987
SAE	1	97.80	172.2	199.58	463	780.12	719.30	92.26	0.923
	2	103.03	60.40	128.1	446	412.25	380.11	95.91	0.959
	3	107.05	48.61	129.6	435	219.27	202.17	97.83	0.978
	5	145.03	29.32	62.92	427	98.25	90.59	99.03	0.990
OE	1	32.25	120.5	185.4	451	620.25	571.89	93.85	0.938
	2	63.35	53.21	145.4	439	420.69	387.89	95.83	0.958
	3	80.12	55.42	124.8	449	213.54	196.89	97.88	0.979
	5	130.32	34.76	62.53	443	102.35	94.37	98.99	0.990
DSE	1	30.82	113.8	208.4	441	805.9	743.07	92.01	0.920
	2	55.35	86.90	155	433	601.25	554.37	94.04	0.940
	3	63.28	29.80	47.29	436	509.2	469.50	94.95	0.950
	5	88.50	21.23	26.39	437	313.58	289.13	96.89	0.969
DLE	1	30.35	129.5	236	432	815.8	752.20	91.91	0.919
	2	40.05	96.88	200.2	413	723	666.63	92.83	0.928
	3	60.50	27.46	48.69	423	512.04	472.12	94.92	0.949
	5	74.02	1.085	30.29	473	334.8	308.70	96.68	0.967

Table 4
Values of Langmiur adsorption parameters at exposed mild steel to different extracts in 1.0 M H_2SO_4.

Inhibitor	Immersion time	Slope≈	$LogK_{ads}$	R^2	$-\Delta G_{ads.} kjmol^{-1}$
PG	2hrs	0.662	0.704	0.838	13.97
	1 day	1.021	2.41	0.999	23.73
	2 days	1.007	2.40	0.999	23.65
	4 days	1.003	2.50	0.999	24.21
SA	2hrs	0.814	1.17	0.968	16.65
	1 day	0.9913	2.29	0.981	23.02
	2 days	0.992	2.37	0.999	23.46
	4 days	0.990	2.41	0.999	23.72
O	2hrs	0.883	1.22	0.997	16.91
	1 day	0.995	2.20	0.995	22.51
	2 days	0.995	2.30	0.996	23.09
	4 days	0.994	2.38	0.999	23.53
DS	2hrs	1.126	0.79	0.965	14.46
	1 day	1.062	2.18	0.999	22.39
	2 days	1.0481	2.33	0.999	23.24
	4 days	1.0173	2.37	0.982	23.47
DL	2hrs	1.2466	1.31	0.950	17.44
	1 day	1.055	2.67	0.999	25.16
	2 days	1.0404	2.84	0.999	26.13
	4 days	1.0175	2.72	0.999	25.48

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