

Recycling of some plastic waste as green corrosion inhibitors for steel in 1 M phosphoric acid

R.S.Abdel Hameed^{1,2*}, Naser S.Elmetery², Nawaf F.Alshemary², Hassan A.Shehata¹

¹Faculty of Science, Department of Chemistry, Al-Azhar University, 11884, Cairo, (EGYPT)

²Faculty of Science, Department of Chemistry, Hail University, Hail, 1560, KSA, (SAUDI ARABIA)

E-mail: r.abdelghany@uoh.edu.sa, mredars2@yahoo.com

ABSTRACT

The reaction of soft drink bottles waste with ethylene di amine (EA) and ethylene glycol (EG) in the presence of sodium acetate and manganese acetate as a catalyst, the depolymerization products were separated, characterized, and evaluated as corrosion inhibitors of C-steel alloy in 1M phosphoric acid corrosive medium using simple chemical technique (weight loss) and electrochemical technique (open circuit potential) at different concentration of the used inhibitors derived from soft drink waste. The effect of temperature on the corrosion inhibition efficiency was studied; the inhibition efficiency increases with increasing the concentration of the inhibitors and decreases with increasing the temperature, the inhibition occurs through adsorption of the inhibitor molecules on the metal surface without modifying the mechanism of corrosion process. The adsorption of the inhibitor on Steel surface was found to follow the Langmuir adsorption isotherm. Thermodynamic function of activation and adsorption was computed.

© 2016 Trade Science Inc. - INDIA

KEYWORDS

Recycling;
Plastic waste;
Phosphoric acid;
Corrosion inhibitors;
Steel and adsorption
isotherm.

INTRODUCTION

Polyethylene terephthalate (PET) is a worldwide used polymer, and packaging is one of its most important applications. Due to its high resistance to the atmospheric and biological agents, this polymer is not considered as biodegradable. PET is not a hazardous product, but its waste quantity increases drastically and so it is a good candidate for recycling. PET waste can be recycled by different methods like physical recycling and chemical recycling. Chemical recycling is the reaction of PET with

various reagents to obtain products that are used in the chemical industry^[1]. In the 1980s, polyethylene terephthalate, (PET) began to be used popularly for the production of disposable soft drink bottles and in 1987, more than 700 million pounds of PET were consumed in their production^[26]. As a result of the diversity of its applications in a high volume of consumer products, large amount of PET waste is also generated, which includes polymer manufacturing waste as well as the products after the end of their useful life. (PET) is a worldwide used polymer, and packaging is one of its most important applications.

Full Paper

Corrosion problem is one of the major concerns in the oil and gas industry. Therefore protection of metallic structures against corrosion regarded as a critical subject. Acid solutions were widely used in industrial acid cleaning, acid descaling, acid pickling and oil-well acidizing. In these acid solutions corrosion inhibitors are required in order to restrain the acid erosion of metallic materials^[2]. The adsorption of organic molecules at the metal/solution interface is of a great interest in surface science and can markedly change the corrosion resistance of the metal^[3]. Most of the well-known acid inhibitors are organic compounds containing nitrogen, sulfur and/or oxygen atoms^[4-23]. Steel is one of the most widely used materials of construction and frequently used in manufacture of the pipelines due to the aggressiveness of the liquids which carried by them. These liquids may be petroleum containing water and sulfur. All kinds of water passed through these lines contain chlorides, nitrate, sulfate and phosphate anions. For this reason, the injection of corrosion inhibitors through different sites of pipe is very important. Steel is used in fabrication of reaction vessels, storage tanks, etc. by industries, which either manufacture or use phosphoric acid as reactants. Phosphoric acid is a major product which has many important uses, especially in the production of fertilizers. Most of the acid is produced from phosphate rocks and acid solution of H_2SO_4 or HCl . Crude H_3PO_4 from the attack stage is filtered and concentrated in evaporation unit^[24] there is a great need to protect steel material used in phosphoric acid industry from corrosion^[25].

In the previous works^[26-31], PET plastic waste was used as cheap and safe corrosion inhibitors for C-steel in hydrochloric acid, sulphuric acid, nitric acid, acetic acid, and sodium chloride corrosive media.

The present work is one from a series aimed to alleviate of environmental pollution by converting PET waste into useful products, and to evaluation the inhibitive action of plastic waste on the corro-

sion of some metals and alloys, in different aqueous medium. In the present work, Ethyleneglycol (EG) and Ethylene diamine (EA) was used to convert PET waste to water soluble oligomers, the prepared materials (oligomers) as green corrosion inhibitors for mild steel in $1M H_3PO_4$ at different temperatures were evaluated.

EXPERIMENTAL

Materials

Poly(ethyleneterephthalate) (PET) waste is collected from beverage bottles. Ethylene diamine (EA), Ethylene glycol (EG), manganese acetate and sodium acetate were obtained from Aldrich Chemical Co., England. Mild steel specimens were used throughout experiments; mild steel specimens were composed from the following metal, TABLE 1

Aggressive solution

The aggressive solution ($1 M H_3PO_4$) was prepared by appropriate dilution of analytical grade H_3PO_4 with double distilled water.

Recycling process

The reaction of PET with ethylene di amine (EA)

PET waste was depolymerized with ethylene di amine (EA), at weight ratio of PET to amine (EA) 1:2 (wt% of PET: wt% of EA) using 1.0% of sodium acetate as catalyst (by weight based on weight of PET). The reaction mixtures were heated under vigorous stirring in nitrogen atmosphere at temperature about $170-190^\circ C$ for 4 h and at $200^\circ C$ for 3 h. The temperature of the reaction was then lowered to $100^\circ C$ for 1 h. The mixture was allowed to cool to room temperature. At the end of the reaction, distilled water was added in excess to the reaction mixture with vigorous agitation to precipitate out of the product, poly(bis(2-amino ethylene) terephthal) amide (EA). White yellow crystalline powder of (EA) was obtained by recrystallization in distilled water, which have the chemical structure as obtained

TABLE 1 : Composition of the mild steel specimens

Element	C	Mn	P	Mo	Al	Sn	V	Nb	Ni	Cu	Fe
Weight (%)	0.17	0.057	0.011	0.022	0.011	0.005	0.004	0.007	0.027	0.043	Balance

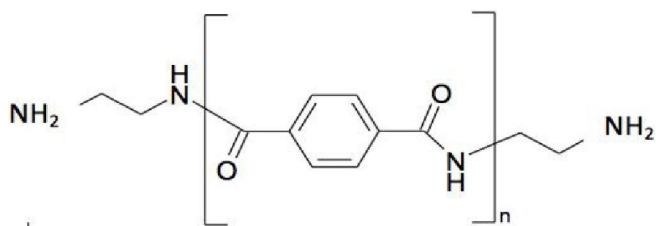


Figure 1a : Chemical Structure of (EA)

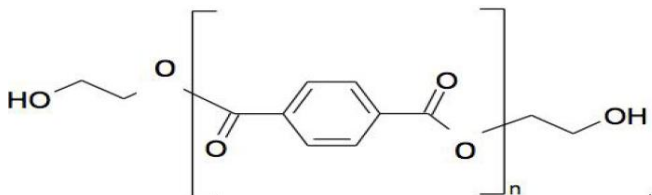


Figure 1b : Chemical structure of (EG)

Figure 1 : the chemical structures for the tow inhibitors prepared from recycling of PET plastic waste in Figure (1 a).

The reaction of PET with ethylene glycol (EG)

PET waste was depolymerized with ethylene Glycol (EG), at weight ratio of PET to Glycol (EG) 1:2 (wt% of PET: wt% of EG) using 1.0% of manganese acetate as catalyst (by weight based on weight of PET). The reaction mixtures were heated under vigorous stirring in nitrogen atmosphere at temperature about 170–190 °C for 4 h and at 200 °C for 3 h. The temperature of the reaction was then lowered to 100 °C for 1 h. The mixture was allowed to cool to room temperature. at the end of the reaction, distilled water was added in excess to the reaction mixture with vigorous agitation to precipitate out of the product, poly(bis(2-Hydroxy ethylene) terephthalate) (EG). White crystalline powder of (EG) was obtained by recrystallization in distilled water, which have the chemical structure as obtained in Figure (1 b).

Weight loss measurements

Coupons of steel of 4 x 8x 0.1cm dimensions were used as test specimens, the specimens were polished by 410 and 610 emery papers, respectively, degreased with acetone, washed with distilled water and finally dried using two filter papers. The described treatment was carried out immediately before each measurement. The specimen of the given metal was immersed in 100 ml of the test solution (i.e., 1 M H₃PO₄) with different inhibitors concentrations, samples maintained in test solution upto

8hours. Experiments were carried out under different temperature 25, 35, 45, and 45°C

RESULTS AND DISCUSSION

Evaluation of the prepared oligomers as green corrosion inhibitors

The use of inhibitors is one of the most practical methods for protection against corrosion in acidic media^[28]. The choice of the inhibitor is based on two considerations:

First: it could be synthesized conveniently from relatively cheap raw materials,

Secondly: it contains the electron cloud on the aromatic ring or the electronegative atoms such as N₂ and O₂ in the relatively long chain compounds^[29].

In this respect, the present corrosion inhibitors were prepared from recycled PET waste and introducing nitrogen and oxygen atoms in their chemical structure to increase their abilities to dissolve in water and to use as green corrosion inhibitors for steel. The corrosion of steel in 1 M phosphoric acid solution at different temperatures was studied by weight loss as a simple chemical techniques.

Weight loss measurements

Effects of inhibitor concentration on inhibition efficiency

Effects of the inhibitors concentrations (EA) and (EG) on the steel corrosion rate were summarized on TABLE 1, and graphically represented in Figures 2, 3.

Figure 2,3 shows the results of the Weight-loss time curves of steel in 1.0 M H₃PO₄ in presence of different concentration of EA and EG inhibitors

In general, a decrease in the weight loss of steel occurred in the presence of a different concentration of additive compared to the blank was observed.

Data in TABLE 2 used to calculate the values of surface coverage area (θ), efficiency of inhibitor (P%) were calculated using equations 1 and 2^[28-30]

$$\theta = \frac{W^\circ - W}{W^\circ} \quad (1)$$

$$P(\%) = \frac{W^\circ - W}{W^\circ} \times 100 \quad (2)$$

Full Paper

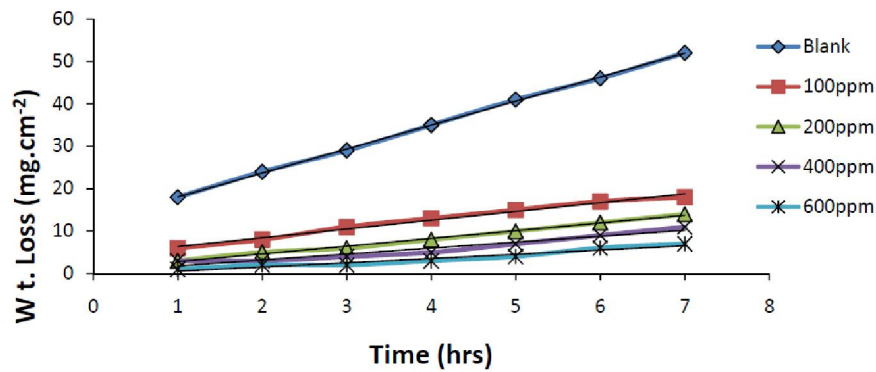


Figure 2 : Weight loss–time curves of steel in 1.0 M H_3PO_4 in absence and presence of different concentrations of EA inhibitor derived from plastic waste

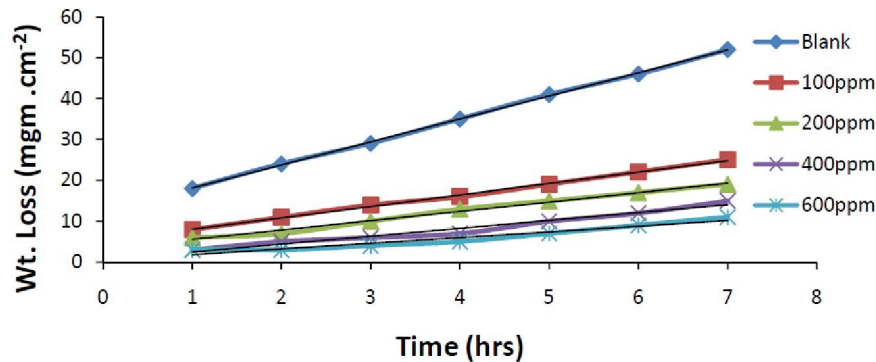


Figure 3 : Weight loss–time curves of steel in 1.0 M H_3PO_4 in absence and presence of different concentrations of EG inhibitor derived from plastic waste

TABLE 2 : Effect of inhibitors concentrations on steel corrosion in 1.0 M H_3PO_4 at 25°C Obtained from weightloss measurements after 8 hours

Sample	Conc. ppm	Wt. loss $mg.cm^{-2}$	CR(mpy)	Corrosion rate (k) $mg.cm^{-2} hr^{-1}$	θ	P%
Blank	0	62.8	170.32	6.48	---	---
	100	13.2	68.53	2.46	0.88	88.7
	200	10.3	63.57	1.9	0.90	90
	400	8.4	47.65	1.5	0.94	94
	600	6.9	32.46	0.9	0.98	98
EA	100	15.3	69.78	2.8	0.86	86
	200	12.5	66.6	2.1	0.88	88
	400	10.2	54.45	1.9	0.92	92
EG	600	9.8	39.84	1.3	0.95	95

Where W^o and W are the weight loss in the absence and presence of inhibitor, respectively. W is the loss of weight after corrosion (mg), A the total area of the specimen (cm^2), t the corrosion time (hr), d the specimen density (g/cm^3).

Corrosion rates (CR) of steel (equation 3) were calculated by considering the total affected sample area and immersion times.

$$CR(mpy) = \frac{3445.15W}{A.d.t} \quad (3)$$

As we can see from TABLE 2, the addition of inhibitors results in a remarkable decrease of the corrosion rate of steel. In fact these results support the inhibitive effect of the added inhibitors on steel corrosion in the acidic solution. Moreover, as we can see from TABLE 2, increasing the inhibitors concentrations increases surface coverage area (θ) and inhibition efficiency ($P\%$).

Adsorption Isotherms

If one supposes that the adsorption of inhibitor

follows the Langmuir adsorption isotherm, the surface coverage could be given by the equation:

$$C_i/\theta = (1/K_{ads}) + C_{inh} \quad (4)$$

Where, C_i is the inhibitor concentration and K_{ads} is the adsorption equilibrium constant. The degree of surface coverage (θ) for different concentrations of the inhibitors in acidic media,^[27].

Careful inspection of these results showed the inhibitors also that, inhibition efficiency increases with increasing inhibitor concentration and surface coverage.

In the present work, it was observed that, all isotherms have slope less than unity the deviation from unity may be explained on the basis of interaction between the adsorption species on metal surface^[23]. The inhibition of these compounds due to their adsorption and formation of adsorbed monolayer at metal surface interface. The surface coverage values (θ) were tested graphically for fitting a suitable adsorption isotherm. The relation C_i/θ against C_i illustrated in Figure (4).

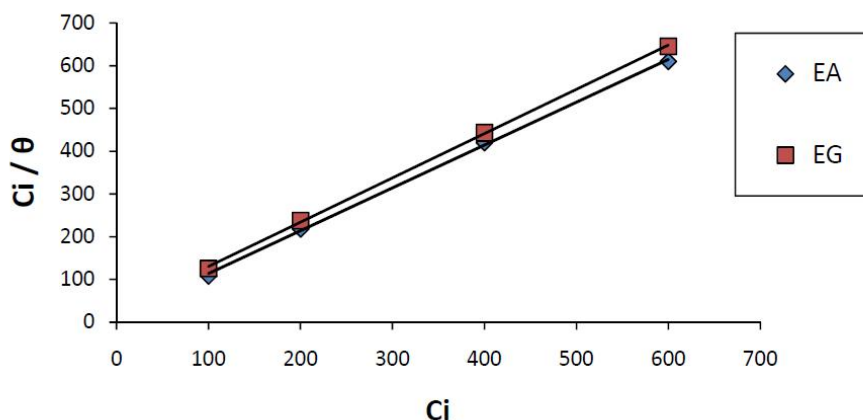


Figure 4 : Langmuir adsorption isotherm for EA and EG Inhibitors

TABLE 3 : Degree of surface coverage (θ) and percentage of inhibition efficiency (P %) in the presence of 600 ppm of the inhibitors EA and EG in 1M H_3PO_4 at different temperatures calculated from weight loss data

Compound	T °C	T, °K	(1/T) × 10 ⁻³	C.R. mg.cm ² .hr ⁻¹	Log C.R.	P%	θ	E* _{active} , k.J.mol ⁻¹
EA	25	298	3.3	0.9	-0.045	98	0.98	118.34
	35	308	3.24	1.1	-0.041	97	0.97	
	45	318	3.14	1.2	-0.079	95	0.95	
	55	328	3.04	1.6	-0.204	93	0.93	
EG	25	298	3.3	1.2	-0.079	95	0.95	104.56
	35	308	3.24	1.6	-0.204	93	0.93	
	45	318	3.14	1.9	-0.278	91	0.91	
	55	328	3.04	2.2	-0.34	87	0.87	

Effects of the reaction temperature on inhibition efficiency

Temperature effects on inhibition efficiency were also investigated, TABLE 3 summarizes the results of surface coverage area (θ), inhibition efficiencies (P%), and corrosion rates (CR) of inhibitors EA, EG for the corrosion of steel at different temperatures. Results of the effect of EA surfactant on the corrosion behavior of steel in 1.0M H_3PO_4 solution at different temperatures using weight loss technique are shown in Figure 5. It has been found that the weight loss of steel increased with increasing temperature. TABLE 3 gives the values. Surface coverage area and inhibition efficiency were decreased with increase in temperature, and the corrosion rate of steel increases with increasing temperature.

Activation energy of corrosion

Corrosion is an electrochemical phenomenon and consequently follows the laws in chemical kinetics. The corrosion rate increases with temperature as a result of decreasing the apparent activation energy,

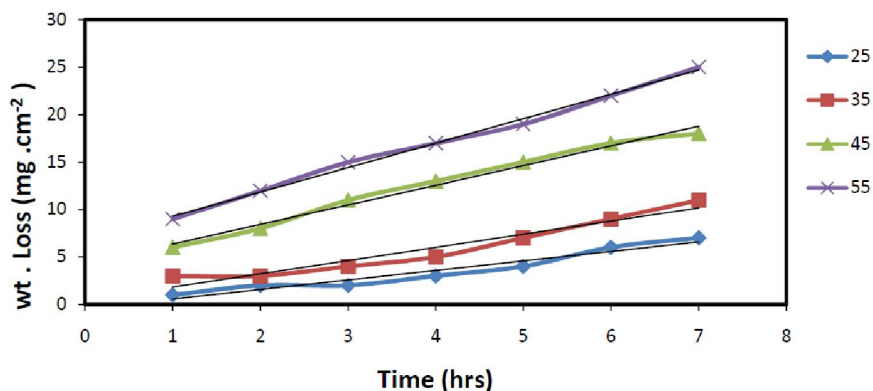


Figure 5 : Weight loss–time curves of steel in 1.0 M H_3PO_4 in presence of 600 ppm of EA inhibitor at different temperatures

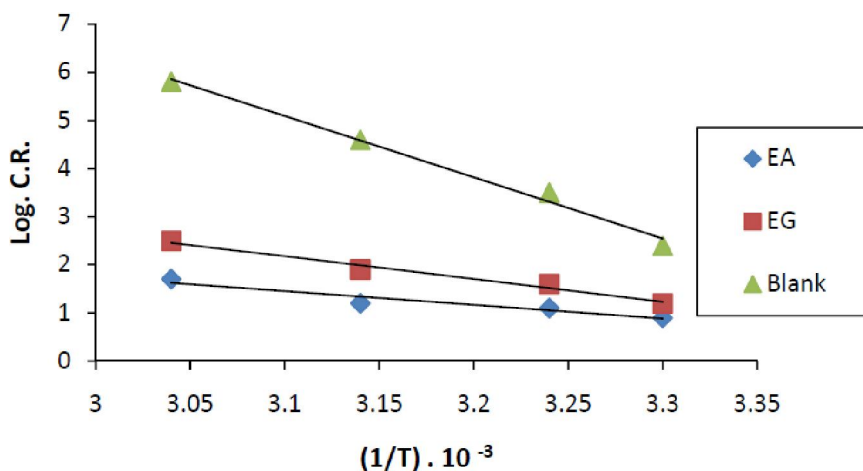


Figure 6 : Relation between log C.R. against $1/T$ at different temperature in presence and absence of 600 ppm of EA and EG

E_a of the charge transfer reactions. Increasing temperature also enhances the rate of H^+ ion diffusion to the metal surface beside the ionic mobility, which increases the conductivity of the electrolyte. The effect of temperature on the inhibition efficiency of corrosion inhibitors is important in elucidation of the mechanism and kinetics of their action. Moreover, at lower temperature, adsorbed hydrogen atoms (exothermic process) blocked the cathodic area.

The activation energy (E_a) can be calculated from Arrhenius equation^[27].

$$\text{Log } K_{\text{rate}} = \text{log } A - E_a/2.303RT \quad (5)$$

A is pre-exponential factor related to concentration, steric effect and metal surface characteristics, etc. Plotting of log C.R. against $1/T$ for the free acid solution (blank) and 600 ppm of the EA and EG inhibitors were represented in Figure 6. The activation energies were calculated and listed in TABLE 3. The results showed that, the values of activation

energy (E_a) increase in the same order of increasing the inhibition efficiency of the inhibitors. It is also indicated that the whole process is controlled by surface reaction^[26].

Thermodynamic parameters

The free energy of adsorption (ΔG_{ads}^*) at different temperatures was calculated from the following equation:

$$\Delta G_{\text{ads}}^* = RT (\text{log } K_{\text{rate}} - \text{log } KT/h) \quad (6)$$

K , is equilibrium constant, which given by:

$$K = \theta / Ci (1 - \theta) \quad (7)$$

The values of E_a^* and ΔG_{ads}^* were listed in TABLE (4). The negative values of ΔG_{ads}^* indicate that the spontaneous adsorption of inhibitors on the surface of carbon steel. The negative values of ΔG_{ads}^* also suggest the strong interaction of the inhibitor molecules onto the steel surface^[26]. The thermodynamic functions of inhibitors adsorption, entropy, ΔS_{ads}^* , enthalpy, ΔH_{ads}^* and free energy, ΔG_{ads}^*

are calculated from the equations:

$$\Delta H_{\text{ads}}^* = E_a^* + RT \quad (8)$$

Where, R is the universal gas constant. Entropy of inhibitor adsorption (ΔS_{ads}^*) can be calculated using the following equation:

$$\Delta S_{\text{ads}}^* = (\Delta H_{\text{ads}}^* - \Delta G_{\text{ads}}^*)/T \quad (9)$$

The calculated ΔS_{ads}^* data were listed in TABLE (4). All obtained data show that ΔS_{ads}^* values are positive and increased by increasing temperature which indicate that the inhibitors more oriented and more ordered on the surface of the metal. TABLE (4) showed being that ΔH_{ads}^* has negative values, indicating that, the reaction is exothermic. On the other hand, the negative value of ΔH_{ads}^* indicated that the adsorption of inhibitors is a chemical one and the surfactant forms stable layer at the surface of

steel. This behavior protects steel from the environmental conditions (H_3PO_4) and decreased the corrosion rate.

Open circuit potential measurements

The potential of mild steel electrodes immersed in 1 M H_3PO_4 solution was measured as a function of immersion time in the absence and presence of 600 ppm of the two inhibitors derived from plastic waste as shown in Figure (7) It is clear that the potential of mild steel electrode immersed in 1M H_3PO_4 solution (blank curve) tends towards more negative potential firstly, giving rise to short step. This behavior due to breakdown of the pre-immersion air formed oxide film presents on the surface followed by the growth of a new oxide film inside the solu-

TABLE 4 : Thermodynamic activation parameters of adsorption for steel in 1.0 M H_3PO_4 in absence and presence of 600 ppm of the inhibitors at different temperatures

Compound	Temp., K	E^*	$-\Delta G^*$, $\text{kJ}\cdot\text{mol}^{-1}$	$-\Delta H^*$, $\text{kJ}\cdot\text{mol}^{-1}$	ΔS , $\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$
Blank	298	62.4	29.232	15.86	0.1395
	308		29.256	16.23	0.1467
	318		30.08	16.85	0.1454
	328		31.44	17.63	0.1429
	298		43.45	17.74	0.102
EA	308	118.34	45.94	18.66	0.104
	318		48.26	19.14	0.105
	328		50.10	20.42	0.107
	298		50.34	17.26	0.110
EG	308	104.56	55.82	18.34	0.118
	318		57.06	20.04	0.120
	328		58.14	21.25	0.129

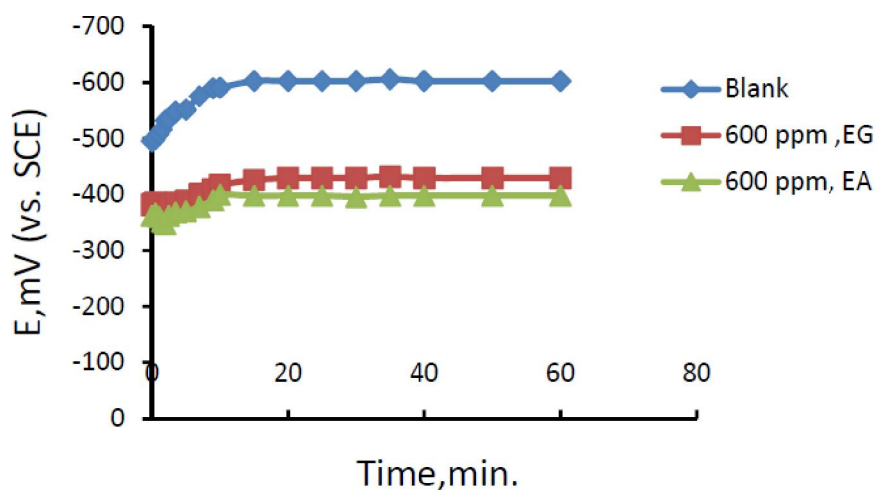


Figure 7 : Potential–time curves for carbon steel immersed in 1M phosphoric acid solution in the absence and presence of 600 ppm of the used inhibitors

Full Paper

tion, so that the potential was shifted again to more noble direction until steady state potential is established^[31-33]. Addition of inhibitor molecules to the aggressive medium produces a negative shift in the open circuit potential due to the retardation of the cathodic reaction. The shift to noble direction by amide inhibitor, EA more than ester inhibitor, EG, derived from plastic waste.

CONCLUSION

Based on the obtained results, the following conclusions are accomplished: -

1. Soft drink bottles can be depolymerized (recycled) into water soluble oligomers when it react with ethylene diamine and ethylene glycol in the presence of catalyst
2. The recycling process without solvent considered as green process
3. The oligomers obtained from waste are excellent inhibitors for steel corrosion in phosphoric acid solution.
4. Inhibition efficiencies increased by increasing inhibitor concentration and by decreasing temperatures.
5. The activation parameters of the adsorption (E^* , ΔH^* and ΔS^*) were calculated and showed that the used inhibitors decrease the rate of corrosion.
6. The adsorption of these inhibitors on steel surface obeys Langmuir's adsorption isotherm.
7. The corrosion inhibition efficiency of the prepared oligomer of EA is greater than EG. Due to the presence of amide bond (CONH) and amino group (NH_2) in its structure.
8. The two inhibitors obtained from plastic waste considered as green corrosion inhibitors for steel in acidic medium at low concentration and high temperature up to 55 °C.
9. Addition of inhibitor molecules to the aggressive medium produces a negative shift in the open circuit potential due to the retardation of the cathodic reaction.

REFERENCES

- [1] G.Colomines, J.Robin, G.Tersac; *Polymer*, **46**, 3230 (2005).
- [2] S.Yao, X.Jiang, L.Zhou, Y.Lv, X.Hu; *Mater.Chem.Phys.*, **104**, 301 (2007).
- [3] R.Solmaz, G.Kardas, M.C.ulha, B.Yazici, M.Erbil; *Electrochim.Acta*, **53**, 5941 (2008).
- [4] F.Bentiss, M.Lebrini, M.Lagrenée, M.Traisnel, A.Elfarouk, H.Vezin; *Electrochim.Acta*, **52**, 6865 (2007).
- [5] R.S.Abdel Hameed, H.I.Al-Shafey, A.S.AbulMagd, H.A.Shehata; *Journal of Materials and Environmental Science*, **3**(2), 294 (2012).
- [6] R.S.Abdel Hameed; *Portugaliae Electrochimica Acta*, **29**(4), 273 (2011).
- [7] R.S.Abdel Hameed; *Al Azhar Bull Science*, **20**(1), 151 (2009).
- [8] R.S.Abdel Hameed et al.; *Der Chemica Sinica*, **3**(1), 236-248 (2012).
- [9] M.A.Migahed, M.A.Abd El Raouf, A.M.Al Sabagh, H.M.Abd El Bary; *J.Appl.Electrochem*, **36**, 395 (2006).
- [10] M.A.Migahed, I.F.Nassar; *Electrochim.Acta*, **53**, 2877 (2008).
- [11] M.A.Migahed, A.M.Al Sabagh, N.M.Nasser, Gh.N.Kandile; *J.Disp.Sci.Tech.*, **29**, 161 (2008).
- [12] M.Tariq Saeed, Sk.Asrof Ali, S.U.Rahman; *Anti-Corros.Meth.Mater.*, **50**, 201 (2003).
- [13] M.Elachouri, M.S.Hajji, S.Kertit, E.M.Essasi, M.Salem, R.Coundert; *Corros.Sci.*, **37**, 381 (1995).
- [14] B.Mernari, H.El Attari, M.Traisnel, F.Bentiss, M.Lagrennee; *Corros.Sci.*, **40**, 391 (1998).
- [15] G.Schmitt; *Brit.Corros.J.*, **19**, 165 (1984).
- [16] K.F.Khaled, K.Babic-Samardzija, N.Hackerman; *J.Appl.Electrochem.*, **34**, 697 (2004).
- [17] K.F.Khaled; *Electrochim.Acta*, **48**, 2493 (2003).
- [18] L.Tang, X.Li, Q.Qu, G.Mu, G.Lin; *Mater.Chem.Phys.*, **94**, 353 (2005).
- [19] O.K.Abiola; *Corros.Sci.*, **48**, 3078 (2006).
- [20] F.Bentiss, F.Gassama, D.Barbry, L.Gengembre, H.Vezin, M.Lagrennee, M.Traisnel; *Appl.Surf.Sci.*, **252**, 2684 (2006).
- [21] A.Popova, M.Christov, S.Raicheva, E.Sokolova; *Corros.Sci.*, **46**, 1333 (2004).
- [22] T.Arslan, F.Kandemirli, E.E.Ebenso, I.Love, H.Alemu; *Corros.Sci.*, **51**, 35 (2009).
- [23] J.Cruz, T.Pangarasu, E.Garcia ochoa; *J.Electroanal.Chem.*, **583**, 8 (2005).
- [24] T.Sydberger, S.Nordin; *Corrosion*, **34**, 16 (1978).
- [25] S.T.Arab, A.M.Al Turkustani; *Portugaliae Electrochimica Acta*; **24**(53), 275 (2006).
- [26] R.S.Abdel Hameed; *Advances in applied science*

- research, **2(3)**, 483 (2011).
- [27] R.S.Abdel Hameed; H.I.Alshafy, O.Farghaly; Research & Reviews in Electrochemistry, **3(2)**, 1-14 (2012).
- [28] R.S.Abd El Hameed, H.I.AL Shafey, S.A.Soliman, M.S.Metwally; Al Azhar Bull.Sci., **19**, 283 (2008).
- [29] R.S.Abd El Hameed, Ph.D.Thesis; Al Azhar Uni., (2006).
- [30] R.S.Abd El Hameed, H.I.AL Shafey, E.A.Ismail; Al Azhar Bull.Sci., **20**, 185 (2009).
- [31] R.S.Abdel Hameed, Hussin IAl-Shafey, E.A.Ismail, Abd H.Alhakeem, Abu-Nawwas, El O.E.Azabawy; International Journal of Engineering Research and Applications, **3(6)**, 1094-1103 (2013).
- [32] R.S.Abdel Hameed, HI Al Shafey, A.H.Abu-Nawwas; International Journal of Electrochemical Science, **9**, 6006-6019 (2014).
- [33] R.S.Abdel Hameed, Hussin IAl-Shafey, E.A.Ismail, Abd H.Alhakeem, Abu-Nawwas; Expired voltaren drugs as corrosion inhibitor for aluminium in hydrochloric acid, Int J Electrochem Sci., **10**, 2098 – 2109 (2015).