The Inhibitive Effect of Some Amino Acids on the Corrosion Behaviour of 316L Stainless Steel in Sulfuric Acid Solution

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Abstract

The inhibition effect of four amino acids on the corrosion of 316L stainless steel in 1.0 M H_2SO_4 has been studied by open-circuit potential and potentiodynamic polarization measurements. Corrosion data such as corrosion rate, corrosion potential ($E_{Corr.}$) and corrosion current ($I_{Corr.}$) were determined by extrapolation of the cathodic and anodic Tafel region. Glycine, Leucine and Valine inhibit the corrosion process, but Arginine accelerate the corrosion phenomenon. Glycine has the highest inhibition efficiency, its efficiency increases with increasing the concentration to attain 84.2% at 0.1M. Different behaviours were observed, in case of Arginine, corrosion rate increase by increasing concentration. These results show that the presence of Arginine at high concentration turns the surface of 316L stainless steel electrochemically active, probably dissolving the passivation layer and promoting the stainless steel anodic dissolution. Results obtained from potentiodymaic and open-circuit potential measurements are in good agreement.

Keywords: Amino acids, 316L stainless steel, Sulfuric acid

1. Introduction

Austenitic stainless steel are widely used in various chemical industries, due to their superior corrosion protection through a passive oxide layer forms on their surfaces, when exposing to an aqueous environments. Although the main characteristic of these materials is high resistance to corrosion, but when coming in contact with chloride ions, they undergo of localized corrosion and lose their passivity properties (A. Pardon, M.C. Merino, A.E. Coy, F. Viejo, R. Arrabal, E. Matykina. 2008)(I. Gurappa. 2002)(T.L. Sudesh, L. Wijesinghe, D.J. Blackwood, 2006) (C.V. Vidal, A.I. Munoz. 1954) (P. Fauvet, F. Balbaud, R. Robin, Q.T.Tran, A. Mugnier, D. Espinoux, 2008). Sulphuric acid is commonly used in removing the undesirable scale and rust in metal working, such as cleaning of boilers and heat exchangers(Muzaffer zcan, Ramazan Solmaz, Gülfeza Kardas, 2008)(F. Bentiss, M. Bouanis, B. Mernari, M. Traisnel, H. Vezin, M. Lagrenée, 2007)(Nabel A. Negm, Mohamed F. Zaki, 2008). Since acids are an aggressive medium for oil and gas well equipment, the effective way to reduce their corrosion attack on tubing and casing materials, is to inject a suitable corrosion inhibitor which could be added to the acid solution during the acidifying process (C. Kustu, K.C. Emregul, O. Atakol. 2007)(M.A. Migahed, I.F. Nassar. 2008)(M.A. Ouraishi, D. Jamal. 2000)(M.A. Ouraishi, D. Jamal, 2001). For austenitic stainless steel, the electrochemical tests revealed generally low resistance to pitting for hydrochloric acid concentrations of 0.1, 0.5, and 0.9 M (J. Alhajji. 1997). Comparative study of surface properties of austenitic stainless steels in sulfuric and hydrochloric acid solutions (K. Varga, P. Baradlai, W.O. Barnard, G. Myburg, P. Halmos and J.H. Potgieter. 1997), from the experimental results, it is obvious that the surface excess values of bisulfate/sulfate ions on both stainless steels (316L and 304L) are much higher (up to 1.5 x 10⁻⁹ mol cm⁻²) and their interaction with the passive oxide layer is substantially stronger than those of chloride ions. The surface concentration of Cl of less than 2 x10⁻¹⁰ mol cm⁻² provides evidence that only a very limited part of the real surface area of the steel samples (less than 5 %) is occupied by Cl ions, ie, pitting corrosion sites are probably formed. The corrosion behavior of stainless steel samples (316L and 304L) in HCl and H₂SO₄ solution has been studied using potentiodynamic, cyclic voltammogram, EDX and SEM techniques(D.J. Shaw, 1966).

Amino acids are nontoxic, relatively cheap and easy to produce with purities greater than 99% (R.C. Bohinski. 1973). The use of chemical inhibitors is the most practical method for the protection against corrosion in acidic solutions (M. Mihit S. El Issami, M. Bouklah, L. Bazzi, B. Hammout, E. Ait Addi, R. Salghi, S. Kertit. 2006)(M. Mihit, R. Salghi, S. El-Issami, L. Bazzi, B. Hammouti, I. Ait Addi, S. Kertit, Pigm. 2006)(A. Dafali, B. Hammouti, R. Mokhlisse, S. Kertit, 2003)(S. Kertit, R. Salghi, L. Bazzi, B. Hammouti, A. Bouchtart, Ann. 2000). Several studies have been performed on the use of organic compounds as inhibitors for the corrosion of metals in aggressive acidic media. Most of the natural amino acids are the alpha amino acids which contain carboxyl and amino groups bonded to the same carbon atom. It was shown that the inhibition action of some organic compounds is based on adsorption phenomenon (L. Bazzi, S. Kertit, M. Hamdani. 1995). Inhibition effect of

hexadecyl pyridinium bromide on the corrosion behavior of some austenitic stainless steels in H_2SO_4 solutions has been studied (M.M. Hamza, S.S. Abd El Rehim, A.M. Ibrahim. 2011). Electrochemical corrosion behavior and hydrogen evolution reaction of 316L stainless steel has been investigated in 0.5 M sulfuric acid solution containing four novel organic inhibitors as derivatives from one family, using potentiodynamic polarization, electrochemical impedance spectroscopy (EIS) measurements and surface examination via scanning electron microscope (SEM) techniques (Nada F. Atta, A.M. Fekry, Hamdi M.Hassaneen, 2011). Fourier-transform infrared spectroscopic analysis and semiempirical molecular orbital calculations were carried out to analyse the ionization states and configuration of amino acid adsorption on a stainless steel surface.

The aim of this research, may throw some light on the corrosion inhibition of some aliphatic amino acids on 316L stainless steel in sulfuric acid solution.

2. Experimental

Circular electrode with working surface area of 1.76cm^2 were used. The chemical composition given in Table 1. Electrode was ground with emery paper and polished with diamond paste (1µm), finally the sample was cleaned by acetone, washed by distilled water, and quickly inserted in the cell. All experiments were carried out under total immersion in 100ml of test solutions. Saturated calomel electrode (SCE) and platinium electrode were used as reference and auxiliary electrodes, respectively. The stirred electrolyte used, as a corrosive media was 1.0 M H₂SO₄ aerated solution, prepared from analytical reagent grade concentrated acid using distilled water at $25\pm$ 2°C. The open-circuit potential of the metal immersed in the test solutions were monitored using the electronic multimeter (Type E scord-EDM-2116). The electrode was allowed to stabilize in the electrolyte until the potential change was < 0.1 mV/min. This potential was taken as the steady state potential (E_{s.s.}). Potentiodynamic polarization studies were carried out in the potential range \pm 250 mV vs corrosion potential (E_{Corr.}) at a scan rate of 0.5 mV/s. The studies were performed with model 352/252 corrosion measurement system, which consists of EG & G potentiostat/galvanostat model 273 A.

The amino acids used in those investigations are Glycine, Leucine, Valine and Arginine as example for aliphatic amino acids at different concentrations range from 0.005 - 0.1 M in 1.0M H₂SO₄ solution.

3. Results and discussion

3.1 Open-circuit potential measurements

The open-circuit potentials of stainless steel electrode were traced about 90 min. from electrode immersion in solutions. Generally the steady state was reached within 60 min. from electrode immersion in the test solution. Steady state potential is shifted either in the negative or positive direction in the presence of the amino acids. The direction of the potential shift depends on the type and concentration of the amino acids. Fig.1, represents the variation of the open-circuit potentials with time of 316L stainless steel electrode in 1.0 M H_2SO_4 solution in absence and in presence of different concentrations of amino acids. The values of the steady state potentials ($E_{s.s.}$) and the immersion potential ($E_{im.}$) in the presence of different concentrations of different amino acids are given in Table 3.

The variation of the steady state potentials ($E_{s.s.}$) with the logarithmic of the molar concentrations of test solutions, is presented by the curves in Fig.2. According to the change of the steady state potentials with log C, the amino acids studied may be classified into two classes. The first one comprised (Arginine, Leucine and Valine), while the second belong Glycine. The results show that the open-circuit potentials and the steady state potentials depend on the type and concentration of the amino acids Table (3).

In class one which include Leucine, Valine and Arginine, steady state potential values were shift towards more negative values with increasing the amino acid concentration. This behaviour can be attributed to the influence of the inhibitor on the anodic process, especially at high inhibitor concentrations (K.M. Ismail. 2003). These results show that the presence of amino acids (Leucine, Valine and Arginine) at high concentrations turns the surface of 316L stainless steel electrochemically active, probably dissolving the passivation layer and promoting the stainless steel anodic dissolution (A.B. Silva, S.M.L. Agostinho, O.E. Barcia, G.G.O. Cordeiro, E.D⁺ Elia, 2006).

Inspection of the potential-time curves in case of Glycine Fig.1 (a), steady state potential shift towards more noble values. This can be attributed to the formation of a protective layer of Glycine on the electrode surface (E.E. Orguzie, S.G. Wang, Y.Li, F.H.Wang. 2008).

3.2 Potentiodynamic polarization measurements

Effect of some amino acids on 316L stainless steel in aqueous 1.0 M H₂SO₄.

Organic corrosion inhibitors function by chemisorption of their molecules on metallic substrate, complexing of the molecules with metal ions, that remain in solid lattices, neutralizing the corrodent and/or absorbing the corrodent. Many organic compounds containing polar group and π electrons are good corrosion inhibitor for steels (A.M. Badawi, S.T. Keera and M.A. Hussein, 2001)(T. Giordano, 1987). In acidic solutions, these compounds exist as protonated species. This protonated species may adsorb on the cathodic sites of the stainless steel surface according to Langmuir isotherm, and decrease the evolution of hydrogen. And also are able to

adsorb on anodic sites through nitrogen atom. The adsorption of these compounds on anodic sites may decrease anodic dissolution of the stainless steel electrodes. The inhibition of organic compounds is strongly depended on the structure and chemical properties of the layer formed on the metal surface under particular experimental conditions(S.A.M. Refaey, F. Taha and A. M. Abd El-Malak. 2004). Such behaviour may also be considered in line with an earlier suggestion, that a molecule may first physically adsorb, and then slowly react with the metal surface to form a chemisorbed layer (D.J. Shaw, 1966).

a- Effect of Glycine

Fig.3 shows the polarization curves for the effect of different concentrations of Glycine on the potentiodynamic behaviour of 316L stainless steel in 1.0 M H_2SO_4 solution. Generally with increasing inhibitor concentration the corrosion current density ($I_{Corr.}$) and corrosion rate decrease. It indicates that, according to increasing, inhibitor concentration its adsorption to metal surface increases and protects the metal from corrosion. The inhibition effect of glycine towards the corrosion of low alloy steel ASTM A213 grade T22 boiler steel was studied in aerated stagnant 0.5 M HCl solutions in the temperature range 20-60 °C , the results show that glycine is good "green", mixed-type inhibitor with cathodic predominance(Mohammed A. Amin, Sayed S. Abd El Rehim, Hesham T.M. Abdel-Fatah, 2009).

The corrosion rate can be calculated from the following equation:

Corrosion rate (mpy) =
$$\frac{0.13 \text{ I}_{\text{corr.}}(\text{E.W})}{\text{A.d.}}$$

Where:

E.W. = equivalent weight (in g.eq.)

 $A = area (in cm^2)$

 $d = density (g./cm^3)$

0.13 = metric and time conversion factor.

Also the inhibitor efficiencies calculated from the polarization method using the equation,

$$IE = \frac{(I_{Corr})_{uninh} - (I_{Corr})_{inh.}}{(I_{Corr})_{uninh}} \times 100$$

Where $(I_{Corr.})_{uninh.}$ and $(I_{Corr.})_{inh.}$ are the corrosion current densities obtained by extrapolation of the cathodic and anodic Tafel lines, in uninhibited and inhibited solutions, respectively. All parameters were measured and reported in Table 4.

According to the results, $E_{Corr.}$ shifts to more negative (less positive) values and that indicates, in this case, the inhibitor have been adsorbed to cathodic areas and acts as cathodic inhibitor (H. Ashassi-Sorkhabi, M.R. Majidi, K. Seyyedi, 2004).

The amino acid presents in its protonated form in acidic solution, therefore, the amino acid in this form can be attracted to the cathodic sites on the metal surface.

It is clear from the potentiodynamic experiments that the presence of Glycine, decrease the corrosion rate, i.e. the values of $I_{Corr.}$ decreases. Furthermore, the addition of Glycine shifts, the potential of metal in the negative direction due to the decrease in the rate of the cathodic reaction. Particularly, the cathodic reaction is inhibited to larger extent than the anodic reaction (K.M. Ismail. 2007).

The inhibition efficiencies increase as the inhibitor concentration increases and reach maximum values of 84.2% (at 0.1M Glycine) (W.A. Badawy, K.M. Ismail, A. M. Fathi. 2006).

b- Effect of Leucine

Leucine is a passivating inhibitor, as it is found to decrease the passivation current (Puja Singh, Kalpana Bhrara Gurmeet Singh. 2008). The various corrosion parameters, like $E_{Corr.}$, $I_{Corr.}$, corrosion rate and inhibition efficiency in presence of different concentrations of Leucine calculated and tabulated in Table 5. The recorded results in Fig. 4 indicated that, the corrosion current ($I_{Corr.}$) are lower in the presence of Leucine as compared to the acid free solution. From the results, found that E_{Corr} shift towards more negative values by increase the concentration of Leucine, indicate that, Leucine act as cathodic inhibitor. The corrosion inhibition process is based on the adsorption of the amino acid molecules on the active sites and/or deposition of the corrosion products on the metal surface(W.A. Badawy, K.M. Isamil, A.M., Fathi, 2005)(G. Bereket, A. Yurt, 2001). In the present case owing to high acidity of the medium, Leucine is present in the protonated form as(J.P. Greenstein, M. Winitz, 1961):



Therefore, it is clear that in the strong acid medium under consideration both the carboxylic and the amino groups of the additive are expected to be fully protonated. Both the functional groups get adsorbed on the cathodic sites of the corroding metal. The following figure represents the schematic presentation of the mode of adsorption of amino acids over the metal surface (A.S. Fouda, M.M.E. Semongy. 1982).



C-Effect of Valine

The anodic and cathodic curves observed in Fig. 5, in absence and presence of Valine and the results obtained tabulated in Table 6, show that the corrosion rate of 316L stainless steel decrease in case of presence of Valine than in case absence of Valine. However, the addition of Valine inhibits the corrosion phenomenon.

In the case of Valine, nitrogen atom in aliphatic chain increases the interaction of molecule with metal surface (J.P. Greenstein, M. Winitz, 1961). Inspection of the results in Table 6, $E_{Corr.}$ shifts to more positive values by increasing the concentration of Valine, in this case, Valine acts as anodic inhibitor (W.A. Badawy, K.M. Ismail, A. M. Fathi. 2006). Further increase in the inhibitor concentration shows a slight decrease in the inhibition efficiencies. This phenomenon is consistent with results obtained for other inhibitors and is attributed to the dissolution of the adsorbed film (P. Gupta, R.S. Chaudhary, T.K.G. Nam bood hiri, B.Prakash, Br. Corros. J. 1982). The corrosion inhibit behaviour of Valine in acidic solution may be due to the formation of Fe-Valine complex, this complex can form stable passive layer on the stainless steel surface, leads to the decreasing of corrosion rate(E.E. Orguzie, S.G. Wang, Y.Li, F.H.Wang. 2008). From the results, found that, the inhibition efficiency depends on the concentration of the inhibitor.

d-Effect of Arginine

Arginine accelerates the corrosion rate and it relate to complexing effect of Arginine. Glycine, Leucine and Valine as example for aliphatic amino acids, while Arginine as basic amino acids (W.A. Badawy, K.M. Ismail, A. M. Fathi. 2006).

In case of Arginine solution, as the formation of the complexes between metal and amino acid (Arginine), since Arginine acting as bidentate ligand, this behyajour, results in change in the corrosion potential of 316L stainless steel in the active direction, followed by increase in the corrosion rate. All parameters were observed and are given in Table 7. Inspection of this results, observed that, corrosion potential of 316L stainless steel shifts to more negative values, while I_{Corr} and corrosion rate increase by increasing the concentration of Arginine. These results suggest that the presence of Arginine, promoted an more active surface. Fig. 6, shows polarization curves for the effect of different concentrations of Arginine in 1.0 M H₂SO₄. From the Fig. 6, observed that, all curves exhibit active dissolution without any distinctive transition to passivation within the studied potential range(D.J. Shaw. 1966). The presence of the active region suggests, a dissolution of the passivation layer, probably due to the Arginine complexes formed with the metallic ions present in this film. Consequently, the knowledge of their corrosion activity is highly essential. Further, the effect of length of C-atom chain(O. Olivares – Xometl, N.V. Likhanova, M.A. Dominguez-Aguilar, E. Arce, H. Dorantes, P. 2008), along aliphatic chain promoted not only higher corrosion efficiency, but also a higher toxicity. Arginine has a radical R which contains nitrogen, effectiveness remains low because the existence of a tautomeric and a steric hindrance at nitrogen atoms(M.Zerfaoui, H.Oudda, B. Hammouti, S.Kertit, M. Benkaddour. 2004). These results in Table 7, were in agreement with some authors, have shown that some amino acids have been demonstrated to be able to enhance the corrosion process(W. Sand. 1997)(H.H.P. Fang, L.C. Xu, K.Y. Chan. 2002).

4. Conclusion

The present work involves open-circuit potential and potentiodynamic polarization techniques for studying the effect of different amino acids on the corrosion of 316L stainless steel in $1.0M H_2SO_4$ solution.

From the obtained results, we may conclude:

1) In general, amino acids with longer hydrocarbon chain showed greater inhibition, addition of amino groups or groups which increase electron density on alph amino group also increased the inhibition efficiency.

2) It is evident from the obtained values of $E_{Corr.}$, $I_{Corr.}$ and inhibition efficiencies that, the decrease in the corrosion rate associated with positive or negative shift in the corrosion potential.

3) Glycine, Valine and Leucine act as corrosion inhibitors but Arginine act as corrosion accelerator.

4) The inhibition efficiency follows the following order:

Glycine (84.2%) > Valine (38.2%) > Leucine (33.5%). So Glycine..is the best inhibitor

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Table 1. Chemical composition of 316L stainless steel electrode.

Amount of elements (%)									
Si	Cr	Ni	Mn	Мо	Fe				
0.56	17.28	10.57	1.14	2.62	67.83				

The structure of the amino acids given in Table 2.

Table 2.	Structure and	molecular	weight	of the	amino	acids	under	study
			~					

Name	Molecular formula	Structure
Glycine	C ₂ H ₅ NO ₂	СН ₂ —СООН NH ₂
Leucine	C ₆ H ₁₃ NO ₂	CH ₃ CH-CH ₂ -CH-COOH CH ₃ NH ₂
Valine	C ₅ H ₁₁ NO ₂	CH ₃ CH-CH-COOH CH ₃ NH ₂
Arginine	$C_6H_{14}N_4O_2$	$NH = C - NH - CH_2 - CH_2 - CH_2 - CH - COOH NH_2$

Amino acids	Concentration	E _{im.} (mV)	E _{s.s.} (mV)
	Μ		
Blank	_	- 195	- 140
	0.005	- 185	- 120
Clusing	0.05	- 182	- 100
Gryeine	0.1	- 190	- 91
Tanaina	0.005	- 230	- 114
Leucine	0.05	- 180	- 137
	0.1	- 225	- 186
Valina	0.005	- 230	- 160
vanne	0.05	- 198	- 185
	0.1	- 235	- 190
	0.005	- 150	- 190
Argining	0.05	- 226	- 205
Argillille	0.1	- 220	- 242

Table 3. Values of E_{im} and $E_{s.s.}$ for 316L stainless steel in 1.0.M H_2SO_4 in absence and in presence of different amino acids.

Table 4. Corrosion parameters and inhibition efficiency for 316L stainless steel in absence and presence of different concentrations of Glycine

Conc.	E _{Corr.}	I _{Corr.}	Connection note (mny)	Inhibition efficiency
Μ	mV	µA/cm ²	Corrosion rate (mpy)	IE%
Blank	66.43	3.280	1485 x10 ⁻³	
Blank + 0.005M Glycine	-57.84	1.879	850.7 x10 ⁻³	42.7
Blank + 0.05M Glycine	-31.86	0.894	405.2 x10 ⁻³	72.7
Blank + 0.1M Glycine	-4.458	0.516	233.7 x10 ⁻³	84.2

Table 5. Corrosion	parameters	and	inhibition	efficiency	for	316L	stainless	steel	in	absence	and	presence	of
different concentrat	ons of Leuc	ine											

Conc.	F mV	$\mathbf{I} = \mathbf{u} \mathbf{A} / \mathbf{a} \mathbf{m}^2$	Corresion rate (mny)	Inhibition efficiency
Μ	L _{Corr.} III v	I _{Corr.} μA/cm	Corrosion rate (mpy)	IE%
Blank	66.43	3.280	1485 x10 ⁻³	
Blank + 0.005M Leucine	48.84	1.420	643.1 x10 ⁻³	56.7
Blank + 0.05M Leucine	28.39	1.442	653.1 x10 ⁻³	56.0
Blank + 0.1M Leucine	-35.42	2.180	990.8 x10 ⁻³	33.5

Table 6.	Corrosion	parameters	and	inhibition	efficiency	for	316L	stainless	steel	in	absence	and	presence	of
different	concentrati	ons of Valir	ne											

Conc . M	E _{Corr.} mV	I _{Corr.} μA/cm ²	Corrosion rate (mpy)	Inhibition efficiency IE%
Blank	66.43	3.280	1485 x10 ⁻³	
Blank + 0.005M Valine	9.275	1.523	703.1 x10 ⁻³	53.5
Blank + 0.05M Valine	24.80	1.832	829.4 x10 ⁻³	44.14
Blank + 0.1M Valine	180.0	2.024	916.4 x10 ⁻³	38.29

Table 7. Corrosion parameters and inhibition efficiency for 316L stainless steel in absence and presence of different concentrations of Arginine

Conc . M	E _{Corr.} mV	I _{Corr.} μA/cm ²	Corrosion rate (mpy)
Blank	66.43	3.280	1485 x10 ⁻³
Blank + 0.005M Arginine	-28.56	4.323	1958 x10 ⁻³
Blank + 0.05M Arginine	-25.75	5.809	2631 x10 ⁻³
Blank + 0.1M Arginine	-44.99	7.882	3569 x10 ⁻³



Figure 1. Variation of the open-circuit potentials of 316L stainless steel with time in 1.0M H₂SO₄ containing (a) Glycine, (b) Leucine, (c) Valine and (d) Arginine



Figure 2. Es.s- log Conc.curves of 316L stainless steel electrode in different amino acids solutions



Figure 3. Polarization curves for the effect of different concentrations of Glycine on the potentiodynamic behaviour of 316L stainless steel in 1.0M H₂SO₄



Protonated form (Acidic solution)



Figure 4. Polarization curves for the effect of different concentrations of Leucine on the potentiodynamic behaviour of 316L stainless steel in 1.0 M H₂SO₄



Figure 5. Polarization curves for the effect of different concentrations of Valine on the potentiodynamic behaviour of 316L stainless steel in 1.0M H₂SO₄



Figure 6. Polarization curves for the effect of different concentrations of Arginine on the potentiodynamic behaviour of 316L stainless steel in 1.0M H₂SO₄