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Amino acid compounds as eco-friendly corrosion inhibitor in acidic media- Review

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Abstract

Corrosion control of metallic materials is of technical, economical, and environmental importance. The most used way to protect metals and alloys against corrosion is to add inhibitors to the aggressive solution. The organic inhibitors act by adsorption to form a barrier on the metal surface and block or retard the reaction of corrosion. The environmental toxicity of organic corrosion inhibitors has reoriented research for green corrosion inhibitors as they are biodegradable and containing the necessary elements such as O, C, N, and S, which are active in organic compounds, assist in adsorption of these compounds on metals or alloys. Amino acids inhibitors fall into this category since they are cheap; most of them are soluble in aqueous media and are easy to produce at high purity. It's the objective of this review.

Keywords: Corrosion, Green inhibitor, Metals, Aminoacid, Aminoester

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1. Introduction

Corrosion is a naturally occurring phenomenon i.e. spontaneous and thermodynamically to return metal at oxidized states as in nature. The economic cost of corrosion may be estimated directly from the application, operation and maintenance of anti-corrosion technologies or indirectly from the loss of productivity, compensation for casualties and environmental pollution, and any other cost that is not directly incurred within that industry [1–3].

The use of inhibitors is one of the best options of protecting metals and alloys against corrosion. According to a standard definition, a corrosion inhibitor is a "chemical substance that when present in the corrosion system at a suitable concentration decreases the corrosion rate, without significantly changing the concentration of any corrosive agent." It is generally effective in small concentrations [4–6]. Corrosion scientists and engineers are more inclined towards the green corrosion inhibitors that are inexpensive, readily available, environmentally friendly and ecologically acceptable, and renewable [7–9].

The known hazardous effects of most synthetic organic inhibitors and restrictive environmental regulations have now made researchers to focus on the need to develop cheap and environmentally benign natural products as corrosion inhibitors. These natural organic compounds are either synthesized or extracted from aromatic herbs, spices and medicinal plants. Plant extracts are viewed as an incredibly rich source of naturally synthesized chemical compounds that can be extracted by simple procedures with low cost and are biodegradable in nature. The use of these natural products such as extracted compounds from leaves or seeds as corrosion inhibitors have been widely reported by several authors [8–10].

Alfa amino acids (AA) form a class of non-toxic organic compounds which are completely soluble in aqueous media and produced with high purity at low cost. Have two polar groups, namely, one amino group and one carboxyl group (Figure 1), AA compounds can coordinate with metals through nitrogen atom and oxygen atom. These properties would justify their use as corrosion inhibitors [9–11].

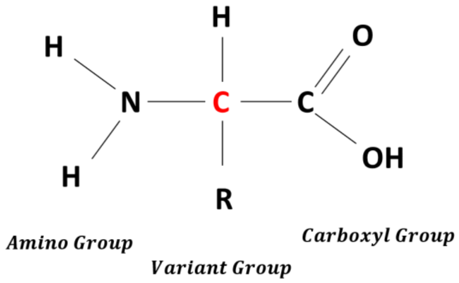


Figure 1 : α-amino acid structure

2. Importance of amino-acids

Amino acids are the monomers that make up proteins. Specifically, a protein is made up of one or more linear chains of amino acids, each of which is called a polypeptide. There are 20 types of amino acids commonly found in proteins [11].

The importance of amino-acids (AA) in viable biological systems is due to the unusual properties of this class of chemical compounds. These substances, possessing both acidic and basic properties within the same molecule, afford the basic linkages leading to complex peptides and proteins, the basic materials of life. Further, ten such amino-acids have been shown to be essential for life processes. Again, due to the basic and acidic character in a given molecule, these substances are dipolar compounds, with either zwitterionic species formation or anionic or cationic species formation depending upon the acidity of the fluid environment [12-14]. The relative concentrations of these species, which coexist with each other, depend completely upon the value of pH. Amino acids can be classified into categories based on the chemistry of their R-groups to non-polar, carboxyl, amine, aromatic,… (Table 1).

Table 1 - Amino acid categories (based on R-group properties)

non-polar	**carboxyl**	**Amine**	**Aromatic**	**Hydroxyl**	**other**
Alanine	Aspartac Acid	Arginine	pjenyalanine	serine	Asparagine
Glycine	Glutamic	Histidine	Tryptophan	Threonine	Cysteine
Isoleucine		Lysin	Tyrosine	Tyrosine	Glutamine
Leucine					Selenocysteine
Methionine					Pyrrolysine
Proline					
Valine					

2. Glycine : most studied

The literature indicated that the most studied amino acid is Glycine as corrosion inhibitor of metals in various aggressive media [15-24]. Since 1968, Grigor'ev and Kusnetsov studied the effect of glycine derivatives as inhibitors of the production of hydrogen in the corrosion of zinc and iron in H_2SO_4 solutions. They found that an increase in the electron acceptor properties of the substituent radical resulted in greater control of the liberation of hydrogen from the metals [15]. Glycine, simplest amino acid: H_2N, CH_2 and COOH (figure 2) can coordinate with metals through the nitrogen atom and oxygen atom of the carboxyl group.

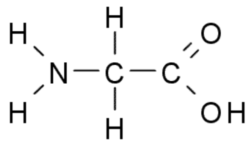


Figure 2 : Glycine structure

Thereafter, Akiyama and Nobe in 1970, The adsorption of amino acid on carbon steel in acidic environment have been investigated by [16]. The presence of heteroatoms, such as N, O, and S and conjugated π-electrons system have made amino acids a significant class of green corrosion inhibitors thanks to their environmental aspect.

3. Methionine

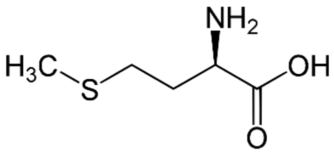
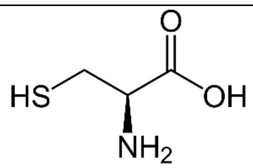
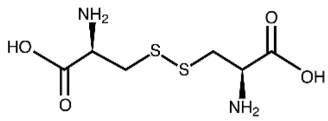
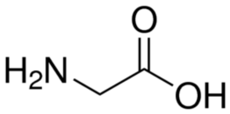
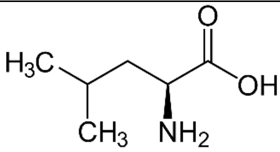
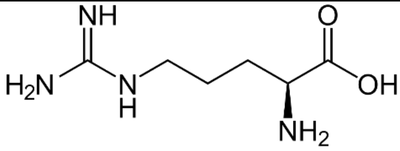
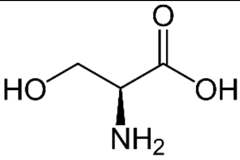
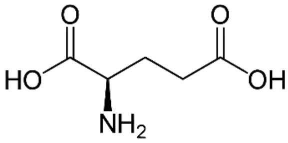
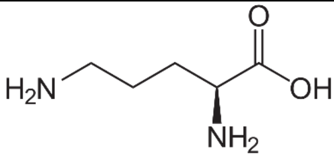
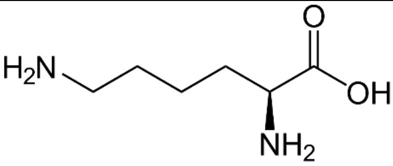
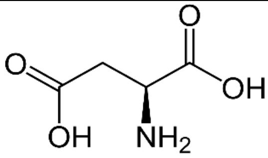
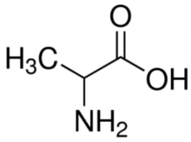
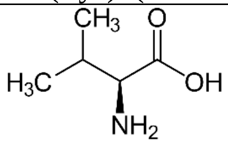
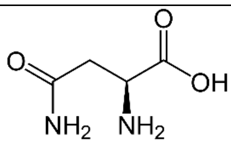
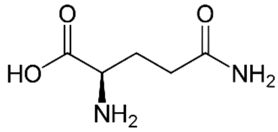
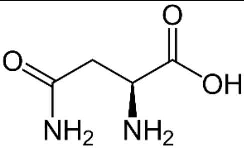
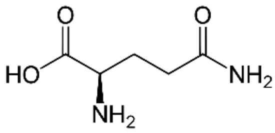
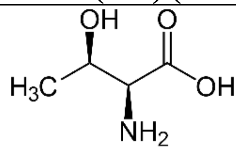
In our laboratory, tests were conducted first on methionine rich in heteroatoms as Sulfur, Nitrogen and oxygen on pure iron in HCl solution. The excellent inhibitory property was patented [25] and published [26]. The methionine can interact with the corroding surface of the steel via the protonated amino groups by adsorbing at the cathodic sites and hindering the hydrogen evolution; or through the sulfur atom in its molecular structure, which may adsorb at the anodic sites reducing the dissolution rate of iron [27-29]. The inhibitory power of methionine is highly increased when carboxyl acid passed to methyl ester, then Its inhibition efficiency (E%) reached a maximum value of 95% at 10^{-2}M. Polarization measurements indicated $METOCH_3$ acted as a cathodic inhibitor without changing the mechanism of the hydrogen evolution reaction. $METOCH_3$ was adsorbed on the iron surface according to a Frumkin isotherm model [26]. The intramolecular synergistic effect of carboxyl and amino groups as well as aliphatic chain containing SH group favoured the adsorption of methionine and methionine methyl ester on the iron surface.

4. Various amino acid compounds

Corrosion tests and electrochemical measurements were performed on Armco iron in HCl solution in the presence of some amino acids containing an active sulphur (methionine: Met, cysteine: Cys, cystine: Cyt) and a sulphur-free amino acids (glycine: Gly, leucine: Leu, arginine: Arg, serine: Ser, glutamic acid: Glu, ornithine: Orn, Lysine: Lys, aspartic acid: Asp, alanine: Ala, valine: Val, asparagine: Asn, glutamine: Gln, and threonine: Thr). Table 2 summarized their molecular structure and corresponding efficiency against corrosion of Armco iron by hydrochloric acid [30].

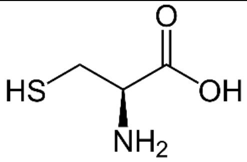
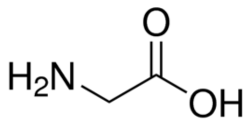
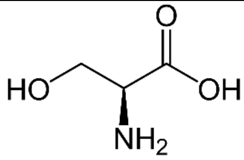
Results obtained indicated that there is a net difference between pure iron and Armco iron [26,30]. Moreover, the efficiency depended on the variable group of amino acid compound. Details were more discussed in Ref. [30]. The efficiency obtained was interpreted on the basis of the nature of variable group ® using the computational method. The highest protection occurred in the presence of methionine. The kind of group also reorient the adsorption phenomenon on the metal surface to form barrier against the aggressive ions and thereafter towards various isotherms such as Langmuir, Temkin and Frumkin … Also, the effectiveness of amino acid on steel corrosion depends on the nature of acid. One AA more efficient in HCl becomes less protector in other acid as H_2SO_4 or H_3PO_4.

Table 2 – 18 Amino acids tested and their efficiency

 methionine: (Met) (E%=72.7)	 cysteine: (Cys) (E%=58.2)	 cystine: Cyt) (E%=60.7)
 Glycine (Gly) (E%=1.6)	 Leucine (Leu) (E%=58.0)	 Agrinine (Arg) (E%=66.9)
 Serine (Ser) (E%=-1.2)	 glutamic acid: (Glu) (E%=7.3)	 Ornithine (Orn) (E%=24.8)
 Lysine (Lys) (E%=36.4)	 aspartic acid (Asp) (E%=14.5)	 Alanine (Al) (E%=11.5)
 Valine (Val) (E%=15.7)	 Asparagine (Asp) (E%=14.5)	 Glutamine (Gln) (E%=7.3)
 Asparagine Asn (E%=26.9)	 Glutamine Gln (E%=14.0)	 Threonine Thr (E%=26.9)

It is also important to see the evolution of efficiency when R changed from :

R : CH_2-SH of cysteine to CH_2-NH_2 to CH_2-OH, Efficiency falls to lower values. Serine became catalyser of Corrosion [30].

 cysteine: (Cys) (E%=58.2)	 Glycine (Gly) (E%=1.6)	 Serine (Ser) (E%=-1.2)

In acidic media, the amino group $-NH_2$ is readily protonated to ammonium ion $-NH_3^+$ as well as the carboxyl group can lose the hydrogen to form a zwitterionic behaviour to facilitate adsorption on the metal surface as shown in Figure 3. This theory of the zwitterion or dipolar type configuration has been further substantiated by Fox and Foster [31]. An important property of the amino acids is that they exhibit an isoelectric point. This is the point in an electric field at which the zwitterion form will not migrate to either the anode or cathode [32-34].

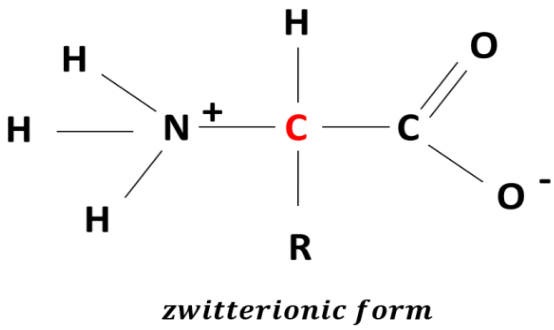


Figure 3 : Protonated amino acid or zwitterion structure

In their study, Gece and Bilgiç compared the calculated quantum chemical parameters of 12 amino acids with their experimental inhibition efficiencies. The quantum chemical study strongly suggests that the amino acids interact with nickel surface in the protonated (zwitterion) rather than in the neutral state, which is compatible with the results of multiple regression analysis. This conclusion is also supported by calculating the energies of the frontier orbitals for (Ni + amino acid) model systems and free energy changes for the interaction of lysine and nickel system [35].

A thorough examination of the available literature (Table 3) revealed that almost tested amino acids compounds have been confirmed a good inhibition performance to use it as the eco-friendly inhibitors against corrosion in various mediums. Then, there is no a universal amino acid compound that is applicable to most of the metal/solution systems. However, the protection capacities of those compounds was depended to their molecular structure, their concentration, nature of corrosive medium, and metallic surface nature and to other factors. Yet, Hammouti *et al* [26], show that some amino esters derivatives can increase the corrosion rate. Except, the compounds contain S atom such as: Methionine and cysteine methyl esters, protect iron against corrosion in molar hydrochloric acid solution.

Table 3 - Amino acids and their derivatives used as corrosion inhibitors in acidic medium

Amino acids inhibitor	Metal/ Electrolyte Conc. Inh	Inhibition Efficiency (%)			Isotherm	Nature of adsorption	Ref
		IE_{EIS}	IE_{PDP}	IE_{WL}			
Cysteine	Copper/ 1M HCl 18mM	78.6	84.1	-	Langmuir	Physical adsorption	[36]
L-phenylalanine + Ce^{4+}	Copper/ 0.5M HCl 5mM	82.7	71.8	-	-	-	[37]
Cysteine-cysteine	Mild steel/ 0.5M H2SO4 5mM	86.0	-	-	Langmuir	Double	[38]
Tetra-n-butyl ammonium methioninate	Mild steel/ 1M HCl 10^{-3}M	-	95.1	-	Freundlich	Physical	[39]
Dodecyl amine of tyrosine	Carbon steel/ 2M H3PO4 50ppm	90.0	91.5	-	Temkin	Physical	[40]
Methionine methyl ester	Iron/ 1M HCl 10^{-2}M	-	91.9	93.0	Frumkin	Chemical	[26]
glycine methyl ester		-	-151	-	-	-	
leucine methyl ester		-	-361	-	-	-	
phenylalanine methyl ester		-	-90.0	-	-	-	
cysteine methyl ester		-	65.6	-	-	-	
L-Alanine	Copper/1M HNO3	-	24.5	18.5	-	-	[41]
Acid Aspartic		-	28.1	35.2	-	-	
L-Leucine		-	33.8	40.7	-	-	
Acid Glutamic		-	37.5	37.0	-	-	
Glutamine		-	40.2	42.6	-	-	
Asparagine		-	45.0	31.5	-	-	
Threonine		-	50.4	48.1	-	-	
Methionine		-	80.4	93.9	-	-	

The inhibition of corrosion of mild steel in sulfuric acid solution by vanillin and several amino acids has been studied by Shaaban et al. [42]. After obtention of moderate efficiency of vanillin and cysteine separately, the authors proposed to study the synergistic effect. It has been found that the highest inhibition efficiency is obtained in the presence of vanillin and cysteine mixture. The inhibition efficiency of 87 % is obtained in the presence of vanillin and cysteine, 0.5 mM each. A significant synergism (synergism parameter = 1.53) between vanillin and cysteine for the corrosion inhibition is exerted. Comparing the corrosion inhibition efficiencies of vanillin and cysteine mixture with the Schiff base synthesized from those compounds probed tentatively the active sites of adsorption of the two species [42]. The synergism parameter was calculated by the following equation proposed by Aramaki and Hackerman [43]:

$$S_\theta = \frac{1-\theta_{1+2}}{1-\theta'_{1+2}}$$

where : θ_{1+2} = $(\theta_1 + \theta_2)$-($\theta_1\theta_2$); θ_1 = surface coverage by inhibitor1; θ_2 = surface coverage by inhibitor2; θ'_{1+2}= measured surface coverage by both inhibitor1 and inhibitor2. S_θ approaches unity when there are no interactions between the inhibitor compounds, while $S_\theta > 1$ points to a synergistic effect; in the case of $S_\theta < 1$, the antagonistic interaction prevails.

The use of theoretical methods such as: Molecular Dynamic (MD) simulation to calculate the adsorption energy and to find the most stable adsorption configuration of amino acid derivatives on metal surface has reported [44-49]. From the results obtained by MD simulation method indicated that the protonated amino acid in amino group was oriented away from the metal surface into the solution (Figure 4), which can explain its lower adsorption energy comparing with neutral form. That may be explaining the predominance of physical adsorption nature of these compounds onto the surface of materials.

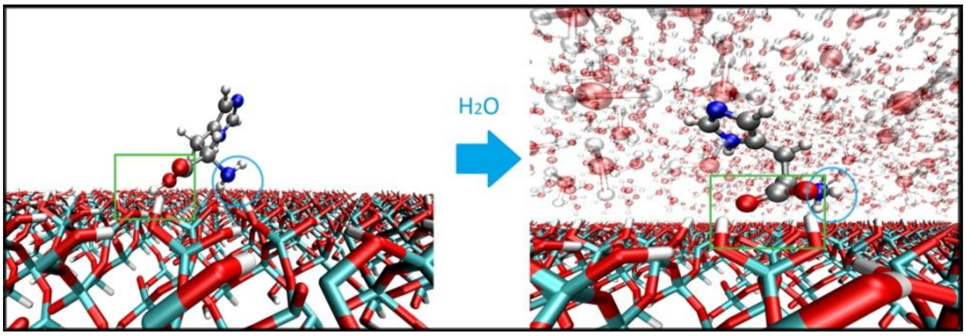


Figure 4: Orientation of amino group in the presence and absence of water

Mendonça et al. investigated recently six amino acids (Met; Cys; Ser; Arg; Gln and Asn) as inhibitor of the corrosion of carbon steel and copper in 0.5 mol L^{-1} H_2SO_4 using electrochemical techniques. Results allowed to ranking the corrosion inhibitors according their corrosion inhibition efficiency: For copper, the descending order was:

Arg > Gln > Asn > Met > Cys > Ser,

while for carbon steel the descending order was:

Met > Cys > Ser >Arg > Gln >Asn.

They concluded that DFT calculation allowed correlation of the energies of the frontiers orbitals with the experimental corrosion inhibition ranking only inside of every set, indicating that this quantum model is suitable to rank corrosion inhibitors only when they have similar molecular skeletons. On the other hand, the classical Monte Carlo method was successful to rank the studied corrosion inhibitors, suggesting that such method can be applied as a computational protocol to forecast the corrosion inhibitor rating. However, more prospective studies are necessary to validate a computational protocol using Monte Carlo method [50].

Conclusion

In this review, we reported the ability of some amino acids and their derivatives as nontoxic inhibitors to protect several materials against corrosion in different acidic environments. Despite that, this ability has been poorly investigated, especially in the case of the smart coating. In this paper, we encourage researchers to develop new amino acids derivatives and evaluate their inhibition behaviour against corrosion, which we believed that will be promising.

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