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Review

Pyrimidine derivatives as environmentally-friendly corrosion inhibitors: A review

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The perpetual quest for more efficient and environment friendly corrosion inhibitors remain a focal point in corrosion control. The use of organic compounds to inhibit corrosion has assumed great significance due to their vast applications in counteracting wastage of ferrous alloys. These compounds have shown great effectiveness for inhibiting aqueous corrosion due to film formation by adsorption on the metal surface. This paper reviews the inhibitive effect of pyrimidine derivatives as corrosion inhibitor. This class of heterocyclic aromatic organic compound has very important theoretical and practical applications. Their effect on the corrosion of metallic alloys was evaluated through assessment of various techniques and articles on their use in various investigations. The corrosion rate was found to be a function of different variables. Due attention was paid to the systematic study of inhibitor action of derivatives with much emphasis on the inhibitor type and functional groups of the molecular structure. The comprehensive discourse presented concludes, that the pyrimidine derivatives fulfill the basic requirements for consideration as an efficient corrosion inhibitor.

Key words: Pyrimidine, acid, inhibitor, steel, corrosion, organic compounds.

INTRODUCTION

The use of inhibitors is one of the most practical methods for protection against corrosion in acidic environments (Maqsood et al., 2011). A great number of studies have being devoted to the subject of corrosion inhibitors but most of what is known is as a result of trial and error, both in the laboratory and in the fields. Corrosion inhibition is of great practical importance, being extensively employed in curtailing wastage of engineering materials and minimizing corrosion control costs. Inhibitor applications is quite varied often playing an important role in oil extraction and processing industries, heavy industrial manufacturing, water treatment facility, water-containing hydraulic fluids, water treatmentchemicals, engine coolants, ferrous metal cleaners, automatic transmission fluids, automotive component manufacture, cutting fluids etc. to minimize localized corrosions and unexpected sudden failures. Historically, the development of corrosion inhibitors was determined by their effective-ness, and they were often based on ecologically problematic heavy metals (corrosion inhibitors Bodo Moeller Chemie). Acid solutions are widely used in industry, the most important fields of application being acid pickling and industrial acid cleaning due to the general aggressivity of acid solutions; thus enhancing corrosive attack on metallic materials.

The most important aspect of inhibition normally considered by corrosion scientists is the relation between molecular structure and corrosion inhibition efficiency. Such effects were studied by many authors. Lorenz et al. (1985) classified the modes of inhibition effect of interface inhibitors into three categories: (i) the geometric blocking effect of adsorbed inhibitive species on the metal surface (ii) the effect of blocking the active sites on the metal surface by adsorbed inhibitive species (iii) the electrocatalytic effect of the inhibitor or its reaction products. It has been discussed in the case of the first mode the inhibition effect comes from the reduction of the reaction area on the surface of the corroding metal, whereas for

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the other two modes the inhibition effects are due to the changes in the average activation energy barriers of the anodic and cathodic reactions of the corrosion process (Wang et al., 2001).

Most of the well-known acid inhibitors are organic compounds containing π bonds, phosphorus, sulfur, oxygen and nitrogen as well as aromatic rings in their structure which are the major adsorption centers (Abd El-Maksoud, 2003; Lopez-Sesenes, 2011). The compounds containing both nitrogen and sulfur can provide excellent inhibition, compared with compounds containing only nitrogen or sulfur (Aljourani et al., 2010; Abboud et al., 2007). Generally, a strong interaction causes higher inhibition efficiency, the inhibition increases in the sequence O < N < S (Awad et al., 2005; Chetouani et al 2003). Heteroatoms such as nitrogen, oxygen and sulphur are capable of forming coordinate covalent bond with metal owing to their free electron pairs and thus acting as inhibitor. Compounds with π-bonds also generally exhibit good inhibitive properties due to interaction of π orbital with metal surface.

Different classes from organic compounds are used as corrosion inhibitors for metallic alloys in various acid media (Growcock et al., 1988; Hettiarachchl et al., 1989; Gad Alla et al., 1990; Agrawal et al., 1990; Moretti et al., 1996; Agrawal et al., 1997; Abdel-Aal et al., 2001; Mthar et al., 2002; Selvi et al., 2003; Bentiss et al., 2004; Noor et al., 2005; Yurt et al., 2005; Li et al., 2006). Most of these organic inhibitors are nitrogen, sulfur or oxygen containing compounds. The first stage in the action mechanism of these organic inhibitors in aggressive acid media is their adsorption on the metal surface (Rozenfeld, 1981; Rudresh and Mayanna I., 1977). The effectiveness of the functional atom in the adsorption processes varies because of the very different situation created by changing various factors such as medium and inhibitor in the system metal/aggressive medium/inhibitor, various inhibition mechanisms must be considered (Trabanelli, 1986; Rozenfeld et al., 1981; Thomas, 1981).

Unfortunately, many common corrosion inhibitors are health hazards for the inhibition of corrosion in aqueous media (Flick, 1987; Uhlig and Revie, 1985). Development of effective and environmentally acceptable corrosion inhibitors as alternatives of chromates which are toxic and carcinogenic (Sax and Lewis, 1989; Katz, 1993) are been researched into for wide scale applications. Choice of inhibitor is based on two considerations. First it can be synthesized conveniently from relatively cheap raw materials. Secondly the presence of an electron cloud on the aromatic ring, the electronegative nitrogen, oxygen atoms and the relatively long chain compound in aqueous acid medium may likely induce greater adsorption of the compound on the steel surface promoting effective inhibition (Ajaal et al., 1994).

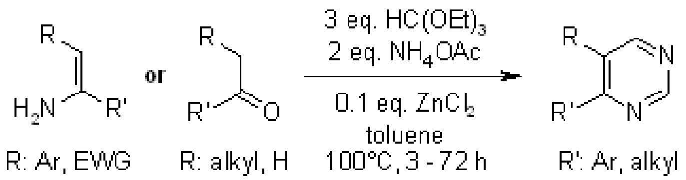
Owing to the increasing ecological awareness as well as the strict environmental regulations, and consequently the need to develop environmentally friendly processes, attention is currently focused on the development of “green” alternatives to mitigating corrosion. Green approaches to corrosion mitigation entail the use of substances, techniques and methodologies that reduce or eliminate the use of/generation of feedstocks, products, by-products, solvents, reagents, and so forth that are hazardous to human health or the environment in combating corrosion. They include the use of plant extracts, ionic liquids, biochemicals, and biodegradable organic and green inorganic inhibitors, the development of corrosion inhibitors based on waste products, corrosion protection by corrosion by-products, ion-exchange pigments, and coatings, the development of mitigation techniques that enable the detection and prevention of the early stages of corrosion, and so forth (Special Issue on Green approaches to corrosion mitigation, 2011).

Several reports have documented the use of many heterocyclic compounds, such as thiophene derivatives, mercaptobenzoazoles, crown ethers, Schiff base, pyridine derivatives, thio-compound, rhodanine azo-sulpha drugs, and mercaptopyrimidines (Nega et al., 1990; Al-Mayouf et al., 2001; Fouda et al., 2010; Bilgic et al., 2001; Abd El-Maksoud et al., 2005; Elayyachy et al., 2006; Abdallah, 2002; Reznik, 2008), however, studies on pyrimidine are limited in spite of their ease of availability and corrosion inhibition properties. They find diverse applications in pharmaceutical applications such as analgesic, antipyretic, antihypertensive, anti-inflammatory drugs, pesticides, herbicides, plant growth regulators, and organic calcium channel modulators (Hassan, 2000; Smith and Kan, 1964; Cannito et al., 1964; Nega et al., 1990; Chakaravorty et al., 1992; Shishoo and Jain, 1992). Pyrimidine derivatives are promising with respect to corrosion inhibition (Reznik et al., 2008; Abdallah et al., 2006).

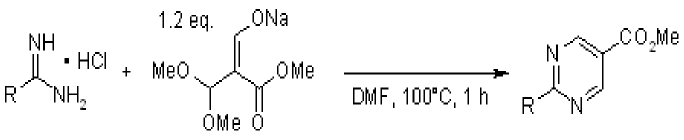
Pyrimidine is a heterocyclic aromatic organic compound similar to benzene and pyridine, containing two nitrogen atoms at positions 1 and 3 of the six-member ring (Gilchrist, 1997). It is isomeric with two other forms of diazine. It is single-ringed, crystalline organic base, $C_4H_4N_2$ that forms uracil, cytosine, or thymine and is the parent compound of many drugs, including the barbiturates.

SYNTHESIS OF PYRIMIDINE DERIVATIVES

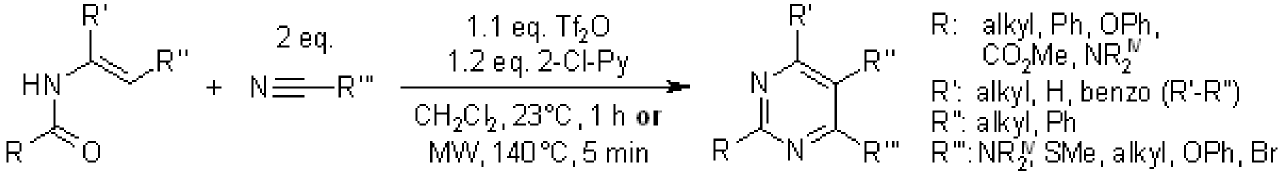
A $ZnCl_2$-catalyzed three-component coupling reaction allows the synthesis of various 4, 5-disubstituted pyrimidine derivatives in a single step from functionalized enamines, triethyl orthoformate, and ammonium acetate. The procedure can be successfully applied to the efficient synthesis of mono- and disubstituted pyrimidine derivatives, using methyl ketone derivatives instead of enamines (Sasada et al., 2009).



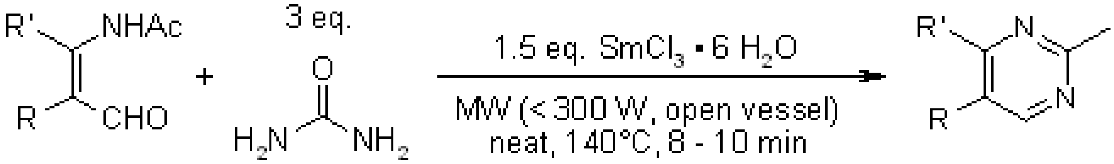
A method for the synthesis of 2-substituted pyrimidine-5-carboxylic esters is described. The sodium salt of 3, 3-dimethoxy-2-methoxycarbonylpropen-1-ol has been found to react with a variety of amidinium salts to yield the corresponding 2-substituted pyrimidine-5-carboxylic esters (Zhichkin et al., 2002).



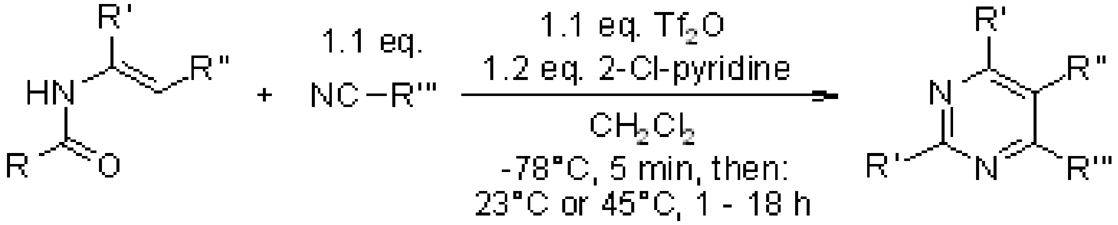
The direct condensation of cyanic acid derivatives with N-vinyl/aryl amides affords the corresponding C4-heteroatom substituted pyrimidines. The use of cyanic bromide and thiocyanatomethane in this chemistry provides versatile azaheterocycles poised for further derivatization (Ahmad et al., 2009).



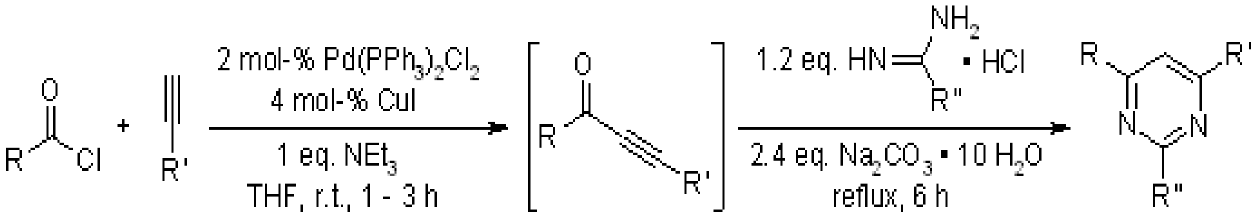
A novel and efficient synthesis of pyrimidine from β-formyl enamide involves samarium chloride catalysed cyclisation of β-formyl enamides using urea as source of ammonia under microwave irradiation (Barthakur et al., 2007).



The coupling of acid chlorides with terminal alkynes using one equivalent of triethylamine under Sonogashira conditions followed by subsequent addition of amines or amidinium salts to the intermediate alkynones allows a straightforward access to enaminones and pyrimidines under mild conditions and in excellent yields (Karpov and Müller, 2003).



A single-step conversion of various *N*-vinyl and *N*-aryl amides to the corresponding pyrimidine and quinazoline derivatives involves amide activation with 2-chloropyridine and trifluoromethanesulfonic anhydride followed by nitrile addition into the reactive intermediate and cycloisomerization (Movassaghi et al., 2006).



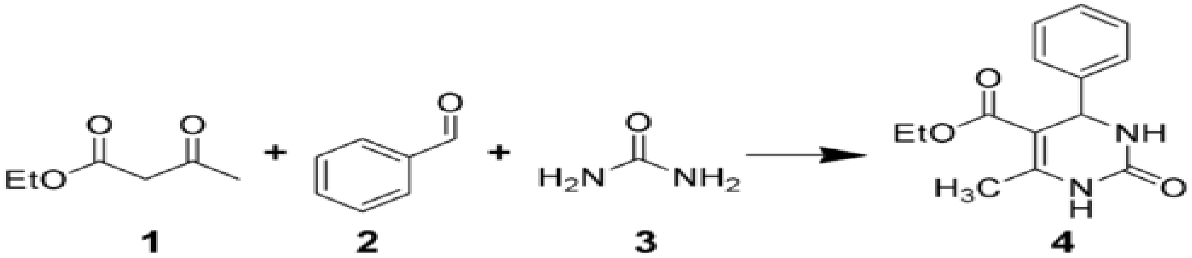
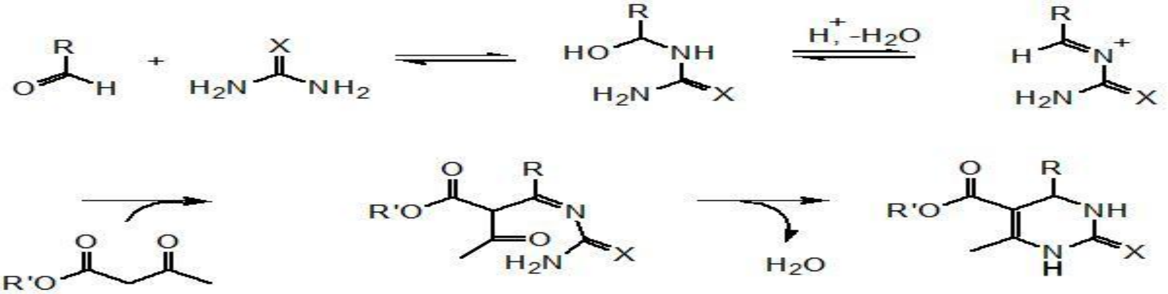


Figure 1. Biginelli reaction (Biginelli, 1891).

Biginelli reaction

The Biginelli reaction is a multiple-component chemical reaction that creates 3, 4-dihydropyrimidin-2(1*H*)-ones 4 from ethyl acetoacetate 1, an aryl aldehyde (such as benzaldehyde 2), and urea 3. (Biginelli, 1891, 1893; Zaugg and Martin, 1965; Kappe, 1993) (Figure 1).

The reaction is catalyzed by Brønsted acids and/or by Lewis acids such as boron trifluoride (Hu et al., 1998). Dihydropyrimidinones, the products of the Biginelli reaction, are widely used in the pharmaceutical industry as calcium channel blockers, antihypertensive agents and alpha-1-a-antagonists. The biginelli reaction was increased two steps further by kappe (1997) which results in higher yield output. This scheme begins with rate determining nucleophilic addition by the urea to the aldehyde (Rovnyak et al., 1992; Folkers et al., 1932; Kappe, 1997). The ensuing condensation step is catalyzed by the addition of acid, resulting in the imine nitrogen. The β-ketoester then adds to the imine bond and consequently the ring is closed by the nucleophilic attack by the amine onto the carbonyl group. This final step ensues a second condensation and results in the Biginelli compound.



MECHANISM OF INHIBITION

Corrosion inhibition by pyrimidine derivatives may involve either physisorption or chemisorption of theinhibitors to the metal surface and subsequent interference with either cathodic or anodic or both reactions occurring at the adsorption sites. The electrostatic attraction between the charged hydrophilic groups and the charged active centers on the metal surface leads to physisorption. Existing data show that most organic inhibitors adsorb on the metal surface and form a compact barrier film (Muralidharan et al., 1995). Moreover, many *N*-heterocyclic compounds have been proved to be effective inhibitors for the corrosion of metals and alloys in aqueous media. The influence of organic compounds containing nitrogen on the corrosion of steel in acidic solutions has been investigated by several authors (Arshadi et al., 2002; Quafi, 2002).

Pyrimidine compounds absorbs on the metal surface, blocking the active sites and thereby reducing the corrosion attack. The efficiency of these compounds as corrosion inhibitors is attributed to the number of mobile electron pair present (Arora et al., 2007), the π orbital character of free electrons (Hackerman and Hurd, 1960) and the electron density around nitrogen atom (Riggs and Every., 1962). The effectiveness of the functional atom in the adsorption processes varies.

Galvanostatic polarization technique in acidic medium reveals these compounds to behave as mixed type inhibitors and they inhibit corrosion by parallel adsorption on the surface of stainless steels due to the presence of more than one adsorption active centre in the inhibitor

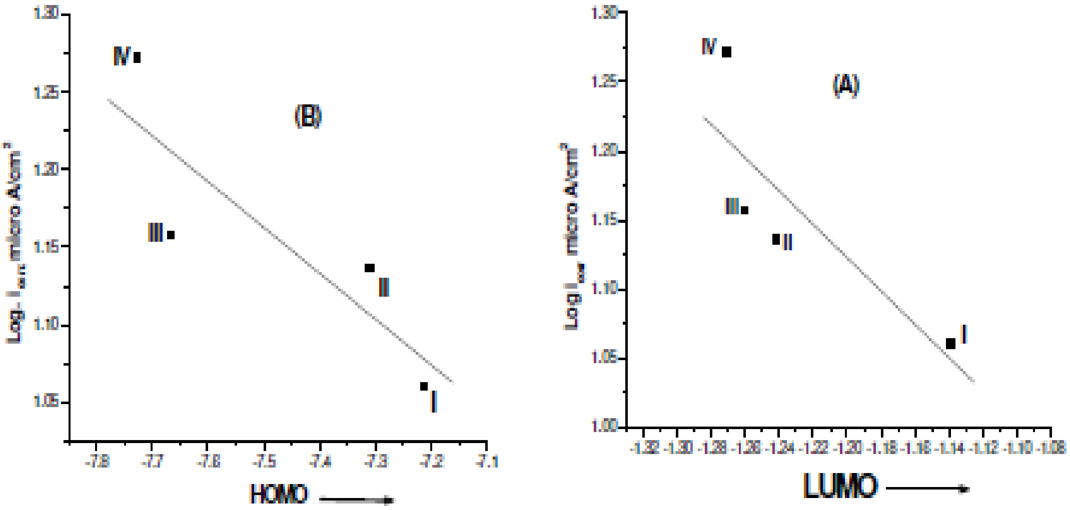


Figure 2. Correlation of LUMO (A) and HOMO (B) energies with log icorr for the investigated pyrimidine derivatives (Fouda et al., 2007).

molecule. The inhibition mechanism is a combination of surface blockageand electrostatic repulsion between adsorbed species and chloride ions which strongly depends on the inhibitor concentrations (Fouda et al., 2007).

The relationship between inhibition characteristics and quantum chemical data shows that corrosion rate mostly depend upon the energies of the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) (Fouda et al. 2007) (Figure 2). The inhibition efficiency increases with increasing the energy of the HOMO, that is, the increase of the ionization potential. It is further evident that the inhibition efficiency increases with the decrease of ionization of the molecule, which means that the molecule acts as an electron donor when blocking the corrosion reaction. Four types of adsorption may take place involving organic molecules at the metal solution interface (i) electrostatic attraction between charged molecules and the charged metal, (ii) interaction of πelectrons with the metal, (iii) interaction of uncharged electron pairs in the molecule with the metal and (iv) a combination of the above (Schweinsgberg et al., 1988). It is apparent that theadsorption of pyrimidine compounds on the Fe surface could occur directly on the basis of donor acceptor between the lone pairs of the heteroatoms and the extensively delocalized πelectrons of the pyrimidine molecule and the vacant d-orbitals of iron surface atoms. Moreover, the presence of the electron releasing methyl groups in the molecules increase the electron density on the pyrimidine heterocyclic ring.

In acidic solution, these compounds can exist as protonated species. These protonated species may adsorb on the cathodic sites of carbon steel surface and decrease the evolution of hydrogen. These compounds are able to absorb on anodic sites through N atoms, azomethine group, heterocyclic and aromatic rings which are electron donating groups. The adsorption of these compounds on anodic sitesmay also decreases anodic dissolution of ferrous alloy.

The performance of pyrimidine compounds as corrosion inhibitor mainly depends on the size and the active centres of the compound. This is enhanced due to the presence of OH and NO_2 groups in the compound. The electrophilic (electron withdrawing) character of NO_2 group is higher than the nucleophilic (electron releasing) character of OH group which hinders the delocalized πelectrons on the aromatic ring. Potentiostatic polarization results clearly revealed that pyrimidine derivatives do behave as a mixed type but slightly anodic in nature (Elewady, 2008).

The presence of heteroatoms (N) and -SH groups causes effective adsorption process leading to the formation of an insoluble protective surface film which suppress the metal dissolution reaction. The adsorption of inhibitor on the surface of mild steel also obeys Langmuir adsorption isotherm. Potentiodynamic polarization results has shown that all the inhibitors behave predominantly as anodic type in dilute sulphuric acid and the inhibitor concentration causes a positive shift in corrosion potential

Aminopyrimidine derivatives as corrosion inhibitors also depend on the number of adsorption sites in the molecule, their charge density, molecular size and stability of these derivatives in acidic solutions. The adsorption is assumed to take place mainly through the nitrogen atom of amino group (active centers). Transfer of lone pairs of electrons on the nitrogen to the surface to form coordinate type linkage is favored by the presence of vacant orbital in iron atom of low energy. Polar character of substituent in the changing part of the inhibitor molecule has prominent effect on the electron charge density of the molecule. This fact can be expressed that these compounds acts as adsorption inhibitors. An increase in the concentration of the inhibitors, more effectively inhibits the anodic and cathodic currents but greater reduction in the anodic current than the cathodic current. Anodic dissolution and hydrogen evolution reaction are also effectively retarded. Negative values of ΔG_{ads} in the acidic media ensured the spontaneity of the adsorption process (Abdallah, 2006).

PYRIMIDINE DERIVATIVES AS CORROSION INHIBITOR

As far as the inhibition process is concerned, it is generally assumed that the adsorption of inhibitors at the metal solution interface is the first step in the action mechanism of inhibitors in aggressive acid media. G.Y. Elewady investigated the effect 2, 6-Dimethylpyrimidine-2-amine and two of its derivatives on the corrosion inhibition for carbon-steel (C-steel) in 2 M HCl solution using electrochemical impedance spectroscopy (EIS) and weight loss techniques. Results obtained reveal that the used pyrimidine derivatives perform as effective corrosion inhibitors with the efficiency increasing with increase in the inhibitor concentration. Double layer capacitance (Cdl) and charge transform resistance (Rct) values derived from Nyquist and Bode plots obtained from A.C. impedance studies are indicative of the adsorption of these inhibitors on the iron surface (Abdallah., 2006).

Inhibition efficiency mainly depends on the nature of the investigated compounds. It is apparent that the adsorption of pyrimidine compounds on the surface of ferrous alloys could occur directly on the basis of donor acceptor between the lone pairs of the heteroatoms and the extensively delocalized p-electrons of the pyrimidine molecule and the vacant d-orbitals of iron surface atoms. Moreover, the presence of the electron releasing two methyl groups in the molecules increase the electron density on the pyrimidine heterocyclic ring.

In acidic solution, these compounds can exist as protonated species. These protonated species may adsorb on the cathodic sites of ferrous alloys surface and decrease the evolution of hydrogen. These compounds are able to adsorb on anodic sites through N atoms, azomethine group, heterocyclic and aromatic rings which are electron donating groups.

Caliskan and Akbas (2010) studied the inhibition of 5-Benzoyl-4-(substituted phenyl)-6-phenyl-3, 4-dihydropyrimidine-2 on the corrosion of austenitic stainless steel in 1 M HCl using Tafel plot, linear polarization and EIS at 298 K. Results showed that the compound inhibited steel corrosion. Polarization curves indicate the pyrimidine derivative act as mixed type (cathodic/anodic) inhibitors. The adsorption of the inhibitors on the stainless steel surface was found to obey the Langmuir and Dubinin–Radushkevich adsorption isotherm models. Negative values of ΔGads in the acidic media ensured the spontaneity of the adsorption process. A compound can be classified as an anodic- or a cathodic-type inhibitor when the change in the Ecorr value is larger than 85 mV (Musa et al., 2010; Li et al., 2008). If displacement in Ecorr is <85, the inhibitor can be seen as mixed type.

According to Fouda et al. (2007) the inhibitive properties of (I) [3-(4-Methoxy-phenylazo)-1, 4, 11-Ctriaza-Cyclopenta [c] Phenanthren-2-ylamine(II) 3-(4-Methyl-phenylazo)-1, 4, 11-Ctriaza-Cyclopenta [c] Phenanthren-2-ylamine (III) 3-(4-Hydrogen-phenylazo)-1, 4, 11-Ctriaza-Cyclopenta [c] Phenanthren-2-ylamine and (IV) 3-(4-Chlore-phenylazo)-1, 4, 11-Ctriaza-Cyclopenta [c] Phenanthren-2-ylamine on the cyclic stressed specimens of Stainless Steel type 304 in 0.1 M HCl at 30°C using galvanostatic polarization technique was evaluated by recording anodic and cathodic polarization curves of the electrode in the aggressive media. The examined compounds exerted an inhibiting action towards the general corrosion, ranging between 22 to 85%, and the % inhibition efficiency (%IE) for cyclic stressed specimens ranging between 3 to 34%. Polarization studies revealed that these compounds behave as mixed type inhibitors and inhibit corrosion by parallel adsorption on the surface of stainless steel due to the presence of more than one adsorption active centre in the inhibitor molecule. Abdallah et al. (2006) studies the effect of some amino-pyrimidine derivatives on the corrosion of 1018 carbon steel in 0.05 M HNO_3 solution using weight loss and polarization techniques showed the percentage inhibition efficiency to increase with increasing concentration of inhibitor and with decreasing temperature. The addition of KI to aminopyrimidine derivatives enhanced the inhibition efficiency due to synergistic effect. The inhibitors are adsorbed on the steel surface according to Temkin isotherm. It was found that the aminopyrimidine derivatives provide a good protection to steel against pitting corrosion in chloride containing solutions.

Several derivatives of 2-mercaptopyrimidine (containing -OH, -SH, $-NH_2$ or $-CH_3$ substituents) were studied by (Zucch et al., 1993) as corrosion inhibitors of ARMCO iron and low alloy steels in 1 M sulphuric acid solutions. Polarisation resistance was used to determine the trends of the corrosion rates as a function of time by measurements and analytical determinations of dissolved iron. 2-mercaptopyrimidine, 2, 4-dimercaptopyrimidine and 2-mercap-to-4-methylpyrimidine showed the most satisfactory action, reaching efficiencies of 98% on ARMCO iron at 90°C. The presence of Cr and Mo in the alloys exerted a negative influence on the efficiency of pyrimidine derivatives, by diminishing their action at 90°C. Tentative interpretation of the action mechanisms of the additives has been given on the basis of their molecular characteristics.

Investigation of the Inhibitive and adsorption properties of synthesized triazolo-pryimidine derivatives [triazolo-pyrimidine thione (P2), pyrimidinones (P1, P3)] for the corrosion of mild steel was using weight loss and electrochemical methods revealed triazolopyrimidines to act as adsorption inhibitors for the corrosion of mild steel. Inhibition efficiency increased as the concentration of the inhibitor increased but decreased with increase in temperature. The adsorption mechanism of the triazolo-pyrimidine was found to be physisorption, spontaneous and exothermic. Also, the adsorption followed Langmuir adsorption isotherm. Polarisation studies showed that the inhibitors behave as cathodic type with pyrimidine thione showing greater IE than pyrimidinones (Parameswari et al., 2011).

Increase in temperature accelerates the rate of corrosion and hence the inhibition efficiency of the additives decreases due desorption, thus the adsorption of pyrimidine derivatives on ferrous alloy's surface is most likely to occur through physical adsorption.

Inhibition by pyrimidine derivatives depend on the number of adsorption sites in the molecule and their charge density, molecular size and stability of these derivatives in acidic solutions. The inhibition effect of these compounds is attributed to the adsorption of the inhibitor molecules on the metal surface. The adsorption is assumed to take place mainly through the nitrogen atom of amino group (active centers). Transfer of lone pairs of electrons on the nitrogen to the surface to form coordinate type linkage is favored by the presence of vacant orbital in iron atom of low energy. Polar character of substituent in the changing part of the inhibitor molecule seems to have a prominent effect on the electron charge density of the molecule.

CONCLUSION

The use of inhibitors for the control of corrosion of metals and alloys applied in aggressive environment is an accepted practice. Large numbers of organic compounds are being studied to ascertain their corrosion inhibition potential. All these studies reveal that organic compounds especially those with N, S and O showed significant inhibition efficiency. But, unfortunately most of these compounds are not only expensive but also toxic. Pyrimidine compounds fulfill the basic requirements to be considered as a more environmentally-friendly formulations due to its low toxicity and acceptable inhibition power. These compounds show efficiency similar to that obtained using classical Cr(VI) based inhibitors. Pyrimidine derivatives have proved to be efficient inhibitors for corrosion of ferrous alloys in dilute acidic solutions. The derivatives act as mixed type inhibitors and the %IE was found to increase with increasing the inhibitor concentration. Molecular structure of the reviewed inhibitor is the main factor determining its characteristics. As a result of the presence of heteroatoms (S, N and O) with free electron pairs, aromatic rings with delocalized πelectrons, molecular weight, alkyl chains and substituent groups in general greatly improves inhibition efficiency. The heavy restrictions on the use and disposal of chromate containing materials has lead to an exhaustive search to find a suitable replacement ,thus its reasonably justified to base future research and development of new high-performance and environmentally-friendly anti-corrosion coatings for ferrous alloys on pyrimidine compounds.

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