



## Schiff bases and their metal complexes as corrosion inhibitors for steel alloys in acidic media

Mohamed Mustafa Ibrahim<sup>1,4</sup>, R.S.Abdel Hameed<sup>1,2\*</sup>, Abd-Alhakeem H.Abu-Nawwas<sup>1,3</sup>

<sup>1</sup>Chemistry Department, Hail College of Science, Hail University 2440, (SAUDIARABIA)

<sup>2</sup>Chemistry Department, Faculty of Science, Al – Azhar University, 11884, Cairo, (EGYPT)

<sup>3</sup>The School of Chemistry, The University of Manchester, Oxford Road, Manchester M13 9PL, (UK)

<sup>4</sup>Department of Chemistry, Faculty of Science, University Malaya, 50603 Kuala Lumpur, (MALAYSIA)

E-mail: rsabdelhameed@yahoo.com

### ABSTRACT

The importance of steel protection against corrosion in acidic solutions is increased since iron materials are the commonly exposed metals in industrial environments. This review highlights the application of Schiff bases and their metal complexes as potential corrosion inhibitors and the factors govern their use. © 2013 Trade Science Inc. - INDIA

### KEYWORDS

Corrosion;  
Inhibitors;  
Schiff bases;  
Metal complexes.

### INTRODUCTION

Corrosion is an afflicting problem associated with every use of metals. The damage by corrosion results in highly cost for maintenance and protection of materials used. It is a constant and continuous problem, often difficult to eliminate completely because Metals generally tend to move to its original state by corrosion process. Prevention would be more practical and achievable than -complete elimination<sup>[1-9]</sup>. Among the several methods of corrosion control and prevention, the use of corrosion inhibitors is very popular.

Metals are widely used as the constructional material in most of the major industries particularly in food, petroleum, power production, chemical and electrochemical industries, especially steel due to its excellent mechanical properties and low cost. The major problem of steel is its dissolution in acidic media. Corrosion of metals and steel in acidic aqueous solutions is one of the major areas of concern in many industries where in acids are widely used for applications such as acid pick-

ling, acid cleaning, acid descaling, and oil well acidizing. Because of general aggressiveness of acid solution, the materials of construction are being corroded easily.

It is very important to add corrosion inhibitors to prevent metal dissolution and minimize acid consumption<sup>[1-14]</sup>. Most well known acid inhibitors are organic compounds containing nitrogen, sulfur and oxygen atoms. The inhibiting action exercised by organic compounds on the dissolution of metallic species is normally related to interactions by adsorption between the inhibitors and the metal surface. The good inhibitor has many advantages such as high inhibition efficiency, low price, low toxicity and easy production<sup>[13,14]</sup>.

Corrosion inhibitors are compounds which control, reduce or prevent reactions between metals and corrosive media. Many efficient organic inhibitors have  $\pi$ -bonds in their structures as well as heteroatoms such as nitrogen, oxygen and sulphur. Among those are Schiff bases and their metal complexes. Lone pair of electrons on nitrogen in a compound has been reported to render a compound an efficient corrosion inhibitor for

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metals and alloys. In acidic environment, organic compounds with more than one heteroatom containing  $\pi$ -electrons exhibit high inhibiting properties by providing electrons to interact with metal surface. Other factors including the size of the molecule, the environment and nature of the metal, experimental parameters such as inhibitor concentration, molecular structure and nature of the substituents in the molecule itself should also be considered.

Schiff bases can act as Lewis bases by donating their lone pair(s) of electrons as well as any  $\pi$ -electrons to the metal. Adsorption of Schiff bases on the metal surface will then protect and cover the metal from corrosive environment and hence inhibit corrosion.

### Schiff bases as corrosion inhibitors for steels

As a representative type of these organic inhibitors, imidazoline derivatives and their quaternary ammonium salts. Inhibition of these organic compounds is attributed to the interaction of mild steel surface and inhibitor molecules via their adsorption on the metal surface. Inhibitor adsorption is influenced by:

- (a) Nature and surface charge on the metal
- (b) Type of aggressive media
- (c) Nature of interaction with metal surface
- (d) Presence of heteroatoms such as nitrogen, oxygen phosphorous and sulphur.
- (e) Multiple bonds and aromatic rings

N. A Negm and coworkers<sup>[15]</sup> investigates the corrosion inhibition properties of two quaternary isoxazolium Schiff bases against hydrochloric acid media using gravimetric, polarization and electrochemical impedance measurements. Results indicate that the corrosion of carbon steel in 0.5M HCl solutions is significantly reduced upon the addition of the cationic isoxazolium Schiff base inhibitors. The inhibition efficiency increased with increasing concentration and the alkyl chain length of the inhibitor. The potentiodynamic polarization results show that the tested inhibitors acted as a mixed-type inhibitors and effectively suppress both anodic and cathodic processes through the adsorption on carbon steel surface.

Electrochemical impedance spectroscopy showed the formation of high capacity double layer acts as capacitor at the metal surface. Data obtained from weight loss and electrochemical measurements showed that the

compounds have good inhibiting properties for carbon steel in 0.5M HCl solution.

A. Yurt<sup>[16]</sup> and coworkers studies the inhibition activities on a series of Schiff bases containing heteroaromatic substituents. The compounds act as anodic inhibitors and the variation in inhibitive efficiency mainly depends on the type and nature of the substituents present in the inhibitor molecule.

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 inhibitors. Compounds with  $\pi$ -bonds also generally exhibit good inhibitive properties due to interaction of  $\pi$ -orbital with metal surface. Schiff bases with  $RC=NR'$  as general formula have both the features combined with their structure which may then give rise particularly potential inhibitors.

In the study, polarization measurements showed that all tested Schiff bases causes a decrease in the corrosion current and shifting of corrosion potentials to noble direction in such a way that the inhibition be under anodic control.

Inhibition efficiency of the Schiff bases decrease with temperature and its decrease leads to increase activation energy of the corrosion process. Impedance measurements at  $E_{corr}$  showed a capacitive loop related to dielectric properties of the surface film.

Increasing the charge transfer resistance,  $R_p$  values of inhibitor with increase in the concentration of Schiff bases shows that inhibitive abilities of inhibitors depends on the adsorption of molecule on metal surface. The adsorption of Schiff bases on carbon steel in 0.1M HCl solution obeys Temkin's adsorption isotherm.

Four types of adsorption may take place by organic molecules at metal/solution interface. (1) Electrostatic attraction between the charged molecules and charged metal. (2) Interaction of uncharged electron pairs in the molecule with the metal. (3) Interaction of  $\pi$ -electrons with the metal. (4) Combination of (1) and (3)<sup>[17]</sup>. In acidic solution imine group as well as nitrogen atoms in heteroaromatic ring can be protonated. Physical adsorption may take place due to electrostatic interaction between protonated forms of Schiff bases and  $(FeCl^+)_{ads}$  species. Coordinate covalent bond formation between electron pairs of unprotonated S atoms in heteroaromatic ring and metal surface can take place.

Chemisorption of Schiff bases due to interaction of their  $\pi$ -orbitals with metal surface occurs following deprotonisation step of the physically adsorbed protonated forms of Schiff bases. The difference in protection action can be attributed to the presence of different substituents to azomethine ( $-\text{C}=\text{N}-$ ) group. Presence of electron releasing  $\pi$ -electron excess ring causes increase of electron density of  $-\text{C}=\text{N}-$  group which gives a better protective action of steel surface. The temperature effect over the range 20 - 60°C illustrate that the inhibition efficiency were found to decrease as temperatures were increased. A decrease in inhibition efficiencies with the increase in temperature might be due to weakening of physical adsorption. Physical adsorption in inhibition of corrosion of carbon steel in acidic solution is small but important because it is preceding stage of chemisorption of Schiff bases on carbon steel.

Aysel Yurt and co-workers in 2010 examined the effect of three Schiff bases derived from phenoxypropanes on the corrosion of mild steel in HCl<sup>[18]</sup>. Electrochemical measurement show that inhibition efficiencies increase with increase in inhibitor concentration. This implies that inhibition action of inhibitors were mainly due to adsorption on steel surface. The adsorbed molecules mechanically screen the coated part of the metal surface from the action of the corrodent. Additionally, calculated quantum chemical parameters indicate that Schiff bases adsorbed on steel surface by chemical mechanism.

S. Bilgic and N. Caliskan studied the effect of two Schiff bases N-(1-toluidine)salicylaldimine and N-(2-hydroxyphenyl)salicylaldimine on chromium-nickel steel in sulphuric acid media<sup>[19]</sup>. The difference in the adsorption behaviour of the two Schiff bases can be tentatively attributed to the structures of these molecules. Supposing these molecules are attached to the solid surface through coordination bonds formed by the electron-donating N or O atoms, N-(1-toluidine)salicylaldimine might be attached to the surface with one site leaving the other donor atom free to form intermolecular hydrogen bonds with the molecules of the second layer. However, the Schiff base N-(2-hydroxyphenyl)salicylaldimine has two closely-spaced OH groups that may well be used in forming an intramolecular hydrogen bond, in which case only the  $\text{C}=\text{N}$  group would be available to link the molecule to

the surface. In this circumstance multilayer adsorption would be unlikely and obedience to the Langmuir isotherm becomes reasonable. Chemisorption involves charge sharing or charge transfer from the inhibitor molecules to the surface to form a coordination bond. In fact, electron transfer is typical for transition metals having vacant, low-energy electron orbitals. Electron transfer can be expected with compounds having relatively loosely bound electrons<sup>[20]</sup>. The number of  $\pi$ -electrons in the system is then likely to be the determining factor in the adsorption process. Iron is well known for its coordination affinity to nitrogen and oxygen bearing ligands<sup>[21-23]</sup>. Therefore adsorption on steel may also be attributed to coordination through phenolic OH and  $\text{C}=\text{N}$  groups. Efficient adsorption of the inhibitor molecules may be either due to  $\pi$ -electrons of the aromatic system or electronegative donor atoms, N and O. The inhibition efficiencies of both Schiff bases show almost the same pattern. This is expected, taking into account the similar structures of the two molecules.

Chloride-substituted salicyaldamine Schiff bases were investigated for their inhibiting properties for steel by Kaan C and coworkers<sup>[24]</sup>. It was found that when the concentrations of the inhibitors were increased, the inhibitor efficiencies were also increased with increasing surface coverage. This suggests that the molecules may first be chemically adsorbed on the surface and cover some sites of the electrode surface, then probably form monomolecular layers, on which the insoluble product (by forming a complex) of the iron ions form and the inhibitor grows, thereby protecting these sites from attack by chloride ions. The ortho- $\rightarrow$  para- $\rightarrow$ meta-relationship in terms of inhibitor efficiency is due to the complexation and coordination effect. When an electron donor group, such as a Cl<sup>-</sup> ion, is substituted in the ortho-position, the Schiff base tends to behave like a tridentate ligand in the form ONCl, which radically increases the complex formation constant. Iron (II) in this case tends to form a complex with the tridentate ligand in octahedral coordination<sup>[25,26]</sup>. Thus the main factor affecting the inhibition characteristics of 2Cl-R is the tridentate ligand which tends to form a mononuclear tridentate Fe (II) complex. Apart from this, the ortho-position is closest to the  $\text{C}=\text{N}$  group. The presence of the electron donating group in this position increases the electron density on the nitrogen of the  $\text{C}=\text{N}$  group<sup>[27]</sup>.

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On the other hand the para-position is similar to the ortho-position through resonance but is further away from the C= N group. The ligand tends to form a didentate structure in the form ON. In this case rather than formation of a tridentate complex a didentate complex results and the main factor affecting the inhibition efficiency is the basicity of the complex alone.

The meta-position has no effect through resonance and shows the lowest inhibition although it too can form a didentate structure.

S. Chitra and coworkers work on four dianiline Schiff bases<sup>[28]</sup>. Their inhibiting action on corrosion of mild steel in 1M sulphuric acid at 30°C was investigated by various corrosion monitoring techniques. A preliminary screening of the inhibition efficiency was carried out using weight loss measurements. Potentiodynamic polarization studies showed that the Schiff bases were mixed type inhibitors. The effect of temperature on the corrosion behaviour of mild steel in 1M sulphuric acid with the addition of the Schiff bases was studied in the temperature range from 40-60°C. The adsorption of these compounds on a mild steel surface from sulphuric acid obeyed the Langmuir adsorption isotherm. The decrease in inhibition efficiency with increase in temperature and the less negative  $\Delta G^{\circ}_{ads}$  values suggest predominant physisorption of the Schiff base molecules on the steel surface. Weight loss data reveal that the loss in weight of mild steel specimens decreases with increasing inhibitor concentration. Hence for the four inhibitors the inhibition efficiency (IE) increases with the increase in concentration. In relation to temperature dependence, it was found that the weight loss increased linearly with the increase of temperature in the absence and in the presence of inhibitors.

The inhibitor efficiency was also determined for the dianiline Schiff bases using gasometric method from the volume of gas collected in the absence and presence of inhibitors at 30±1°C for the corrosion of mild steel in 1M sulphuric acid. The volume of gas collected decreased with the addition of inhibitors. The rate of cathodic hydrogen evolution reaction due to the increased level of inhibition leads to the decrease in the volume of hydrogen gas evolved suggesting that the inhibitor efficiency increases with increase in the concentration of the inhibitors similar to weight loss method.

M. N. Desai and his coworkers studied the corro-

sion inhibition of seven different Schiff bases derived principally from benzaldehyde, anisaldehyde, salicylaldehyde and cinnamaldehyde<sup>[29]</sup> in 1–6M HCl media against mild steel. The Schiff bases have been found to be very effective inhibitors for the corrosion of mild steel in HCl solutions. In general the protective action of the inhibitor improves with an increase in inhibitor concentration and with an increase in HCl concentration. All these compounds seem to function as inhibitors by being adsorbed on metal surface. They obey the Freundlich adsorption isotherm.

The activation energies of the Schiff bases studied are less than those in uninhibited acid, indicating that the efficiency of these inhibitors improves with an increase in temperature.

All the compounds reduce cathodic current needed for the protection of the metal. It is interesting to note that in many cases the efficiency of the inhibitors are identical in the absence of external cathodic current but in the presence of impressed cathodic current the adsorption characteristics of one of the substances improves, so that the protective current is reduced.

### Schiff base complexes as corrosion inhibitors

Relatively few studies on Schiff base metal complexes as corrosion inhibitors appear in the literature. A. M. Abdel Gaber and coworkers reports the effect of the Schiff base N,N'-bis (salicylaldehyde)-1,3-diaminopropane (Salpr) and its corresponding cobalt complex on the corrosion behavior of steel in 1M sulphuric acid solution<sup>[30]</sup>. Results reveal that there might be non-ideal behaviour in the adsorption processes of Co (Salpr) complex on the steel surface. The Co(Salpr) could displace more water molecules from the steel surface than the corresponding Salpr. The bulky Co(Salpr) molecule could cover more than one active site. If the adsorption process involves overlap of occupied ligand non-bonding orbital with metal empty inner d or f orbital, a coordinate type bond formed and the process is termed chemisorption<sup>[31]</sup>. This situation can arise in cases where the inhibitor molecules contain lone pairs of electrons, multiple bonds, or conjugated  $\pi$ -type bond system<sup>[32-34]</sup>.

On the other hand, some organic inhibitors could react with metal ions resulting from the corrosion process to form analogous complexes spontaneously that

may decrease or increase the corrosion rate. The inhibitive effect of Salpr and Co(Salpr) was attributed to the adsorption over the metal surface via the free adsorption centers in the molecule. The higher binding (equilibrium) constant value of Co(Salpr) complex indicated that it is more efficient inhibitor than the corresponding Schiff base. It is worthwhile to investigate the stability of Co(Salpr) complex in the test solution before studying its effect on the corrosion behaviour of mild steel. The stability of Co(Salpr) complex at 20 °C was studied using spectrophotometry technique in which the UV-visible absorption spectra was measured for the complex in absolute ethanol and in test solution (1 M sulphuric acid containing 10% (v/v) ethanol), the resultant curves were thereafter compared to that of alcoholic Salpr solution. The co-complex is of similar trend in both ethanol and sulphuric acid solutions in which a

shoulder appear at 219 nm followed by a distinct band at 251–256 nm and another broad band at 379 nm. The complex-acid solution bands have lower intensity than the complex-alcoholic solution that may be attributed to a partial hydrolysis of the complex in such media. The absorption bands of the Salpr appeared at 215, 258, and 332 nm. The difference in the absorption behaviour for the Salpr and its corresponding complex confirms the stability of Co(Salpr) complex in the acid solution. The potentiodynamic polarization curves and Nyquist plots for steel in 1.0 M sulphuric acid solution containing different concentrations of Co(Salpr) at 20 °C gave the same characteristics that have been discussed in the presence of Salpr suggesting that the Co(Salpr) could be classified as cathodic type inhibitor and the dissolution process occurs under activation control.

TABLE 1

No.	Compound	Media	Metal	Reference No.
1	2-aminoisoxazole Schiff base derivatives	HCl	Carbon steel	1
2	Thiophene and Benzothiazole derivatives	HCl	Carbon steel	2
3	Phenoxypropane Schiff bases	HCl	Mild steel	4
4	Salicylaldehyde Schiff bases	H <sub>2</sub> SO <sub>4</sub>	Chromium-nickel steel	5
5	Salicylaldamine Schiff bases	HCl	Steel	10
6	Dianiline Schiff bases	H <sub>2</sub> SO <sub>4</sub>	Mild steel	14
7	Benzaldehyde, anisaldehyde, salicylaldehyde and cinnamaldehyde Schiff bases	HCl	Mild steel	15
8	Benzylidene and Salicylidine Schiff bases	HCl	Aluminum	16
9	N,N'-bis (salicylaldehyde)-1,3-diaminopropane	H <sub>2</sub> SO <sub>4</sub>	Steel	20
10	N,N'-bis (salicylaldehyde)-1,3-diaminopropane cobalt complex	H <sub>2</sub> SO <sub>4</sub>	Steel	20
11	ACBAI and its metal complexes	HNO <sub>3</sub>	Mild steel	25
12	2-((pyridin-2-ylimino)methyl)phenol, 2-((hexadecylimino) methyl)phenol, 2-((4hydroxyphenylimino) methyl)phenol, and 1-(4-(2 hydroxy benzylidene amino) phenyl)ethanone	HCl	Carbon steel	37

Suraj B. Ade and coworkers studies the effect of the Schiff base 4-Chloro-2-(2-oxo-1, 2-dihydroindol-3-ylidene amino)-benzoic acid (ACBAI) and their Cd (II), Ti (IV), Zr (IV) and Hg (II) metal complexes on mild steel in 0.1N HNO<sub>3</sub> acid solutions using weight loss method<sup>[35]</sup>. The study reveals that the compounds have inhibition properties due to adsorption to the metal surface, thus acting as blanket preventing mild steel from coming into contact with the acidic corrosive environment. The results of percentage inhibition efficiency indicate that [Cd(II) ACBAI] complex is a good inhibitor. [Ti(IV) ACBAI], [Zr(IV) ACBAI], and [Hg(II)

ACBAI] complexes have less inhibition efficiency.

However, studies about the effect of metal complexes as corrosion inhibitor for steel in acid solution appeared in the literature are extremely limited<sup>[36,37]</sup>.

The table below summarizes the reviewed work in terms of compounds, media and the metals studied

## CONCLUSIONS

- 1 This review highlights the application of Schiff bases and their metal complexes as potential corrosion inhibitors for steel, and the factors govern their use.

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- 2 Most of Schiff's base reported here act as good corrosion inhibitors for steel in acidic media.
- 3 The corrosion inhibition efficiency depend on the corrosive medium, acid type, acid concentration, PH, type of inhibitors used, and their interaction with the metal.
- 4 The use of Schiff's base metal complexes as Corrosion inhibitors for metals are limited and are of real promise.

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