Novel metal complexes as antimicrobial and anticorrosion in acid media

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Abstract

New Cu(II),Co(II),Ni(II),Mn(II),Pt(IV),Fe(III) and Ag(I) complexes of Schiff base N-(2-hydroxyphenyl)-3-(2-hydroxyphenylimino) were prepared and characterized by elemental analyses, IR, UV–Vis spectra, molar conductance, 1H NMR and mass spectra. Electronic spectral and magnetic moment studies indicate an octahedral geometry for the Cu(II),Ni(II), Co(II) and Fe(III) complexes while square planar geometry for the Pt(IV) and Ag(I) complexes. The Schiff base and their metal complexes were tested for their antimicrobial activities. The activity show that the complexes [LCu(H$_2$O)$_2$]2H$_2$O, [L Mn$_2$.(H$_2$O)$_2$]Cl$_2$.4H$_2$O and [LCo$_2$(NO$_3$)$_2$] are more active than the Schiff base and Ampicillin towards Esherichia col and Staphylococcus aureus. The anticorrosion potency of H$_2$L Schiff base and [LCu(H$_2$O)$_2$]2H$_2$O, [(HL)Co(H$_2$O)Cl]5H$_2$O and [LNi(H$_2$O)$_2$]2H$_2$O complexes was investigated for carbon steel in 1.0 mol L^{-1} H$_2$SO$_4$ solution using electrochemical methods. Detailed electrochemical polarization and impedance studies prove that H$_2$L Schiff base is mixed-type inhibitors and improved adsorption of H$_2$L Schiff base species on carbon steel surface at increased concentrations reaching a maximum efficiency of 83.08%. Inhibition activity of newly synthesized [(HL)Co(H$_2$O)Cl]5H$_2$O complex on carbon steel gets enhanced significantly due to the presence of chloride ion inside the coordination sphere , which possesses many lone pair of electrons which enhances the bond with the steel.

Keywords: Schiff base; transition metal complexes; corrosion inhibition; antimicrobial, polarization; electrochemical impedance; electrical conductivity.

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1. Introduction

Over the years and among various real-life challenges in the world of chemistry, the spontaneous damage of metal is a routine problem faced by aggressive chemicals and/or of various micro-organisms. Carbon steel is extensively applied in various industrial processes such as marine applications, nuclear and fossil fuel power plants, metal-processing equipment, transportation, chemical processing, pipelines, mining and construction [1,2]. Generally, in these applications the acid solutions are used in order to remove unwanted scale and corrosion products from metals. However, these acids attack the metal and launch corrosion which cause serious damage to the metal and its properties, thereby limiting its applications [3-5]. Thus, the chemistry of corrosion and metal protection is of great concern to chemists and materials scientists. Schiff bases are more important in the field of corrosion inhibition of metals. The presence of azomethine >C=N–groups of Schiff bases makes it adsorbed on the metals surface and formed a monolayer on the surface spontaneously [6,7]. Therefore, it used as effective corrosion inhibitor for mild steel [8-10] carbon steel [11,12] stainless steel [13,14], copper [15], aluminum [11,16], and magnesium alloy [17,18] in various aggressive solutions. The metal complexes are more stable than the Schiff bases and contains also azomethine group generate another class of more active compounds for corrosion inhibition [19-21]. The interaction between inhibitor molecules and the metal surface is affected by molecular size, electron charge density, geometry and number of hetero atoms (N, O, S) present in the molecule [10,22]. Additionally, these kinds of Schiff base have significant interest due to their applicability as model compounds in both analytical and bioinorganic studies [23-25]. The metal complexes of Schiff bas have appreciably improved antibacterial [26-28] and antifungal [29-31] activity against microbial strains than the free ligand.

 In our previous studies of Schiff bases as corrosion inhibition, the results indicated the activity of these category of compounds and the complexes are more potent antimicrobials than the parent ligand [32]. Therefore in the present work we synthesized and characterized Cu(II), Co(II), Ni(II), Mn(II), Pt(IV), Fe(III) and Ag(I) complexes of Schiff base N-(2-hydroxyphenyl)-3-(2-hydroxyphenylimino) butanamide. The antimicrobial activities of these compounds was studied as well as screening for the inhibitive action of H2L Schiff base beside Cu(II), Co(II) and Ni(III) complexes. Corresponding inhibition mechanisms are also discussed.

2. Experimental Details

2.1 Chemicals

All commercial reagents, chemicals and solvents were analytical grade (BDH or Aldrich) and used without further purification.

2.2 Synthesis of The Ligands

The Schiff base N-(2-hydroxyphenyl) -3-(2-hydroxyphenylimino) butanamide, Table1 was prepared by adding hot ethanolic solutions of ethylacetoacetate (0.01 mol L^{-1}) and 2-aminophenol (0.02 mol L^{-1}) in presence of (0.01 mol L^{-1}) of sodium acetate. The mixer then refluxed for 3 hours. Ice bath was used for concentrate and cool the resulting solution. The separated precipitate was collected through filtration using a vacuum pump and washed with ethanol, dried in dedicator.

2.3 Synthesis of The Complexes

The hot ethanolic solutions of corresponding metal salt (0.05 M) and Schiff base (0.05 mol L^{-1}) were mixed and refluxed for about 3-8 hours to get the metal complexes, Table1. The resulting solution was cooled for overnight and filtered. The obtained metal complexes dried in a desiccator.
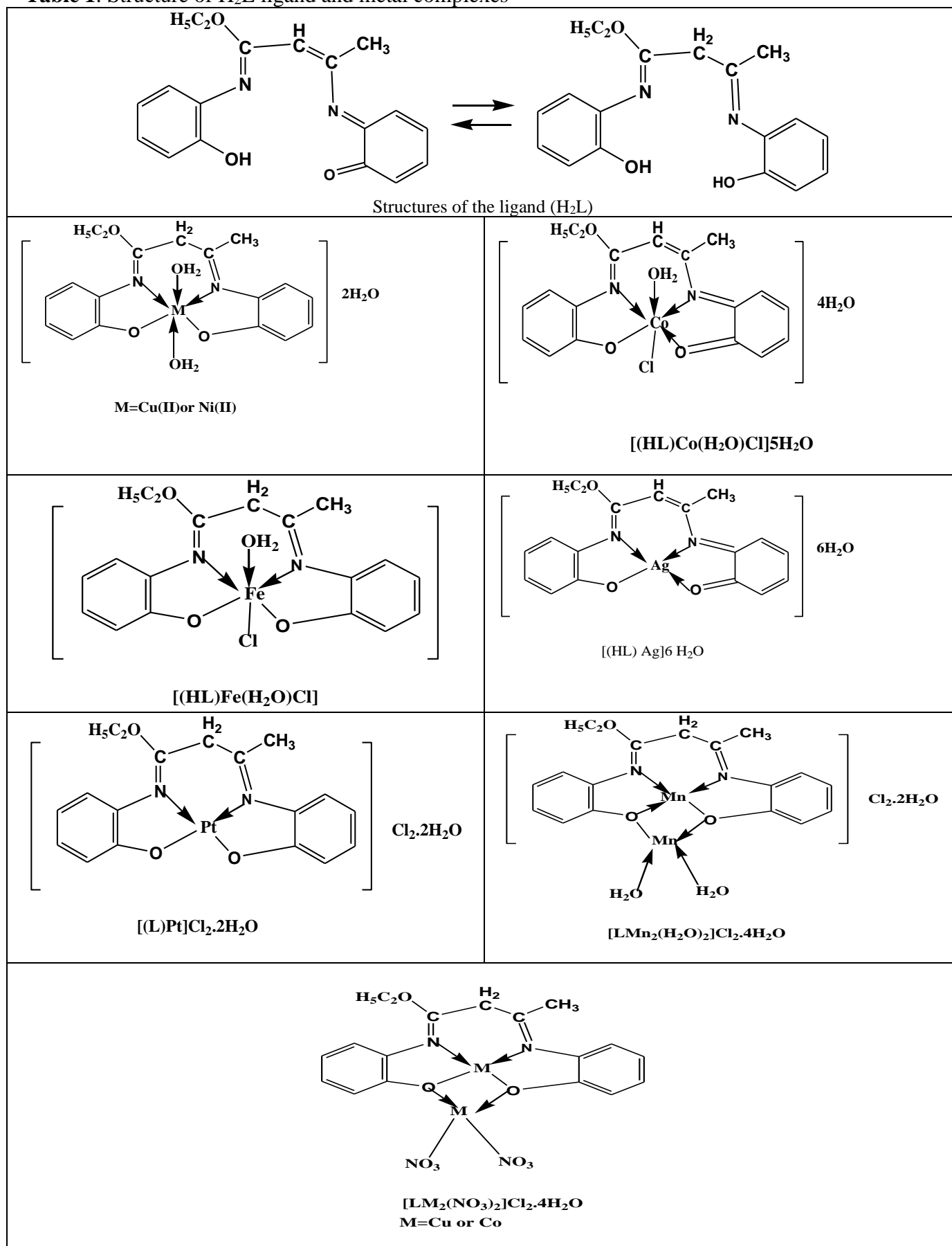
2.4 Measurements

C, H, N and Cl were determined using atomic absorption with a Perkin Elmer (Elemental analyses, EA model 2380) spectrophotometer. The fusions gropes were measured using the IR spectra (infrared spectrometer, IR, Perkin-Elmer 1430) as KBr discs in range 4000-200 cm^{-1}. Electronic absorption spectra in the 200-900 nm region were recorded on a Perkin-Elmer 550 spectro-photometer. The Gouy method were used to measure magnetic susceptibilities at room temperature. A Bibby conductometer MCl was used for conductance measurements.

2.5 Material and Solution for Corrosion Tests

The carbon steel sample is used in this work was machined as a disk of area about 0.785 cm^{2}. The composition (wt%) of carbon steel was: C 3.45%, Si 2.40%, Mn 0.70 %, P 0.17 – 0.26 %, S 0.06 % and Fe balance. Prior to all electrochemical measurements, the samples were abraded with 220-1500 grit SiC paper and then cleaned with deionized water. For corrosion measurements, 1.0 mol L^{-1} H_2SO_4 solution was used to test the uninhibited and inhibited solutions containing test compounds.

2.6 Electrochemical Measurements

The study of corrosion inhibition of the N-(2-hydroxyphenyl)-3-(2-hydroxy-phenylimino) butanamid (H_2L) and their metal Cu(II), Co(II), Ni(II) complexes on carbon steel in aggressive acid medium of 1.0 mol L^{-1} H_2SO_4 in stagnant condition was carried out using potentiodynamic polarization (PDP) and electrochemical impedance spectroscopy (EIS). Three electrode system comprising carbon steel, platinumrod and Ag/AgCl with 3.0 mol L^{-1} KCl were used as working, counter and reference electrodes respectively on Interface 1000ᴛᴍ, Gamry, supported by Potentiostat/Galvanostat/ZRA analyzer. Each experiment was repeated three times to confirm the reliability of the results.

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Table 1: Structure of H₂L ligand and metal complexes

Structures of the ligand (H₂L)

M=Cu(II)or Ni(II)

2H₂O

[(HL)Co(H₂O)Cl]5H₂O

4H₂O

[(HL)Fe(H₂O)Cl]

[(HL) Ag]6 H₂O

6H₂O

[(L)Pt]Cl₂.2H₂O

Cl₂.2H₂O

[LMn₂(H₂O)₂]Cl₂.4H₂O

Cl₂.2H₂O

[LM₂(NO₃)₂]Cl₂.4H₂O
M=Cu or Co

2.7 Antimicrobial Activity

A modified Kirby- Bauer disc diffusion method was used for testing of the Schiff base and their metal complexes against vitro antibacterial activity against Escherichia coli, Staphylococcus aureus and antifungal activity against A spergillus flavus and Candida albicans. A mpicillin (Antibacterial agent) and amphotericin B (Antifungal agent) served as positive controls for antimicrobial activity. For negative control, filter discs impregnated with 10 µL of solvent DMSO (dimethylsulphoxid) was used. For the disc diffusion the zone diameters were measured with slipping calipers of the National Committee for Clinical Laboratory Standards.

3.   Results and Discussion

The ligand may find in more than tautomeric equilibria as shown in Fig.1. The metal complexes are stable, partially soluble in methanol or ethanol and more soluble in dimethylformamide (DMF) and DMSO. The analytical data and spectral studies for the Schiff base and metal complexes are compatible with their proposed molecular formula and summarized in Table 2.
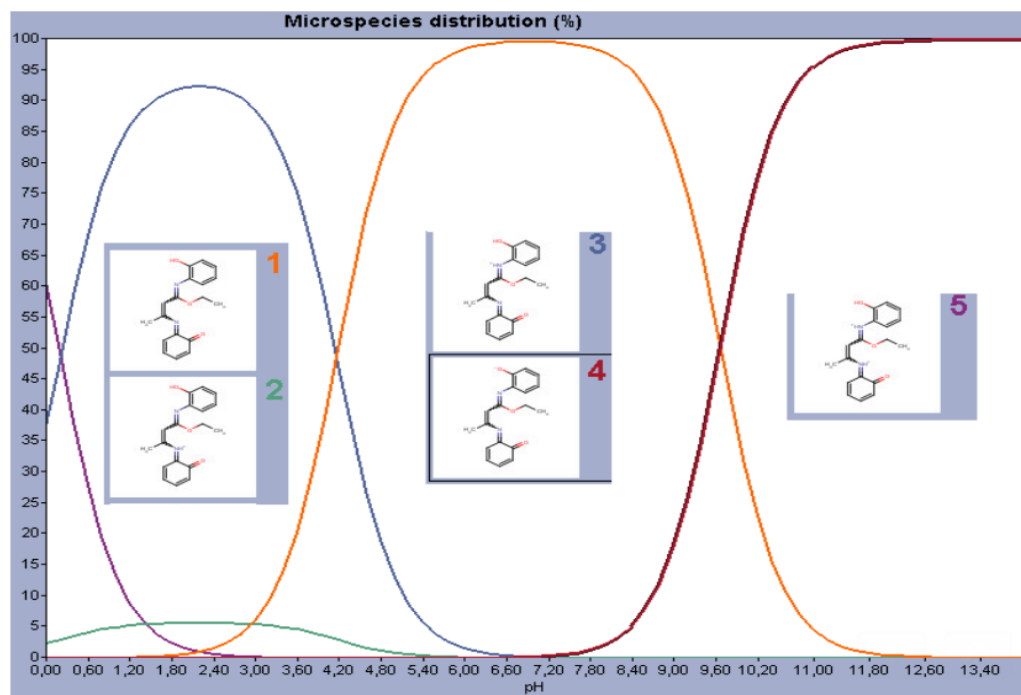
Fig 1. Tautomeric equilibria of the ligand H_2L.

3.1. ^{1}HNMR Spectra

The NMR spectra of the Schiff base **H_2L** show different signals at 9.0 ppm (NH), 4.5 ppm (OH) and 6.4-7.0 ppm (aromatic protons). The triplet and quartet peaks at 2.2 and 3.4 ppm due to CH_3 and CH_2 of ethyl group respectively and the singlet at 3.5 and 2.5 ppm corresponds to CH_2 and CH_3 protons [33].

Table 2: Analytical and physical data of Schiff base H₂L and their metal complexes.

No.	Compound	Colour	M.wt	M.PᵒC	Yield%	Anal.Found (calc.)					Λ $\Omega^{-1}cm^2mol^{-1}$
						C%	H%	N%	M%	Cl	
1	H₂L (C₁₈H₂₀N₂O₃)	Pale brown	312	190	80	68.8 (69.2)	5.9 (6.4)	8.9 (9.0)	-	-	15.0
2	[LCu(H₂O)₂]2H₂O	Deep brown	445.5	300>	90	48.0 (48.5)	6.0 (6.3)	7.1 (6.9)	14.5 (14.3)	-	12.5
3	[(HL)Co(H₂O)Cl] 5H₂O	Deep green	513.4	300>	85	42.4 (42.3)	6.0 (6.0)	6.0 (5.5)	11.0 (11.5)	7.0 (6.9)	18.6
4	[LNi(H₂O)₂]2H₂O	Brown	440.7	300>	80	48.6 (49.0)	4.8 (5.2)	6.0 (6.4)	13.5 (13.3)	-	14.5
5	[L Pt]Cl₂.2 H₂O	Deep gray	614	250	80	36.0 (35.2)	4.2 (4.6)	5.0 (4.6)	31.4 (31.8)	11.8 (11.6)	180.5
6	[(HL)Fe.Cl.H₂O]	Brown	420.5	300>	75	50.7 (51.5)	5.4 (5.0)	7.0 (6.7)	13.7 (13.9)	8.8 (8.4)	12.0
7	[(HL)Ag₂NO₃.H₂O]	Deep brown	608	280	70	36.1 (35.5)	3.9 (3.4)	6.8 (6.9)	35.3 (345.5)	-	30.5
8	[L Mn2.(H₂O)2]Cl₂.4H₂O	Brown	601	300>	85	35.2 (35.9)	3.9 (4.3)	4.9 (4.7)	18.2 (18.3)	11.0 (11.8)	170
9	[LCu₂(NO₃)₂]	Deep green	563	300>	90	38.5 (38.3)	3.5 (3.2)	10.0 (9.9)	22.0 (22.6)	-	25
10	[LCo₂(NO₃)₂]	Brown	553.8	260	80	39.2 (39.0)	2.6 (3.2)	10.4 (10.1)	21.4 (21.3)	-	35.0

3.2 Infrared Spectral (IR)

The characteristic IR absorption bands of the Schiff base **H₂L** and its complexes are summarized in Table 3. The spectra of complexes differ from that of the Schiff base **H₂L**. A strong band appears at 1588cm⁻¹ in the Schiff base **H₂L** due to νC=N vibrations, shifted by about 20-15 cm⁻¹ in all metal complexes indicated that the coordination of nitrogen to metal ion [34]. The band at 1727 cm⁻¹ of C=O in the Schiff base **H₂L** indicated that the ligand has keto-enol form, this band disappearance in the complexes indicated that the coordination occurs through enolic oxygen also the presence of the -OH group in the Schiff base **H₂L** at 3481 cm⁻¹ absent in complexes indicated that the coordination occurs through oxygen of hydroxide and the Schiff base **H₂L** participated as dibasic in the coordination sphere [35]. The bonding of the Schiff base **H₂L** to the central metal ion is provided by the appearance of bands at ~600 cm⁻¹ and ~ 530 cm⁻¹, which assigned to M-O and M-N bands respectively [36]. The presence of water molecules in the complexes is confirmed by band between 3250-3415 cm⁻¹ and is followed by a sharp rocking mode of vibration between 840-850 cm⁻¹ [36].

3.3 Mass Spectra

The mass spectra of the ligands gave the peaks at m/z 312 these values consistent and proofed the molecular weight of the Schiff base **H₂L** calculated. The parent ion and the fragments obtained by cleavage in different positon are 312, 299, 285, 271, 256, 236, 212, 185, 171, 152, 129, 123, 97, 71,57

and 55. The ions support the proposed composition and structure.

Table 3: IR Spectra of the Schiff base H_2L and its complexes.

No.	Molecular Formula	OHν	C=Oν	νC=N	νM-O	M-Nν	M-Clν
1	H_2L ($C_{18}H_{20}N_2O_3$)	3481(s) 3370(s)	1727(s)	1588(s)	-	-	-
2	$[LCu(H_2O)_2]2H_2O$	3380(br)	-	1600(s)	592(m)	550(w)	-
3	$[(HL)Co(H_2O)Cl]5H_2O$	3366(br)	-	1586(s))	678(m)	570(m)	430(w)
4	$[LNi(H_2O)_2]2H_2O$	3370(br)	-	1602(s)	583(m)	546(m)	-
5	$[L Pt]Cl_2.2 H_2O$	3369(br)	-	1601(s)	598(m)	500(m)	-
6	$[(HL)Fe.Cl.H_2O]$	3429(br)	-	1626(s)	589(m)	551(m)	445(w)
7	$[(HL)Ag_2NO_3.H_2O]$	3452(br)	-	1578(m)	587(m)	518(m)	-
8	$[L Mn2.(H_2O)2]Cl_2.4H_2O$	3402(br)	-	1591(s)	595(m)	481(w)	-
9	$[LCu_2(NO_3)_2]$	3428(br)	-	1513(s)	586(m)	470(m)	-
10	$[[LCo_2(NO_3)_2]$	3421(s)	-	1600(s)	543(s)	456(m)	-

3.4 Electronic Spectra and Magnetic Moment

The electronic spectra of the Schiff base **H₂L** and metal complexes were carried out in DMF and corresponding date are shown in Table 4. Three distinct bands were observed at 291 - 226 nm; 346 - 300 nm and 433,414 nm. due to π-π* and n-π* of (C=N) transitions, respectively [37,38]. The absorption band at above 400 nm has been previously assigned to the keto-imine form of ortho – hydroxyl of Schiff base H_2L in polar solvents [38]. Another new peaks appeared at 502 nm in metal complexes assigned to ligand-to-metal charge transfer (LMCT) transition [38].

$[LCu(H_2O)_2]2H_2O$ complex has magnetic moment 1.73 BM indicated octahedral arrangement around copper (II) ion [39]. $[(HL)Co(H_2O)Cl]5H_2O$ complex showed magnetic values 4.2 BM, which was in good agreement with those observed for most high-spin octahedral cobalt(II) complexes [39]. The value for $[LNi(H_2O)_2]2H_2O$ complexes was 3.2 BM range, which indicated octahedral geometry around the nickel(II) ion [40]. The magnetic moment value for manganese(II) complex are 5.6 B.M. suggest tetrahedral geometry around the manganese(II) ion. The value of μeff $[(HL)Fe.Cl.H_2O]$ 5.6 BM, are compatible with high-spin octahedral iron(III) complexes [41]. The complexes $[LPt]Cl_2.2 H_2O$ and $[(HL)Ag_2NO_3.H_2O]$ showed diamagnetic property. $[LCu_2(NO_3)_2]$ and $[LCo_2(NO_3)_2]$ complexes have magnetic moment values 2.2 BM. And 4.6 BM respectively. This could be taken as an evidence for the

presence of square planar complexes [42].

3.5 Molar Conductivity

Molar conductivities of the prepared complexes were measured in 10^{-3} mol /L DMF solutions at room temperature, Table 4. The complexes of the Schiff base **H₂L** showed a lower molar conductivity values in the range 12-35 $\Omega^{-1}cm^2$ mol^{-1} indicated that the complexes are nonelectrolyte in nature [43], except [LPt]Cl₂.2H₂O and [LMn₂.(H₂O)₂]Cl₂.4H₂O complexes have values 180.5 and 170 $\Omega^{-1}cm^2mol^{-1}$ respectively, which indicates the electrolytic nature. This confirm that the chloride ion present in outer coordination sphere [44].

Table 4: UV Spectral data of the Schiff base H_2L and metals complexes.

No.	Compounds	keto-imine form	n-π*	π-π*	C-T
1	H₂L (C₁₈H₂₀N₂O₃)	430,440	346,328,317,309	299,290,280	-
2	[LCu(H₂O)₂]2H₂O	450	340	290,284	540
3	[(HL)Co(H₂O)Cl]5 H₂O	460	330	294,249	530
4	[LNi(H₂O)₂]2H₂O	445	320,350	289,279	510
5	[L Pt]Cl₂.2 H₂O	464,425,414,404	397,390,357,326,302	292,278	-
6	[(HL)Fe.Cl.H₂O]	460	300	290	540
7	[(HL)Ag₂NO₃.H₂O]	420	310	290	-
8	[L Mn₂(H₂O)₂] Cl₂.4H₂O	440	321,310,300	280	520
9	[LCu₂(NO₃)₂]	450	300	279	540
10	[LCo₂(NO₃)₂]	455	310	280	530

3.6 Antimicrobial Activity

The antibacterial and antifungal activity of the Schiff base **H₂L** and their metals complexes were tested against gram-positive *(Staphylococcus aureus)*, gram-negative *(Esherichia coli)* bacteria and fungi *(Aspergillus flavus, Candida albicans)*, using a modified Kirby- Bauer disc diffusion method. The biological activity data were listed   in Table 5.

Table 5: Antimicrobial activity of the Schiff base H_2L and metals complexes.

Organisms	Compound / Inhibition zone diameter(mm)							
	Ampicillin	Amphotericin B	H_2L	$[LCu(H_2O)_2]2H_2O$	$[(HL)Fe.Cl.H_2O]$	$[L Mn_2(H_2O)_2]Cl_2.4H_2O$	$[LCu_2(NO_3)_2]$	$[LCo_2(NO_3)_2]$
Esherichia coli	22	-	18	22	10	22	16	22
Staphylococcus aureus	18	-	18	20	10	20	16	20
Aspergillus flavus	-	17	9	9	0	0	0	0
Candida albicans	-	18	0	0.0	0	0	0	0

According to inhibition zone, the prepared complexes have biological activity higher than the Schiff base **H₂L** toward both bacteria and fungi. The great performance of metals complexes is due to the effect of metal ions chelation on the normal cell process. The antibacterial activity shows that the $[LCu(H_2O)_2]2H_2O$ [45], $[LMn_2(H_2O)_2]Cl_2.4H_2O$ and $[LCo_2(NO_3)_2]$ complexes are more active than the Schiff base and Ampicillin towards *Esherichia coli* and *Staphylococcus aureus* and results are in good agreement with previous findings with respect to comparative activity of free ligand and its complexes [46].

3.7 Electrochemical Studies

The anti-corrosive property of Schiff base **H₂L** and their metals Cu(II),Co(II) and Ni(II) complexes was measured by electrochemical studies. After the carbon steel (WE) was exposed to the 1.0 mol L^{-1} H_2SO_4 solution for 30 min to complete the steady state of the electrode, EIS measurements were implemented at open circuit potential (OCP) by applying a sine wave potential signal with the frequency from 100,000 to 0.1 Hz and amplitude of 10 mV. The measured EIS data were fitted with an equivalent circuit to extract various parameters as solution resistance (R_s), charge transfer resistance (R_{ct}). It can efficiently calculate the resistive and capacitive behavior at the double layer interface between bulk region and the metal from EIS measurements. In order to consider the heterogeneity effect of the electrode surface, constant phase elements (CPE) are used instead of double layer capacitance (C_{dl}) [47,48]. The physical nature of the CPE impedance can be expressed by the following equation [49]:

$$Z_{CPE} = [Yo(j\omega)_n]^{-1}$$

where Y_o is a proportionality factor, ω is an angular frequency, $j^2 = -1$ is an imaginary number. The obtained values of n are in the range of $0.5 < n > 1$, is an evidence for the surface roughness and inhomogeneity of the electrode surface.

Fig 2a shows the Nyquist representation for carbon steel in 1.0 mol L^{-1} H_2SO_4 acid medium in absence and presence of different concentrations of Schiff base **H₂L**. The Nyquist plots exhibit a single depressed capacitive loop over the frequency range studied. The diameter of the Nyquist loop increases with augmentation in the concentration of Ligand. This confirm that the carbon steel corrosion is inhibited

because the inhibitor formed a defensive film on the carbon steel surface, resulting in the increased charge transfer resistance [34,50]. The similar shape of Nyquist plot reveals that the corrosion mechanism is unaffected by the addition of Schiff base **H₂L** and mainly driven by charge transfer [51]. The electrochemical impedance parameters (R_s, R_{ct}) were obtained by fitting the impedance spectra using equivalent circuit and tabulated in Table 6. The impedance parameters which are recorded in Table 6 were used to describe the impedance characteristics of carbon steel corrosion in blank and at various concentrations of the studied Schiff base **H₂L**. The corresponding R_{ct} values for carbon steel are observed to increase with additive in acid solution that may be due to adsorption of the Schiff base **H₂L** molecule from the bulk of the solution to the available surface of carbon steel. This transfer occurs in a large amount with increasing inhibitor concentration. According to that, CPE is decreased, which can result from a decrease in local dielectric constant and/or, adsorption of the inhibitor molecules at the metal/solution interface. The values of n obtained for this inhibitor system were close to unity which reveal homogeneous and capacitive behavior of the interface [52].
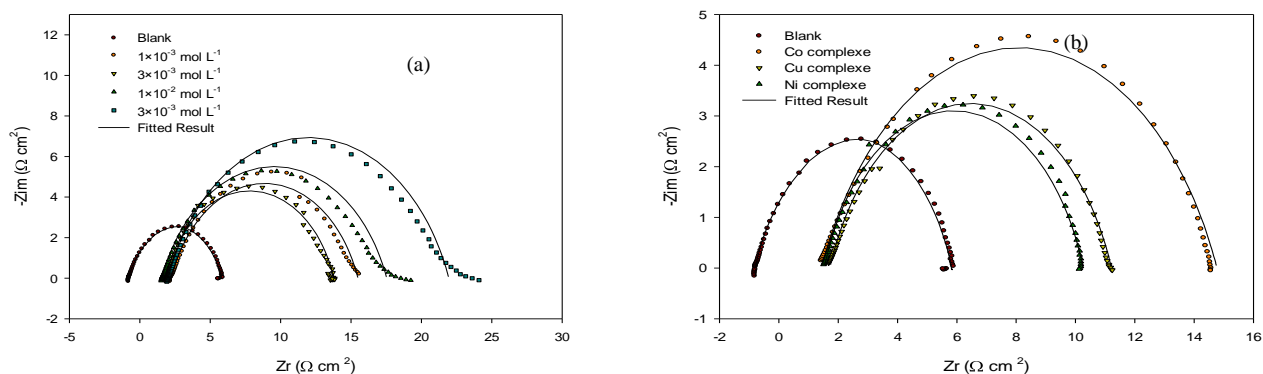
Fig. 2: Nyquist plots of carbon steel in 1.0 mol L^{-1} H₂SO₄ solution (a) at different Schiff base H₂L concentrations (b) at 1.0×10^{-3} mol L^{-1} of metals complexes at 300 K.

The IE% for various concentration was determined from the R_{ct} values by the following equation [51]:

$$\eta_{EIS}\% = \frac{R_{ct} - R_{ct\prime}}{R_{ct}} \times 100$$

where $R_{ct\prime}$ and R_{ct} represents the magnitudes of the inhibited and uninhibited conditions, respectively. From tabulated data in Table 6, it can be inferred that the Schiff base inhibits carbon steel corrosion of mild steel in 1.0 mol L^{-1} M H₂SO₄ at all the studied concentration as well as inhibition efficiency increases as a result of adsorption of the inhibitor molecules on the metal surface. Remarkably, even at the low concentrations (1.0×10^{-3} mol L^{-1}) of the Schiff base **H₂L,** it gave high resistance (≈67%). When the corrosion inhibitor concentration was 3×10^{-2} mol L^{-1}, the corresponding capacitive loop and radius were largest (≈83.0%), which indicated the corrosion inhibition performance for Schiff base H₂L as inhibitor was better at this concentration.

To investigate the durability of metals Cu(II), Co(II), Ni(II) complexes, we conducted an impedance experiment at minimum concentration (1.0×10^{-3} mol L^{-1}), Fig. 1b, and calculated the kinetic parameters in Table 6. These results reassure that the three complexes of Schiff bases possess good corrosion inhibition properties for carbon steel in 1.0 mol L^{-1} M H_2SO_4 solution. The inhibiting efficiency $\eta_{EIS}\%$ was in the following order: $[(HL)Co(H_2O)Cl]5H_2O > [LCu(H_2O)_2]2H_2O > [LNi(H_2O)_2]2H_2O$.

With comparing Schiff base **H₂L** at same concentration, the lower value of capacitance in case $[(HL)Co(H_2O)Cl]5H_2O$ may also result a larger replacement of water molecules by the Schiff base complex through the adsorption at electrode surface. This implies a better interaction of $[(HL)Co(H_2O)Cl]5H_2O$ on carbon steel surface than Schiff base **H₂L** and the other complexes.
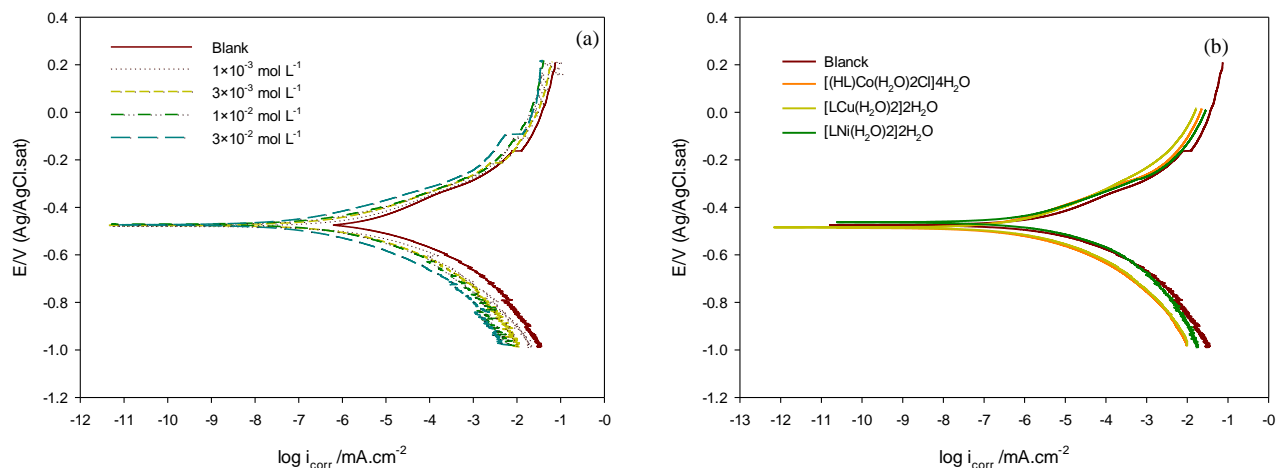
In order to establish the influence of inhibitors and change in concentration on the overall behaviour of carbon steel in 1.0 mol L^{-1} M H_2SO_4 solution, anodic and cathodic potentiodynamic polarization was measured. Regards to the results is presented in Fig. 2a, the individual kinetic descriptors procured from the Tafel curves are recorded in Table 6. Compared with blank solution, the variation in E_{corr} were not obvious between absence and presence of inhibitor. Thus, Schiff base **H₂L** inhibitor was defined as a mixed type inhibitor for carbon steel in test solution. As shown in Table 6, the corrosion current density (i_{corr}) decreased with increased Schiff base inhibitor concentration. Therefore, the corrosion inhibition rate was enhanced and the highest corrosion inhibition efficiency was 80.24 % at 3.0×10^{-2} mol L^{-1} of Schiff base **H₂L**, that agree with resistance data. The surface coverage of inhibitor increased with increased Schiff base **H₂L** inhibitor concentrations because of that. Compared the change of β_c and β_a values with blank solution in Table 6, they were not obvious, which demonstrated that the inhibitor molecules inhibited the corrosion by adsorbing on the surface of carbon steel and reducing the active sites on the metal surface rather than changing the mechanism for anode and cathode reactions [12]. Same trend as impedance data were observed at 1.0×10^{-3} mol L^{-1} of metal complexes in Fig. 3b.

Adsorption isotherm

The adsorption path is usually considered as a substitution process between the organic inhibitor in the aqueous solution [Org_{aq}] and water molecules adsorbed at the metal surface [H_2O_{ads}]. The adsorption isotherms were considered to be a curve that give fundamental information on the interactions between inhibitor and metal surface activity sites [12, 53]. Using impedance data in Table 6, the adsorption of Schiff base **H₂L** molecules preferred the Langmuir isotherm of the form [32]:

$$\theta/(1-\theta) = K_{ads} \, C_{inh}$$

where, θ is the degree of surface coverage; K_{ads} is the equilibrium constant of the adsorption/desorption activity and C_{inh} is Schiff base **H₂L** concentration. The degree of surface coverage, θ (θ= η(%)/100), at different Schiff base **H₂L** inhibitor concentrations in 1.0 mol L^{-1} M H_2SO_4 was evaluated at 300 K.

Fig. 3: Tafel curves of carbon steel in 1.0 mol L^{-1} H_2SO_4 solution (a) at different H₂L Schiff base concentrations; (b) at 1.0×10^{-3} mol L^{-1} of metals complexes at 300 K.

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The adsorption path is usually considered as a substitution process between the organic inhibitor in the aqueous solution [Org$_{aq}$] and water molecules adsorbed at the metal surface [H_2O_{ads}]. The adsorption isotherms were considered to be a curve that give fundamental information on the interactions between inhibitor and metal surface activity sites [12, 53]. Using impedance data in Table 6, the adsorption of Schiff base **H₂L** molecules preferred the Langmuir isotherm of the form [32]:

$\theta/(1-\theta) = K_{ads} C_{inh}$

where, θ is the degree of surface coverage; K_{ads} is the equilibrium constant of the adsorption/desorption activity and C_{inh} is Schiff base **H₂L** concentration. The degree of surface coverage, θ (θ= η(%)/100), at different Schiff base **H₂L** inhibitor concentrations in 1.0 mol L^{-1} M H_2SO_4 was evaluated at 300 K. The fitting lines in Fig. 4 show the Langmuir isotherm with values of slopes, intercepts and linear correlation coefficients (R^2). The K_{ads} value for **H₂L** Schiff base molecules is calculated as 940.366 mol L^{-1} from the slopes of the lines. A higher K_{ads} value connotes a stronger interaction between adsorbate and adsorbent [32,54]. This isotherm assumes that the adsorbed molecules form mono layer on metal surface [53,55]. The K_{ads} was linked to the energy of adsorption (ΔG_{ads}) using the relation [32,56]:

$\Delta G_{ads} = R\ T\ ln\ (55.5 K_{ads})$

where, R and T denoted as per conventional terminology reported in earlier report [32].

The value of ΔG_{ads} for adsorption of the Schiff base **H₂L** molecules was found to be -27.09 kJ mol^{-1}. The negative value of ΔG_{ads} proposes that the Schiff base **H₂L** is spontaneously adsorbed on the carbon steel surface. Literature survey reveals that the values of ΔG_{ads} around -20 kJ mol^{-1} or lower are consistent with physical adsorption, where the electrostatic interaction between the charged molecules and the charged metal occur. Values around -40 kJ mol^{-1} or higher involve charge sharing or transfer from organic molecules to the metal surface to form a coordinate type of metal bond (chemisorption) [57]. The ΔG_{ads} value is around -27 kJ mol^{-1}, revealing that both physisorption and chemisorption processes were involved in the adsorption process of Schiff base H₂L molecules on carbon steel surface.
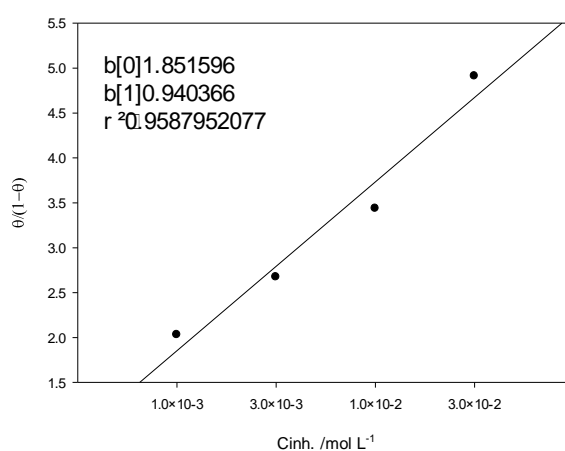
**Fig. 4.** Adsorption isotherm plot of Langmuir for carbon steel in the presence of H₂L Schiff base.

Table 6: Kinetic parameters from EIS and Tafel plots for carbon steel in 1.0 mol L^{-1} H₂SO₄ solution in the absence and presence of H₂L at various concentrations and at 1.0×10^{-3} mol L^{-1} for complexes.

Conc. mol L^{-1}	Impedance measurements					Polarization measurements				
	Rs (Ω cm²)	Rct (Ωcm²)	CPE ($\mu\Omega^{-1}$ s^n cm^{-2})	n	η_{EIS} (%)	-E$_{corr}$ (mV/SCE)	i$_{corr}$ (mAcm^{-2})	B$_a$ (mV/dec)	B$_c$ (mV/dec)	η_p (%)
Blank	-1.88	4.05	22.89	0.809	-	474	4.81	180	134	-
1.0×10^{-3}	1.52	12.27	21.22	0.794	66.99	477	2.12	183	147	55.93
3.0×10^{-3}	2.14	14.88	16.62	0.781	72.78	475	1.45	152	132	69.85
1.0×10^{-2}	1.61	17.97	16.11	0.793	77.46	471	1.14	139	138	76.29
3.0×10^{-2}	1.20	23.94	14.65	0.774	83.08	473	0.95	130	123	80.24

Conc. 1.0×10^{-3} mol L^{-1}	Impedance measurements					Polarization measurements				
[LCu(H₂O)₂]2H₂O	1.52	15.34	16.72	0.788	72.92	474	1.40	173	141	70.89
[(HL)Co(H₂O)Cl]5H₂O	1.48	11.49	18.54	0.763	63.88	477	1.83	175	137	61.95
[LNi(H₂O)₂]2H₂O	1.41	10.39	21.56	0.812	60.06	468	1.93	168	152	59.87

Adsorptive mechanism of Schiff base (H₂L) (Fig. 5)

For adsorption process, four types of adsorption may take place on the metal surface involving organic molecules in aggressive acid media (i) electrostatic attraction between charged molecules and charged metal, (ii) interaction of π-electrons in organic molecules with the metal, (iii) interaction of uncharged

electron pairs in the organic molecules with the vacant *d*-orbital in metal and (iv) a combination of the above [4, 58].
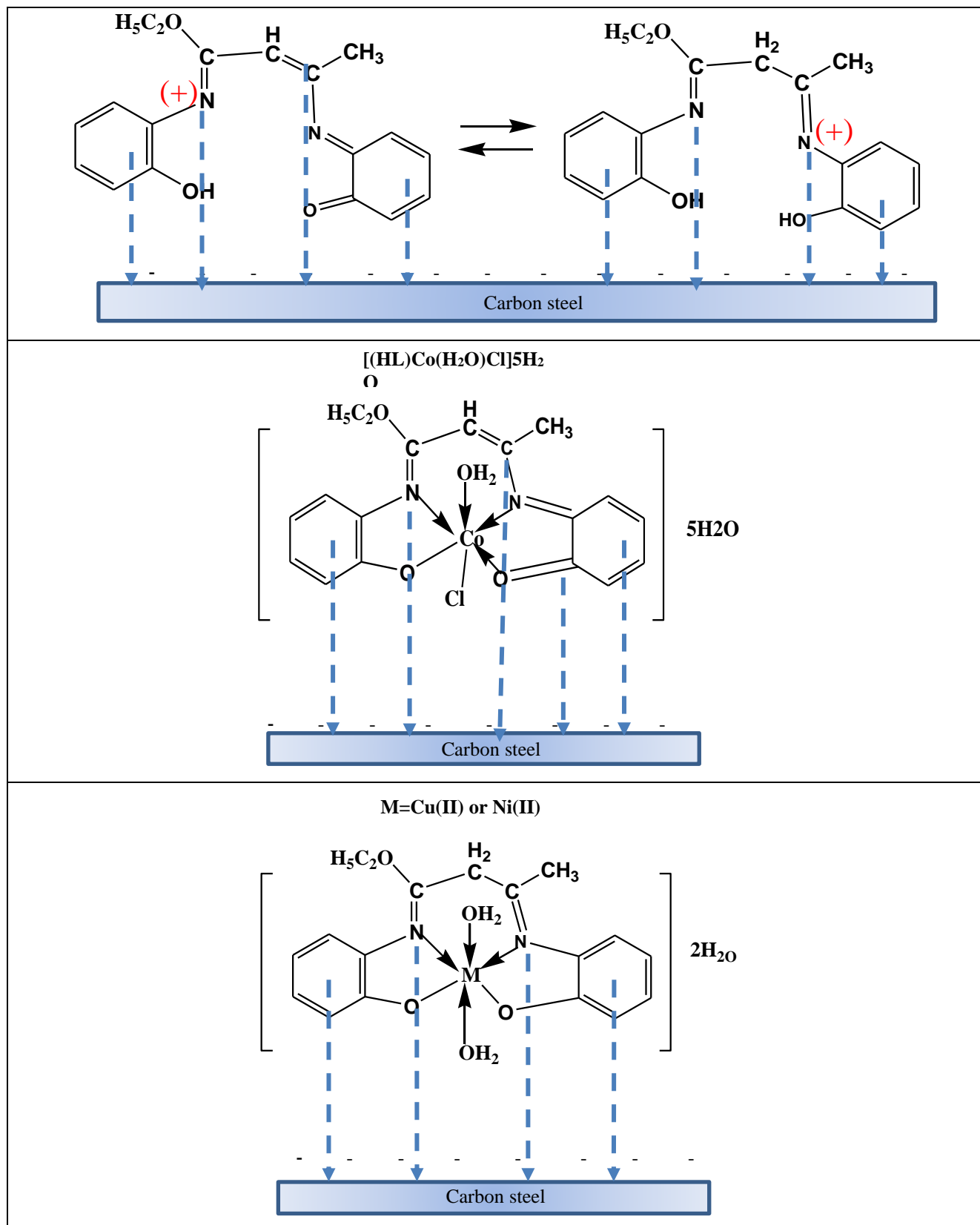
Fig. 5. Schematic diagram representing the adsorption mechanism of Schiff bases H_2L molecules and metals Co(II) and M(II) complexes on carbon steel surface.

In this study, the inhibitor [N-(2-hydroxyphenyl) -3-(2-hydroxyphenylimino) butanamide] **H₂L** contains two aromatic rings, bigger conjugated system nitrogen, sulphur, oxygen electronegative atoms and two-CH=N - groups. As it known, the - CH=N - functional group of Schiff base has good stability, oxidative reduction and excellent coordination ability. The above data shows that the corrosion inhibitors **H₂L** Schiff base have good corrosion inhibition performance for carbon steel in 1.0 mol L^{-1} H_2SO_4 solution, which may result from physical and chemical interaction between **H₂L** Schiff base molecules and carbon steel surface, according ΔG_{ads} value which is –27 kJ mol^{-1}. The adsorption of **H₂L** on the carbon steel surface could be explained in two modes: *firstly*, at low pH, the carbon steel surface (adsorbents) is positively charged as a result of adsorption of H^+ in acidic solution. Then SO_4^{2-} through the interaction of coulomb force was preferentially adsorbed onto the surface of carbon steel. Therefore, carbon steel surface was transformed from negative charge selectivity to positive. The positive surface facilitating the adsorption of protonated **H₂L** molecules (protonation of nitrogen atoms of -CH=N- group) on the cathodic sites of carbon steel surface by the electrostatic action and decreases the evolution of hydrogen. *Secondary*, **H₂L** molecules adsorbed chemically on carbon steel due to interaction of π electrons of the benzene ring of the **H₂L** with vacant *d*-orbitals of iron. This adsorption can be referred to the complex formation between **H₂L** molecule and iron and lead to decrease the anodic dissolution of carbon steel. *Thirdly,* due to the presence of multi adsorption centres, the flat orientation of the entire **H₂L** molecule with respect to the steel surface could lead to the interaction of π-electrons of the aromatic ring as well as -CH=N- groups with the metal surface. It can be explained that **H₂L** molecules adsorb and block the essential part of the active sites on the carbon steel surface by forming strong adsorption [32,57- 60]. The same factors explain the high inhibition efficiency of **H₂L** complexes. Low concentrations of complexes inhibitor in the acidic solution was insufficient to form a compact protective layer formed by the mutual interaction of complex and metal ions.

Finally, the larger size and high molecular weight of Schiff base inhibitors can also cause higher inhibition efficiency. Inhibition activity of [(HL)Co(H₂O)Cl]5H₂O complex on carbon steel gets enhanced significantly due to the presence of chloride ion inside the coordination sphere, which possesses many lone pair of electrons which enhances the bond with the steel.

Conclusion

Cu(II), Co(II), Ni(II), Mn(II), Pt(IV), Fe(III) and Ag(I) complexes of Schiff base N-(2-hydroxyphenyl)-3-(2-hydroxyphenylimino were prepared and characterized using ^{1}HNMR Spectra and IR techniques. [LCu(H₂O)₂]2H₂O and [LMn₂(H₂O)₂]Cl₂.4H₂O Schiff base complexes reveal great biological activity

performance (inhibition zone about 22 mm for Esherichia coli) than the Schiff base and other complexes and Ampicillin towards Esherichia coli and Staphylococcus aureus. Anticorrosion data demonstrate that the **H₂L** Schiff base and its complexes not only forms a protective layer on the electrode surface, but also strongly adsorbs to the surface and hence, increases the durability and corrosion resistance of the electrode in acidic conditions. The adsorption on the carbon steel surface occurs through physical and chemical action, thus forming a dense protective film, preventing the corrosion of carbon steel by acid solution.

Conflict of interest

The authors declare that they have no conflicts of interest.

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