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Synthesis, Characterization and Antimicrobial Activity of Schiff Base Complexes of Copper (II), Cobalt (II) and Nickel (II) Derived from 4,4' - Diamino Diphenyl Methane (DDM) and 4,4'-Diamino Diphenyl Ether (DDE) with salicylaldehyde

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Abstract

Tetra dentate N_2O_2 type complexes of Cu (II), Co (II) and Ni (II) ions have been synthesized using the Schiff bases formed by the condensation of salicylaldehyde (2-hydroxy benzaldehyde) with two diamines, 4,4\ - diamino diphenyl methane(DDM) to give Schiff base (L_1) and 4,4'-diamino diphenyl ether (DDE) to give Schiff base (L_2). Molar conductance , FTIR spectroscopy, UV- visible spectroscopy studies have been carried out to determine the structures of these complexes, from these data it is found that all the complexes posses general stereochemistry [M(L)]2.X,(L= Schiff base (L_1 and L_2) ; $X=Cl_4$ when M= Cu (II), Co (II) and $X=(CH_3COO)_2$ when M= Ni (II), The ratio 2:2 (M:L) complexes showed square planer geometry around Ni(II), Co(II) and Cu(II) ions. FTIR spectra showed that ligands (L_1 and L_2) is coordinated to the metal ions through imino nitrogen (–CH=N–) and phenolic (–OH) oxygen. The Molar conductance data reveal that all synthesized complexes are electrolyte. Another interesting results of this study is the fact that the molar conductance of Cu (II) complexes of Schiff base (L_1 and L_2) (L_1Cu and L_2Cu) are higher than those of Ni(II) and Co (II) complexes with the same ligand, (L_1Ni and L_1Co) and (L_2Ni and L_2Co) respectively. The Ligands ($L_1$and L_2) and their complexes were screened for antimicrobial activity by the well diffusion technique using DMSO as a solvent. The biological activity studies showed that all the prepared complexes are antimicrobially active and showed higher activity than free ligands, the complexes derived from ligand L_2 gave higher activity than L_1 complexes

Keywords: Schiff base, Complexes, DDM, **DDE,** Spectral studies, antimicrobial activity.

الخلاصة

تم تحضير معقدات كل من Cu (I I) , Ni (I I), Co (I I) من نوع N_2O_2 الثنائية السن باستعمال قواعد Schiff المتكونة بمفاعلة الديهايد (2-hydroxy benzaldehyde) مع إثنين من الأمينات الثنائبة الأول هو 4,4\-Diamino diphenyl methane (DDM) ليعطي **Schiff base L_1** والثاني هو 4,4\-Diamino diphenyl ether (DDE) ليعطي **Schiff base L_2**. من دراسة قياسات التوصيلية المولاريه ومطيافية الاشعه تحت الحمراء ومطيافية الاشعه المرئيه وفوق البنفسجيه تم تحديد تراكيب وأشكال المعقدات المحضرة . حيث وجد أن جميع المعقدات المحضرة تمتلك اشكل الهندسي الفراغي $[M(L)]_2.X$ (L= Schiff base (**L_1** and **L_2**) عندماM= Cu (I I),Co(I I) $X=Cl_4$; و M= Ni (I I), عندما $X=(CH_3COO)_2$]. لقد أوضحت النسبة 2:2 (M:L) بأن الشكل الهندسي حول أيونات Ni(I I)،Co(I I),Cu (I I) هو مربع مستوي . كما أوضحت أطياف الأشعة تحت الحمراء بأن الليكاندات المحضرة (**Schiff base L_1** ، **Schiff base L_2**) تتناسق مع ايونات العناصر من خلال ذرة النايتروجين في (–CH=N–) وذرة الأوكسجين في مجموعة(–OH). قياسات التوصيلية المولاريه اوضحت أن جميع المعقدات المحضرة هي الكتروليتات ، من النتائج المهمة الأخرى المستحصلة من هذه الدراسة هي حقيقة أن قيم التوصيليه المولاريه لمعقدات Cu (I I) **(L_1Cu ، L_2Cu)** هي أكبر مقارنة بقيم التوصيليه المولاريه لمعقدات Ni(I I) Co(I I) , مع نفس الليكاندات **(L_1Ni , L_1Co) ,(L_2Ni , L_2Co).** كذلك تمت دراسة الفعالية البايولوجية لقواعد شف المحضرة ومعقداتها ومقارنة فعالية كل من الليكندين المحضرين مع المعقدات المحضرة حيث اثبتت الدراسة ان المعقدات المحضرة اعطت فعالية بايولوجية اكبر من الفعالية البايولوجية لكلا الليكندين وان معقدات الليكند L_2 اعطت فعالية بايولوجية اكبر من معقدات الليكند L_1.

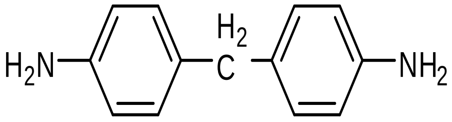
1. Introduction

A large number of Schiff bases and their complexes have been studied for their interesting and important properties, e.g., their ability to reversibly bind oxygen (1), catalytic activity in hydrogenation of olefins (2) and transfer of an amino group (3), photochromic properties (4), and complexing ability towards some toxic metals (5). The high affinity for the chelation of the Schiff bases towards the transition metal ions is utilized in preparing their solid complexes.

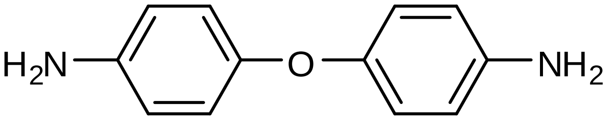
Hydroxyl Schiff base ligands derived from the reaction between salicylaldehyde and mono amine have been studied extensively in complex forming reaction (6).

Schiff base complexes of transition metals or nontransition metal and both of them have a very important role in a different aspect of coordination chemistry to transition metals (6, 7). Schiff bases as a ligand have used to form a very stable complex of metal ions (8). Which have been used in catalysis, magnetic behavior and biological activity (9-11).

Many recent works showed that some drugs have a higher activity if it administered as a metal complexes rather than as an organic compounds (12, 13). Metal complexes derived from Schiff bases of *p*-substituted aniline are becoming increasing important in biological, clinical and analytical fields. (14-16). Diligand complexes of some metal (II) have been reported. This study reported the complexation reactions of the hydroxy Schiff bases derived from aromatic aldehyde (2-Hydroxybenzaldehyde) and aromatic diamine, where two different types of Schiff base derived from diamine [4, 4' -diamino diphenyl methane (DDM) (I) and 4, 4'-diamino diphenyl ether (DDE) (II)] with salicylaldehyde have been used to synthesize compounds containing diligand complaxation metals and compare the biological activity of these ligands and complexes.



DDM (I)



DDE (II)

2. Experimental

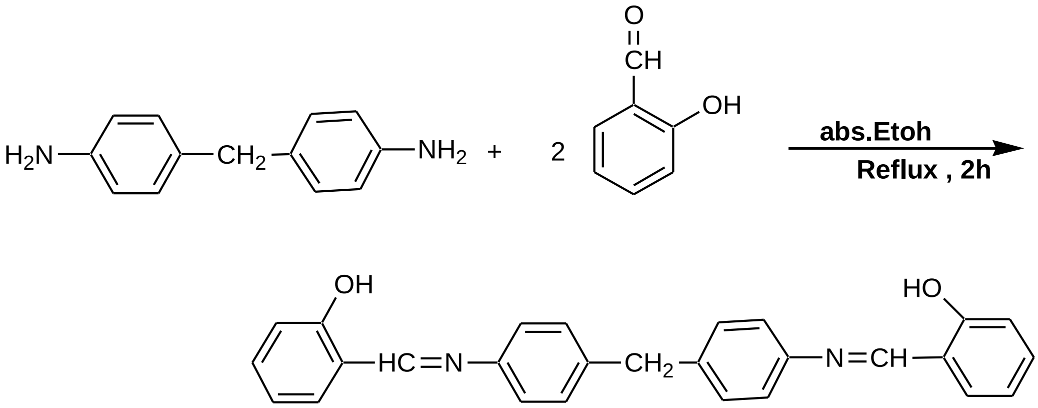
2.1. General

All the chemical compounds, the reagent and the solvents were purchased from BDH and Fluka. Infrared spectra between (400-4000 cm^{-1}) of the synthesized Schiff base compounds and their complexes were recorded by FT-IR spectrophotometer (FTIR-8400 Shimadzu-Japan) as KBr disc. The electronic spectra were recorded on the UV-visible spectrophotometer type (spectro SC) in various solvents. The melting points were recorded on “Gallenkamp Melting Point Apparatus”. The conductance measurements were recorded on W.T.W conductivity Mester LBR meter (Germany). All instruments in chemistry department, science college, Basrah University

2.2 Synthesis of Schiff bases L_1 and L_2

2.2.1 Synthesis of Schiff base (L_1)

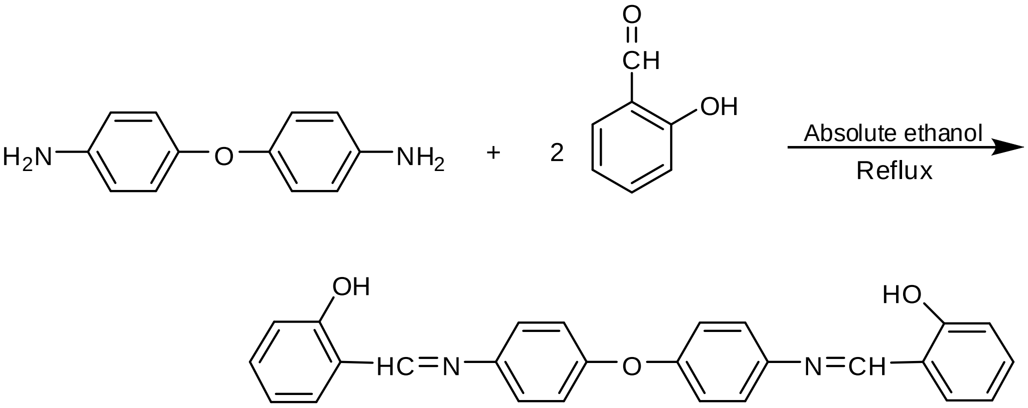
A hot solution (60 °C) of 4,4'-Diamino diphenyl methane (DDM) (3.96 g, 20 mmol) was mixed with a hot solution (60 °C) of 2-Hydroxy benzaldehyde (4.88 g, 40 mmol) in (50 ml) of absolute ethanol. The resulting mixture was left under reflux for 2 h in a water bath in the presence of a few amount of *p*-toluene sulphonic acid as a condensing agent. The solid product formed was separated by filtration, purified by recrystallization from ethanol, washed with diethyl ether and then dried in a vacuum over anhydrous calcium chloride. The dark yellow precipitate is produced in 92.3 % yield. M.p (197-198 °C)



L_1

2.2.2 Synthesis of Schiff base L_2

A hot solution (60 ^{o}C) of $4,4^{\backslash}$-Diamino diphenyl ether (DDE) (4 g, 20 mmol) was mixed with a hot solution (60 ^{o}C) of 2-Hydroxy benzaldehyde (4.88 g, 40 mmol) in ethanol (50 ml). The resulting mixture was left under reflux for 4 h in a water bath in the presence of a few amount of *p*-toluene sulphonic acid as a condensing agent. The solid product formed was separated by filtration, purified by recrystallization from ethanol, washed with diethyl ether and then dried in a vacuum over anhydrous calcium chloride. The pale-yellow precipitate was produced in 86 % yield. M.p (201-202 ^{o}C)



L_2

2.3 Synthesis of metal (II) complexes

The complexes were prepared by the same general method. Details are given for representative case only.

2.3.1 Synthesis of Cu (II), Ni (II) and Co (II) complexes with Schiff base L_1, L_2.

Schiff base **L_1**(2.03 gm ,5 mmol) ,L_2 (2.05 gm,5mmol) dissolved in 25 ml ethanol was added to Copper(II) chloride dihydrate (0.85 gm , 5 mmol) or nickel acetate tetra hydrate (1.24 gm , 50 mmol) or cobalt(II) chloride hexahydrate (1.18 gm , 5 mmol) each dissolved in ethanol(25 ml) . Then, the mixture was refluxed for 3 hrs on a water bath. On cooling to room temperature and keeping at 5 ^{o}C, the complexes L_1Cu, L1Ni and L_1Co which separated out as powdery solids respectively were collected on a filter paper and washed with ethanol and ether and dried in vacuum.

2.4. Biological activity

The biological activity of the prepared complexes and Schiff base were tested in vitro against the bacteria Escherichia coli, Bacillus subtilis and Klebsiella by using the well diffusion method and nutrient agar as the medium. Stock solution (10^{-3}M) was prepared by dissolving the compounds in DMSO solution. The well was made on the agar medium inoculated with microorganisms and filled with the test solution using a micropipette and the plate was incubated at 35 ^{o}C for 24 h. during this period, the test solution diffused and the growth of the inoculated microorganisms was affected. Antibacterial activity was indicated by the presence of clear inhibition of zone around the well.

3. Results and discussion

3.1 Analysis of FTIR spectra and mode of bonding:

The IR spectra provide valuable information regarding the nature of functional group attached to the metal ion atom. In order to study the binding mode and determine the coordination sites that may be involved in chelation of the synthesized Schiff bases **L_1** and **L_2** to the Cu (II), Ni (II) and Co (II) ions in the synthesized complexes, the IR spectrum of the ligands (Schiff bases **L_1** and **L_2**) were compared with the spectra of the synthesized complexes. There were some guide peaks in the spectra of the ligands, which were helpful in achieving this goal. The positions and intensities of these peaks are expected to change upon chelation, The IR data of the spectra of synthesized Schiff bases **L_1** and **L_2** and their synthesized complexes are presented in Table 1. The IR spectra of the Schiff bases **L_1** and **L_2** showed strong bands at 1611 cm^{-1} and 1624 cm^{-1} respectively and they are assigned as due to the –CH=N– stretching frequency (17). The strong but sharp band appearing at 1260 cm^{-1} in Schiff bases **L_1** and at 1264 cm^{-1} in Schiff bases **L_2** are assigned to the C–O (phenolic) stretching frequency, while the broad bands that appearing in the range 3240-3530 cm^{-1} and 3250-3550 cm^{-1} in Schiff bases **L_1** and **L_2** respectively are assigned to O–H stretching frequency. Medium bands, due to in plane and out of plane O–H bending vibrations in (phenolic group) are appeared in 1449 cm^{-1} and 758 cm^{-1} in Schiff bases **L_1** but in 1440 cm^{-1} and 753 cm^{-1} in Schiff bases **L_2** respectively(18). See Figures 1 and 2. In the IR spectrum of the complexes between Schiff bases **L_1** and **L_2** with Cu (II), Ni(II) and Co (II) ions, the prominent infrared spectral bands that assigned as the −CH=N– stretching frequency bands in the synthesized complexes suffered from changing in the shape and position as compared to their free ligands that appeared in the range 1598-1602 cm^{-1}, a lowering by 9-13 cm^{-1} compared to the bands observed for the ligand Schiff base **L_1** and in the range 1608-1620 cm^{-1}, a lowering by 5-18 cm^{-1} compared to the bands observed for the ligand Schiff base **L_2** and this indicating coordination through azomethine nitrogen –CH=N– to the central metal ion. The phenolic C–O band appearing at 1260 cm^{-1} and 1264 cm^{-1} in Schiff bases **L_1** and **L_2** also shift towards higher frequency region at 1272-1276 cm^{-1} in Schiff base **L_1** complexes but at 1275 cm^{-1} in Schiff base **L_2** complexes. The O–H band in the synthesized Schiff bases complexes suffered from changing in the shape and position as compared to their free ligands. Also, from IR spectra we noticed that the in plane and out of plane O–H bending appearing in 1449 cm^{-1} and 758 cm^{-1} respectively for the Schiff base **L_1** shift towards lower frequency region at 1412-1441 cm^{-1} and 748-752 cm^{-1} respectively in Schiff base **L_1** complexes but there are appearing in 1440 cm^{-1} and 753 cm^{-1} respectively in Schiff base **L_2** complexes and shift towards lower frequency region at 1412cm^{-1} and 746-750 cm^{-1} respectively in Schiff base **L_1** complexes. All this suggests coordination through phenolic oxygen there by rendering bidentate chelation of the Schiff bases with the metal ions. Conclusive evidence of the bonding is also shown by the observation that new bands in the IR spectra of the metal complexes appear at 559-581 cm^{-1} and at 566-588 cm^{-1} assigned to M–N stretching vibrations in Schiff bases **L_1** and **L_2** complexes respectively such as (L_1Ni, L_1Cu, L_1Co) and (L_2Ni, L_2Cu, L_2Co) appears at (581 cm^{-1}, 575 cm^{-1} and 559 cm^{-1}) and (588 cm^{-1}, 578 cm^{-1}566 cm^{-1}) respectively. Also new bands appeared at 430-517 cm^{-1} and at 440-528 cm^{-1} assigned to M–O stretching(19) vibrations in Schiff bases **L_1** and **L_2** complexes respectively such as (L_1Ni, L_1Cu, L_1Co) and (L_2Ni, L_2Cu, L_2Co) appears at (517 cm^{-1}, 449cm^{-1} and 430 cm^{-1}) and (528 cm^{-1}, 460 cm^{-1} and 440 cm^{-1}) respectively. See Figures 1 and 2. The M–N and M–O stretching vibrations are not observed in the spectra of the free ligands

Table 1: Selected IR data (4000-400cm^{-1}) of Schiff bases (L_1 and L_2) and their complexes (s=strong, m=medium, w=weak, br=broad).

IR bands	**Compounds**							
	L_1	**L_1Cu**	**L_1Ni**	**L_1Co**	**L_2**	**L_2Cu**	**L_2Ni**	**L_2Co**
O-H Stretching (br,s)	3240 3530	3325- 3500	3232- 3520	3220- 3514	3250- 3550	3231- 3515	3240- 3525	3224- 3521
C=N Stretching (s)	1611	1600	1602	1598	1624	1608	1614	1620
In plane O-H bending (m)	1449	1412	1441	1414	1440	1412	1414	1412
C-O Stretching (s)	1260	1275	1276	1272	1264	1275	1277	1275
Out of plane O-H bending (m)	758	748	752	752	753	748	750	746
M-N Stretching (w)	------	575	581	559	------	578	588	566
M-O Stretching (w)	------	449	517	430	------	460	528	440

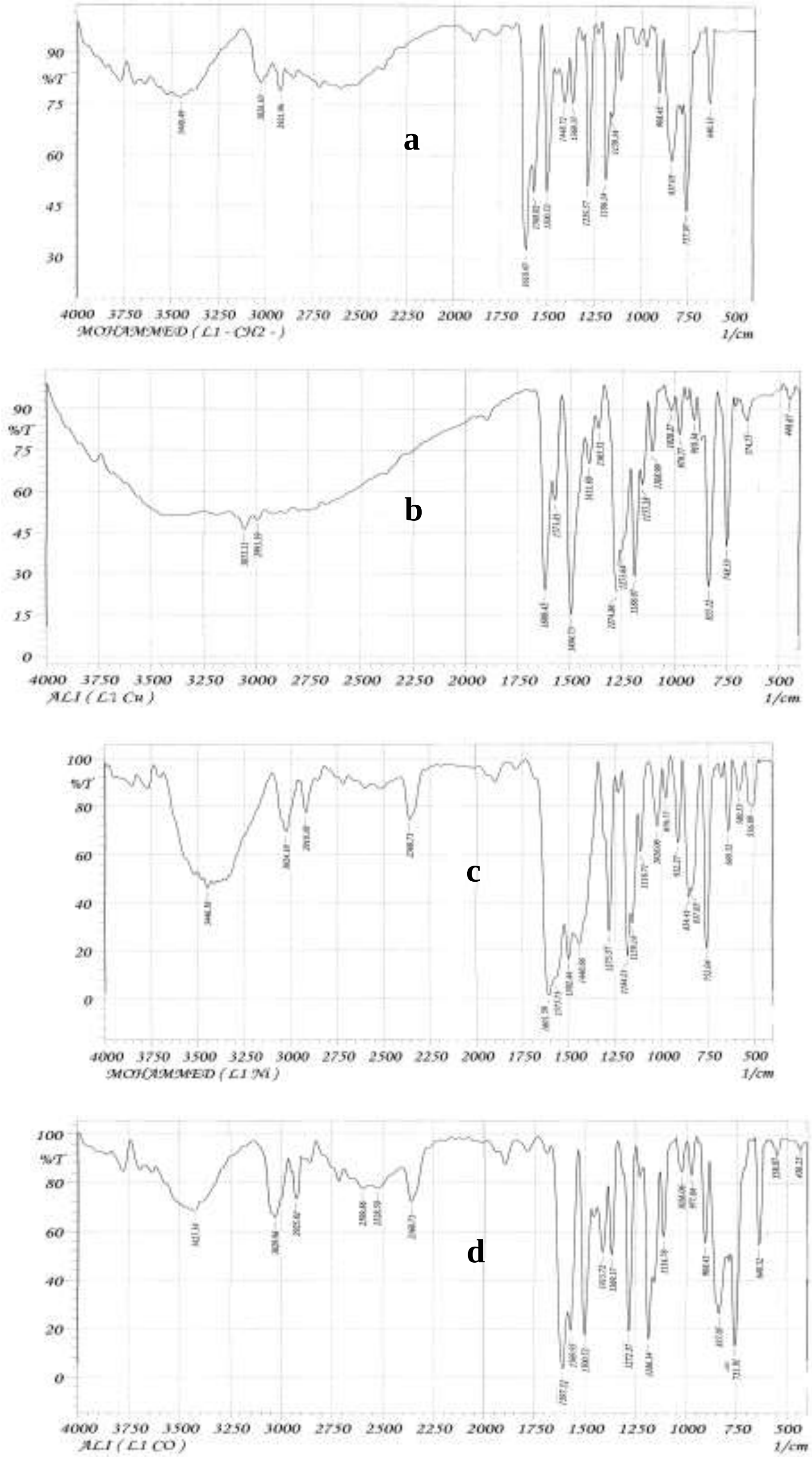


Figure 1: FTIR infrared spectrum for (a): L_1, (b): L_1Cu ,(c): L_1Ni , (d): L_1Co.

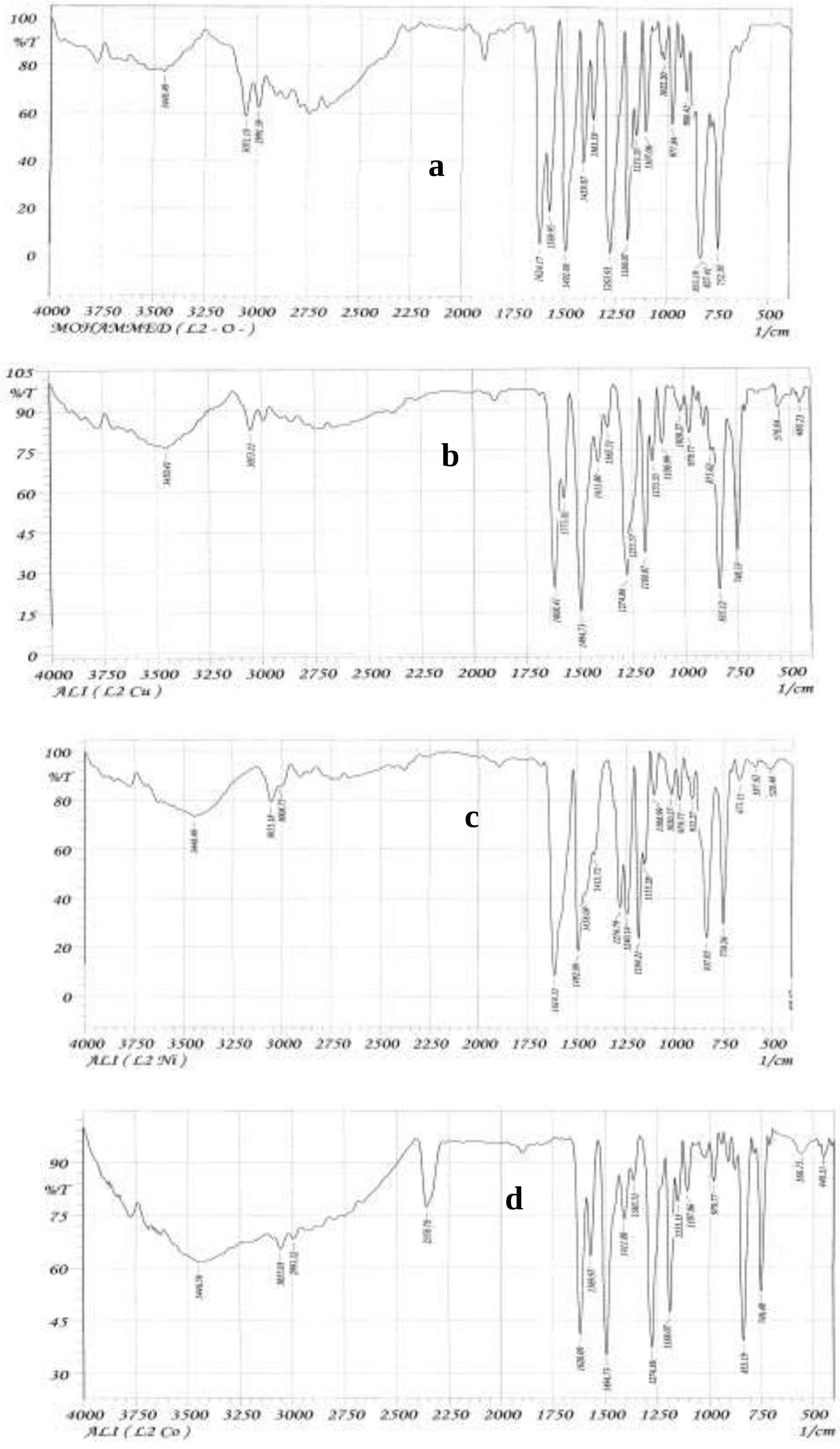


Figure 2:FTIR infrared spectrum for (a): L_2, (b): L_2Cu, (c): L_2Ni, (d): L_2Co.

3.2 Electronic absorption spectra

The electronic absorption spectra of the synthesized Schiff bases $\mathbf{L_1}$ and $\mathbf{L_2}$ and their complexes with Cu (II), Ni (II) and Co (II) ions in DMSO solvent were recorded at 27°C. The absorption region, band assignment and the proposed geometry of the complexes are given in Table 2. The electronic absorption spectra of Schiff bases $\mathbf{L_1}$ and $\mathbf{L_2}$ showed strong to medium absorption peaks in the uv-region of the spectrum. Strong absorption band observed at 230 nm and 245 nm are assignable to $\pi \rightarrow \pi^*$ transition in the Schiff bases $\mathbf{L_1}$ and $\mathbf{L_2}$ respectively whereas the medium absorption bands appearing at 270 nm and 286 nm are assigned to the $n \rightarrow \pi^*$ transition(20). The $\pi \rightarrow \pi^*$ and $n \rightarrow \pi^*$ transitions for the free ligands were shifted to higher wavelength as a consequence of the coordination to the metal ions indicating information of Schiff bases metal complexes(21) .

The electronic spectra of the copper (II) complexes **[** $\mathbf{L_1Cu}$ and $\mathbf{L_2Cu}$ **]** showed strong band at 255 nm and 265 nm ($\pi \rightarrow \pi^*$ transition in ligands $\mathbf{L_1}$ and $\mathbf{L_2}$ and at 300 nm and 310 nm ($n \rightarrow \pi^*$ transition in ligands $\mathbf{L_1}$ and $\mathbf{L_2}$) respectively, in addition these spectra showed two bands at 530 nm and 580 nm in $\mathbf{L_1Cu}$ and at 560 nm and 602 nm in $\mathbf{L_2Cu}$ these bands are assigned to $\mathbf{{}^2B_{1g} \rightarrow {}^2A_{1g}}$ and $\mathbf{{}^2B_{1g} \rightarrow {}^2B_{2g}}$ transitions respectively which strongly favour square-planner geometry around the central metal ion(22). The nickel (II) complexes **[** $\mathbf{L_1Ni}$ and $\mathbf{L_2Ni}$ **]** showed strong band at 248 nm and 255 nm ($\pi \rightarrow \pi^*$ transition in ligands $\mathbf{L_1}$ and $\mathbf{L_2}$**)** and at 290 nm and 300 nm ($n \rightarrow \pi^*$ transition in ligands $\mathbf{L_1}$ and $\mathbf{L_2}$) respectively, in addition these spectra showed two bands at 470 nm and 514 nm in $\mathbf{L_1Ni}$ and at 485 nm and 550 nm in $\mathbf{L_2Ni}$ and these prominent d-d bands observed are assigned as the $\mathbf{{}^1A_{1g} \rightarrow {}^1A_{2g}}$ and $\mathbf{{}^1A_{1g} \rightarrow {}^1B_{1g}}$ transitions(22), which also indicates the square-planner geometry. The cobalt (II) complexes **[** $\mathbf{L_1Co}$ and $\mathbf{L_2Co}$ **]** showed strong band at 263 nm and 259 nm ($\pi \rightarrow \pi^*$ transition in ligands $\mathbf{L_1}$ and $\mathbf{L_2}$ and at 310 nm and 314 nm ($n \rightarrow \pi^*$ transition in ligands $\mathbf{L_1}$ and $\mathbf{L_2}$) respectively, in addition these spectra showed a d-d band at 570 nm and 590 nm in $\mathbf{L_1Co}$ and $\mathbf{L_2Co}$ assigned to $\mathbf{{}^1A_{1g} \rightarrow {}^1B_{1g}}$ transition(22), which confirms square-planner geometry. See Figures 3 and 4.

Table 2: Electronic absorption spectral data of synthesized Schiff bases L_1 and L_2 and their complexes in DMSO solvent.

Compounds	Absorption (nm)λ	ν_{max} ΔE (cm^{-1})	Band assignment	Geometry
L_1	**230**	**43478**	$\pi \rightarrow \pi^*$	………..
	270	**37037**	$n \rightarrow \pi^*$	
L_1Cu	**255**	**39216**	$\pi \rightarrow \pi^*$	Square-planer
	300	**33333**	$n \rightarrow \pi^*$	
	530	**18868**	$^2B_{1g} \rightarrow {}^2A_{1g}$	
	560	**17241**	$^2B_{1g} \rightarrow {}^2B_{2g}$	
L_1Ni	**248**	**40323**	$\pi \rightarrow \pi^*$	Square-planer
	290	**34483**	$n \rightarrow \pi^*$	
	470	**21277**	$^1A_{1g} \rightarrow {}^1A_{2g}$	
	485	**19455**	$^1A_{1g} \rightarrow {}^1B_{1g}$	
L_1Co	**263**	**38023**	$\pi \rightarrow \pi^*$	Square-planer
	310	**32258**	$n \rightarrow \pi^*$	
	570	**17544**	$^1A_{1g} \rightarrow {}^1B_{1g}$	
L_2	**245**	**40816**	$\pi \rightarrow \pi^*$	……….
	286	**34965**	$n \rightarrow \pi^*$	
L_2Cu	**265**	**37736**	$\pi \rightarrow \pi^*$	Square-planer
	310	**32258**	$n \rightarrow \pi^*$	
	580	**17857**	$^2B_{1g} \rightarrow {}^2A_{1g}$	
	602	**16611**	$^2B_{1g} \rightarrow {}^2B_{2g}$	
L_2Ni	**255**	**39216**	$\pi \rightarrow \pi^*$	Square-planer
	300	**33333**	$n \rightarrow \pi^*$	
	514	**20619**	$^1A_{1g} \rightarrow {}^1A_{2g}$	
	550	**18182**	$^1A_{1g} \rightarrow {}^1B_{1g}$	
L_2Co	**259**	**38610**	$\pi \rightarrow \pi^*$	Square-planer
	314	**31847**	$n \rightarrow \pi^*$	
	590	**16949**	$^1A_{1g} \rightarrow {}^1B_{1g}$	

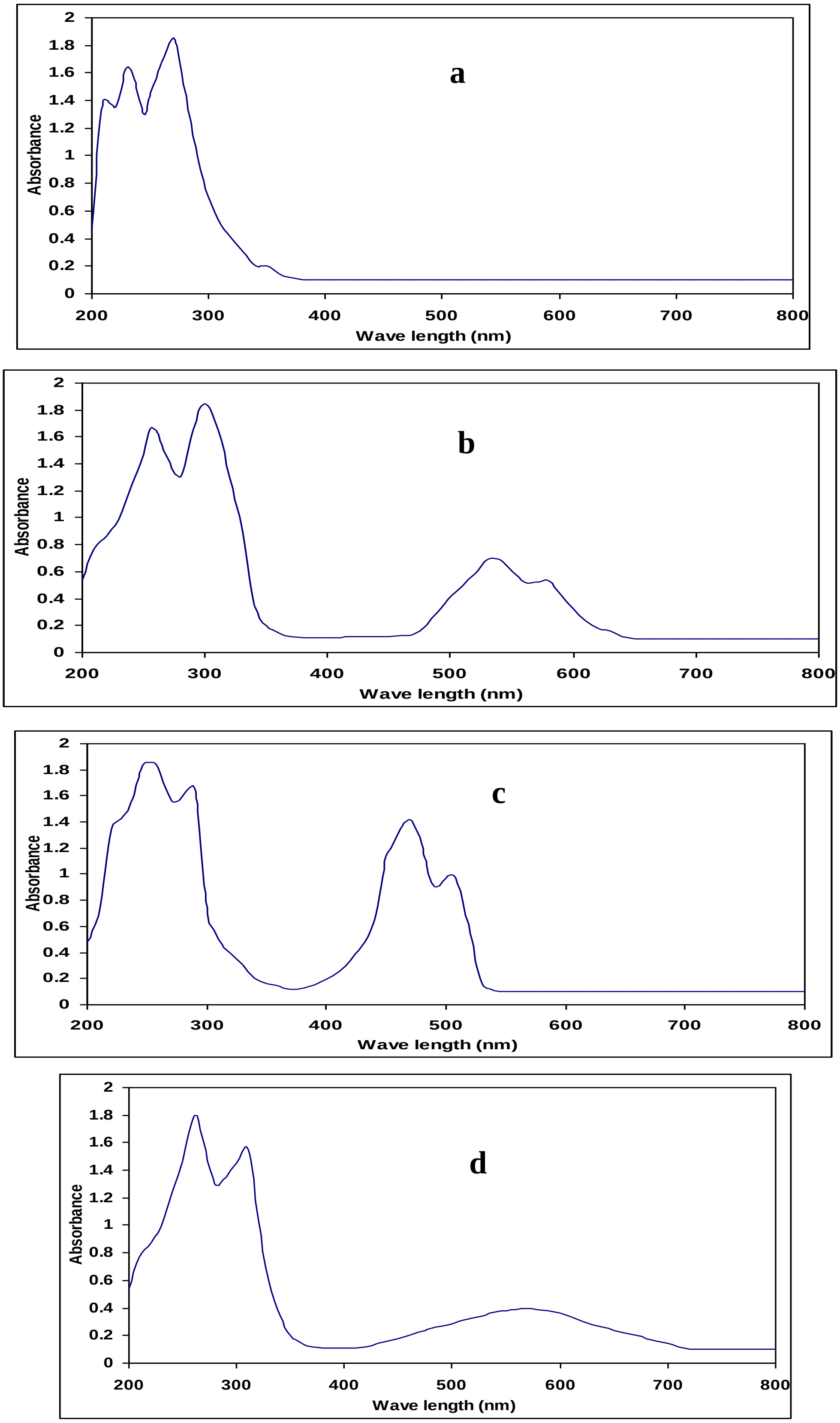


Figure 3UV-Visible Absorption spectra of Schiff base L_1 and its complexes (a):L_1 (b): L_1Cu (c): L_1Ni (d): L_1Co.

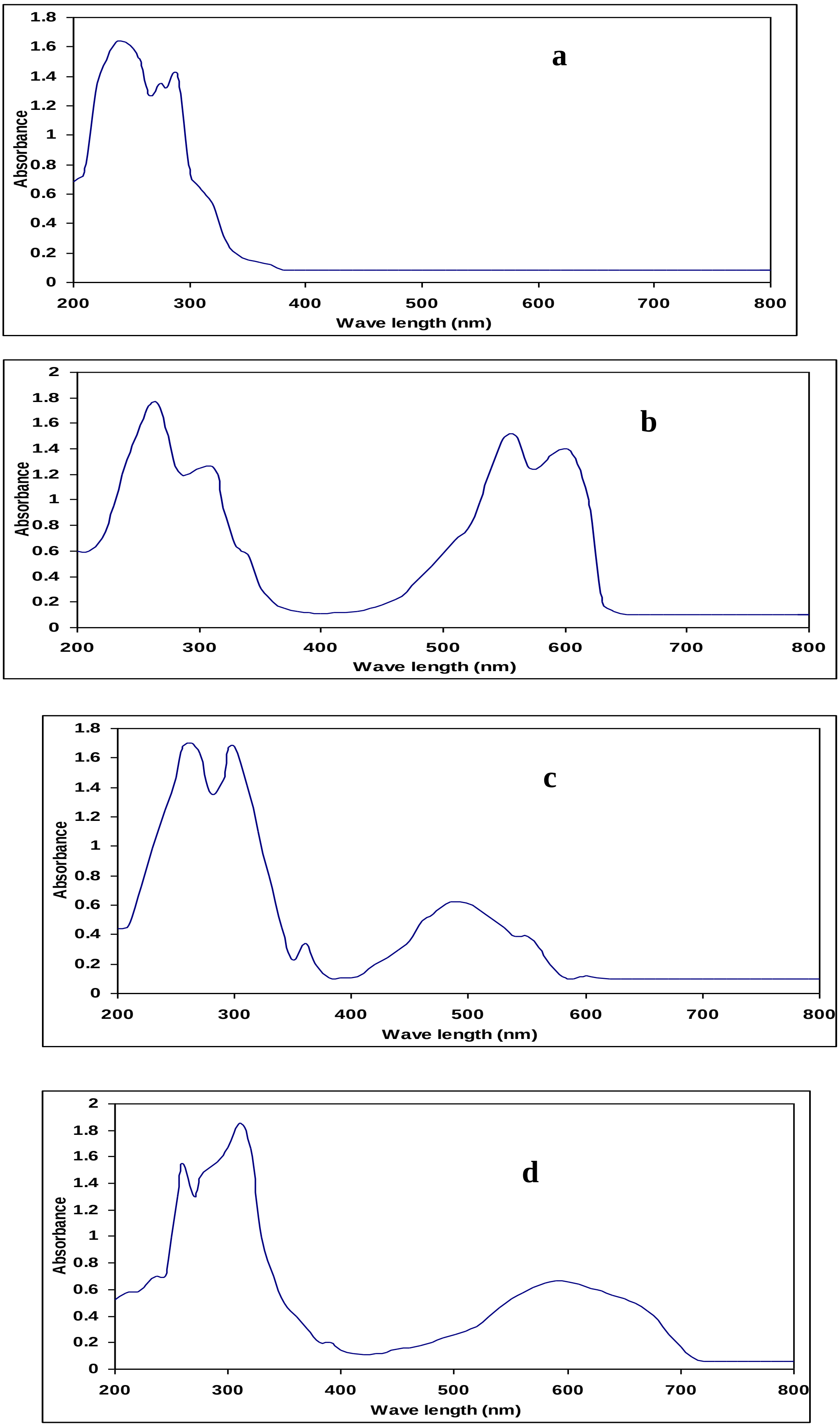


Figure 4:UV-Visible Absorption spectra of Schiff base L_2 and its complexes (a): L_2 (b): L_2Cu (c): L_2Ni (d): L_2Co.

3.3 Molar conductivity measurements

By using the relation $\Lambda_m = K / C$, the molar conductance of the complexes (Λ_m) can be calculated, where C is the molar concentration of the metal complexes solutions. The chelates were dissolved in DMSO and the molar conductivities of 10^{-3}M of their solutions at 25±2 °c were measured. Table (3) shows the molar conductance values of the synthesized complexes between Schiff bases L_1 and L_2 with Cu (II), Ni(II) and Co (II) ions. It is concluded from these values that Cu (II), Ni(II) and Co (II) chelates of Schiff bases L_1 and L_2 ligands are found to have high molar conductance values indicating the ionic nature of these complexes. For example the L_1Cu and L_2Cu have molar conductance values of 83 and 71 $Ohm^{-1}cm^2mol^{-1}$, respectively. Furthermore, it indicates the non-bonding of the chloride anions to synthesized Cu (II) and Co (II) complexes and the non-bonding of the acetate anions to synthesized Ni (II). On the other hand, the molar conductivity values of the L_1Cu and L_1Ni complexes are higher than values of the L_2Cu and L_2Ni complexes respectively. But we found the molar conductivity value L_1Co is lower than L_2Co. For interesting, we concluded that the higher conductance values of chelates support their electrolytic nature of metal complexes.

3.4. Biological activity

Antibacterial activity of the ligands L_1 and L_2 and its complexes with Cu (II), Co (II) and Ni (II) have been carried out against bacteria Escherichia coli, Bacillus subtilis and Klebsiella by the well diffusion method(23). It is observed from this studies that metal chelates have higher activity than the free ligand and its found that the complexes derived from ligand L_2 give higher activity than the complexes derived from ligand L_1 .Table 4 show this effect. Figure 5 shows various antibacterial activities of the Schiff bases and complexes. The data in figure 5 indicates that the Schiff base complexes have an antibacterial activity against the three species.

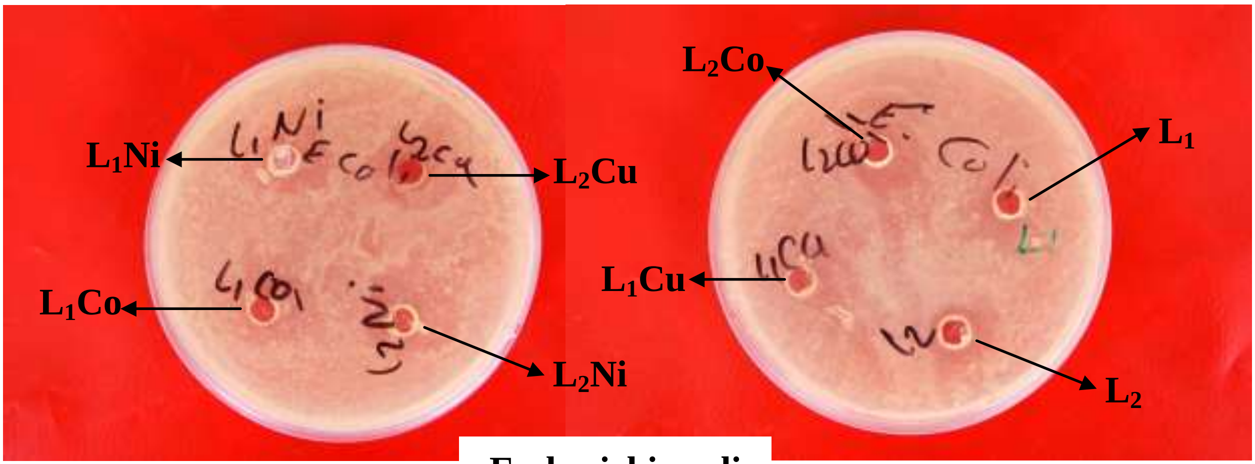
Such increased activity of the metal chelates can be explained on the basis of overtone’s concept of cell permeability the lipid membrance that surrounds the cell favours the passage of only lipid soluble materiala due to which liposolubility is an important factor that controls antimicrobial activity. On chelation, the polarity of the metal ion is reduced to a greater extent due to the overlap of the ligand orbital and partial sharing of the positive charge of the metal ion with donor group. Further, it is increase the delocalization of electron over the whole chelate ring and enhances the lipophilicity of the complex. This increased lipophilicity enhances the penetration of the complexes into lipid membranes and blocking of metal binding sites on the enzymes of the microorganism (24).

Table 3: The formulas, colors, formula weight, yields%, melting points and molar conductance of the ligands L_1 and L_2 and their complexes

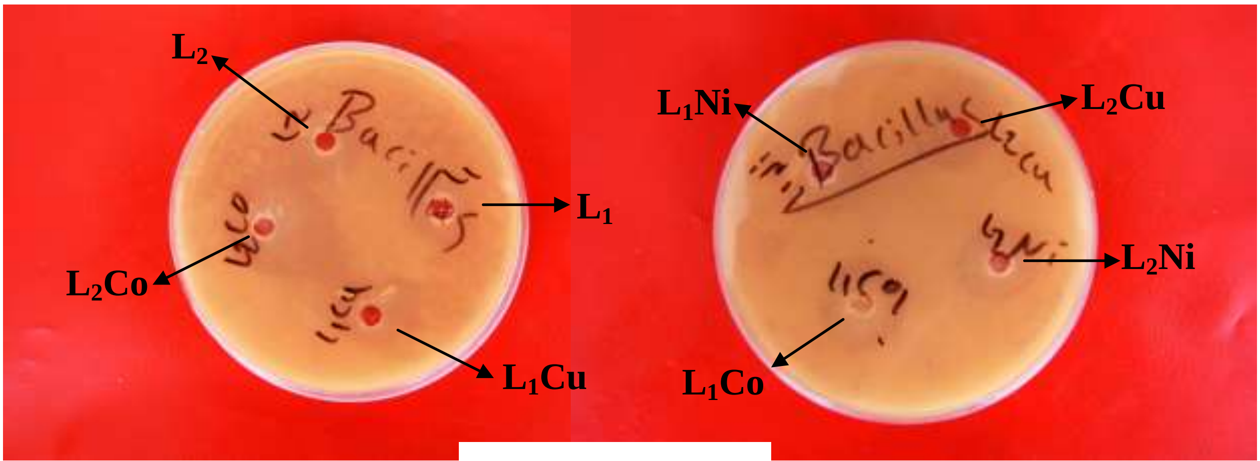
Compounds	Molecular formula	Colour	Formula weight gm / mole	Yeilds %	Mp (C º)	Λ_m $Ohm^{-1}cm^{2}mol^{-1}$
L_1	$C_{27}H_{22}N_2O_2$	Dark yellow	406.5	85	197-198	----------
$[Cu(L_1)]_2Cl_4$	$Cu_2C_{54}H_{44}N_4O_4Cl_4$	Pale brown	1082.1	72	300>	83
$[Ni(L_1)]_2(CH_3COO)_2$	$Ni_2C_{54}H_{44}N_4O_4Cl_4$	greenish-yellow	1166.4	70	300>	62
$[Co(L_1)]_2Cl_4$	$Co_2C_{62}H_{56}N_4O_{12}$	Pale - yellow	1072.8	69	300>	38
L_2	$C_{26}H_{20}N_2O_3$	Pale yellow	408.5	81	201-202	----------
$[Cu(L_2)]_2Cl_4$	$Cu_2C_{52}H_{40}N_4O_6Cl_4$	brown	1085.9	78	300>	71
$[Ni(L_2)]_2(CH_3COO)_2$	$Ni_2C_{60}H_{52}N_4O_{14}$	Pale - green	1170.3	83	300>	54
$[Co(L_2)]_2Cl_4$	$Co_2C_{52}H_{40}N_4O_6Cl_4$	greenish-yellow	1076.7	76	300>	50

Table 4: Antimicrobial activity data of Schiff bases L_1 and L_2 and their complexes

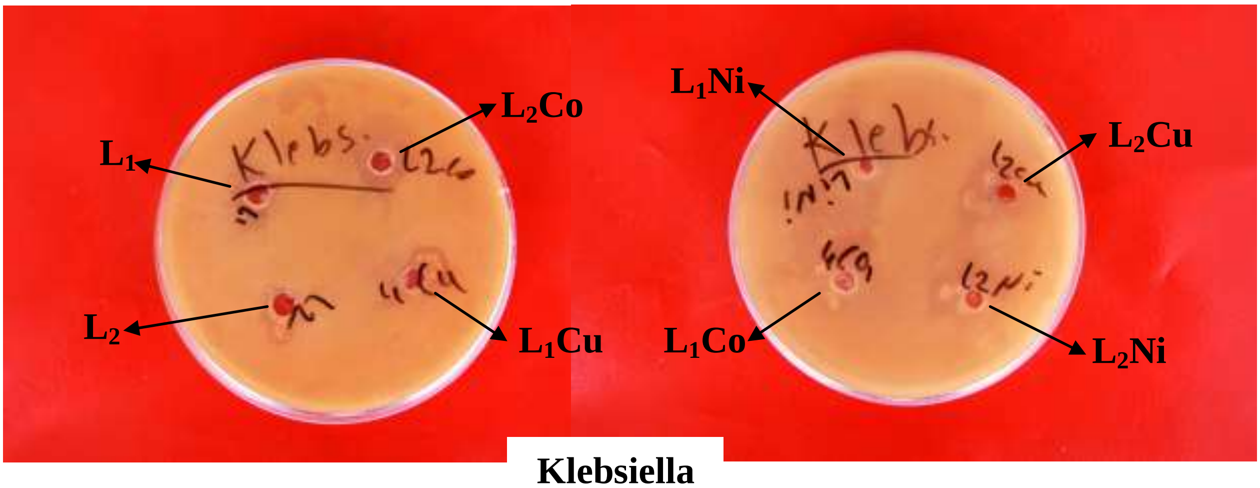
Compounds	Bacteria with zone inhibition (mm)		
	Escherichia coli	Bacillus subtilis	Klebsiella
L_1	9	11	8
L_1Cu	13	22	19
L_1Co	16	22	18
L_1Ni	16	18	17
L_2	12	16	11
L_2Cu	22	19	14
L_2Co	17	32	22
L_2Ni	21	22	18



Escherichia coli



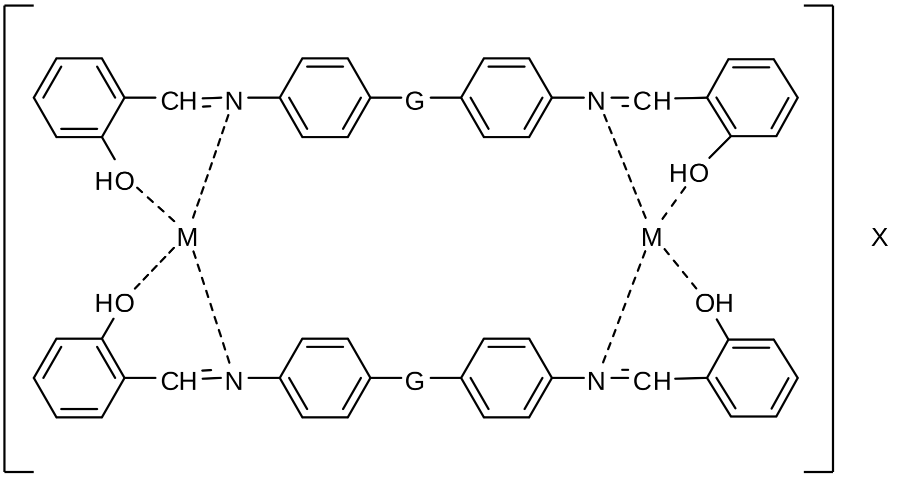
Bacillus subtilis



Klebsiella

Figure 5: Antimicrobacterium activities

From the IR spectra, it is concluded that the Schiff bases **L_1** and **L_2** ligands behaves as a neutral tetra dentate ligand coordinated to the metal ions by nitrogen atom in azomethine group(–CH=N–) and oxygen atom in phenol group (O–H). We concluded from the values of molar conductance that these complexes have ionic structure. It is possible to suggest the geometrical structure for the synthesized complexes between Schiff bases **(L_1 and L_2)** with Ni (II), Co (II), and Cu (II) ions, as shown below:



Where:
G = $-CH_2-$, or –O-
M = Cu (II), Co (II) X = Cl_4
M = Ni (II) X = $(CH_3COO)_2$

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