

## EVALUATION OF ANTIMICROBIAL ACTIVITIES OF MICROWAVE-IRRADIATION SYNTHESIZED TETRADENTATE (N<sub>2</sub>O<sub>2</sub> DONOR) SCHIFF BASE AND ITS Cu(II) COMPLEXES

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### ABSTRACT

The microwave-promoted synthetic method was used to synthesize the dibasic (N<sub>2</sub>O<sub>2</sub> donor) tetradentate unsymmetrical Schiff base [N,N-bis-(o-hydroxy acetophenone)-o-phenylenediimine as primary ligand] and its eight Cu(II) complexes with different monodentate secondary ligands in green ethanol aqueous solvent. Analytical analysis, conductivity measurement, magnetic moment determination and several spectroscopic techniques characterized all the mononuclear and non-electrolytic complexes synthesized through the environmentally benign method. The present study ascertained that the investigated Cu(II) complexes have a distorted octahedral geometry. These complexes have greater antimicrobial activities than the free ligand toward the tested microorganisms as a result of the chelation effect.

**Keywords:** Microwave Irradiation, Copper Complexes, Distorted Octahedral Geometry, Antimicrobial Activity.

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### INTRODUCTION

Recently the synthetic chemistry is at the cutting edge of sustainable development using safe and clean synthetic processes, products and yields.<sup>1-2</sup> Green chemistry or environmentally benign chemistry is means of using basic science to address environmental issues in an economically expedient manner and a special contribution of chemists to the conditions for sustainable development.<sup>3-4</sup> The environmentally benign methodologies are now used in the chemical industry for sustainable development.<sup>5-6</sup> It is found that all environmentally benign methodologies of synthesis of chemicals are less noxious to human health and the environment. The microwave-promoted synthetic approaches using green solvents or dry medium process for the synthesis of metal coordination compounds or complexes are the very significant eco-friendly methods.<sup>7-8</sup>

On screening the literature survey, Schiff bases (SBs), i.e., azomethine compounds have very good complexing capacity towards transition metal ions, and their complexes have innumerable applications in various fields of chemical sciences like biochemical, pharmaceutical, analytical and industrial besides their significant roles in catalysis and organic/inorganic syntheses.<sup>9</sup> These facts actuate the inorganic or coordination chemists to develop environmentally benign methodologies for the synthesis of new Schiff bases and their transition metal complexes having various therapeutic activities.<sup>10-12</sup> Literature survey revealed that unsymmetrical tetradentate Schiff bases derived as the products of condensation of different diamines with different carbonyl compounds (aldehydes and ketones) and their metal complexes have been studied very less and more attention is needed to their studies at present for their chemotherapeutic usages.<sup>13-15</sup>

Microwave irradiated synthetic reactions are a facile, efficient, and environmentally benign synthetic approach for the synthesis of several new Schiff base ligands and their transition metal compounds.<sup>16</sup> Further, copper, the third most abundant essential metallic element, has a biological role in sustaining life and has anti-microbial activity and proper cellular functions.<sup>17</sup> The comprehensive studies revealed that the Schiff bases i.e., azomethine compounds and their metal complexes have fascinating structural and extensive biological applications.

These remarkable activities induced us to utilize the eco-friendly synthetic methodology like microwave irradiation (MWI) in the synthesis of new Schiff base and its Cu(II) complexes. As the part of our continuous research works<sup>18-20</sup> on unsymmetrical tetradentate Schiff base ligands containing N and O donor atoms and considering the significance of Cu<sup>2+</sup> ion, we now report the microwave promoted synthesis and characterization of eight divalent copper complexes of a new salen-type (ONNO donor) unsymmetrical dibasic tetradentate Schiff base [N,N-bis-(*o*-hydroxy acetophenone)-*o*-phenylenediimine] as a primary ligand and several monodentate secondary ligands in this research article which also describes the comparison between conventional synthetic method and environmentally benign synthetic method using aqueous-alcoholic green solvent as green chemistry protocol. Further, the article also presents the evaluation of antibacterial and antifungal activities of the synthesized unsymmetrical Schiff base ligand and its divalent copper complexes against different pathogenic bacteria and fungi.

## EXPERIMENTAL

### Materials and Methods

During the present research project, the high purity analytical or spectroscopic grade reagents/chemical and solvents were used. These chemicals were purchased from Aldrich-Sigma Ltd and used without further purification in these green synthetic methods. Microwave-assisted synthesized ligand characterized as reported earlier.<sup>19</sup>

A melting point apparatus (Electro-thermal 9100) was used to determine the melting points of all eco-friendly synthesized compounds and the melting points were uncorrected. The conductivities of the synthesized complexes at room temperature were determined by using DMF bridge model PW 9501 having Philips PW 9515/10 conductivity cell. Using mercuric tetrathiocyanatocobaltate (II), Hg[Co(SCN)<sub>4</sub>] { $\chi_g = 16.44 \times 10^{-6}$ } as a calibrant, Gouy's method of magnetic susceptibility was used to ascertain the magnetic moments of the bivalent copper complexes at room temperature.<sup>21</sup> The micro-analytical elemental analyzer namely Carlo-Ebra EA1110 CHNO-S was used for elemental analysis of the undertaken compounds. The Shimadzu UV-Visible spectrophotometer (model UV-1900i) spectrophotometer was used to scan the electronic spectra of the investigated compounds, while the Shimadzu Infrared (model IR Affinity-IS) spectrophotometer was used to record the infrared absorption spectra of the compounds as KBr discs between 4000–400 cm<sup>-1</sup> wavenumber. The X-band ESR spectra of bivalent copper complexes in the DMF solution at room temperature were recorded by using Varian E-112 spectrometer. The Kneubuhl approximation was used to determine the *g* values of the Cu (II) complexes.<sup>22</sup>

An improved microwave oven (Model 2001 ETB, power source 230V, output energy 800W and 2459 MHz frequency, Bajaj Electricals Ltd make) fitted with a rotating tray was used to synthesize all the investigated compounds using the green chemistry (MWI) protocol. To control the temperatures of synthesizing reactions, the on/off cycling method was used. The thin-layer chromatography (TLC), having precoated silica gel (GF254) plates, monitored the progress of the microwave irradiated reactions. A common disc diffusion method<sup>23</sup> was used for the evaluation of the antibacterial and antifungal activities of the eco-friendly synthesized tetradentate (ONNO donor) ligand and its Cu (II) complexes. The experimental evaluation method used nutrient agar medium for antibacterial activity and spread dextrose agar medium for antifungal activity respectively.

### Microwave-Promoted Synthesis of Ligand (L<sub>2</sub>H)

A proposed Schiff base was prepared by refluxing a 1:2 molar mixture of aqueous-ethanolic solutions of *o*-phenylene diamine and *o*-hydroxy acetophenone at ambient temperature for about 5-6 minutes in the microwave oven. The obtained yellowish solid was filtered, washed with ethanol, dried at room temperature and recrystallized with ethanol to get the required research quality Schiff base (Scheme-1).

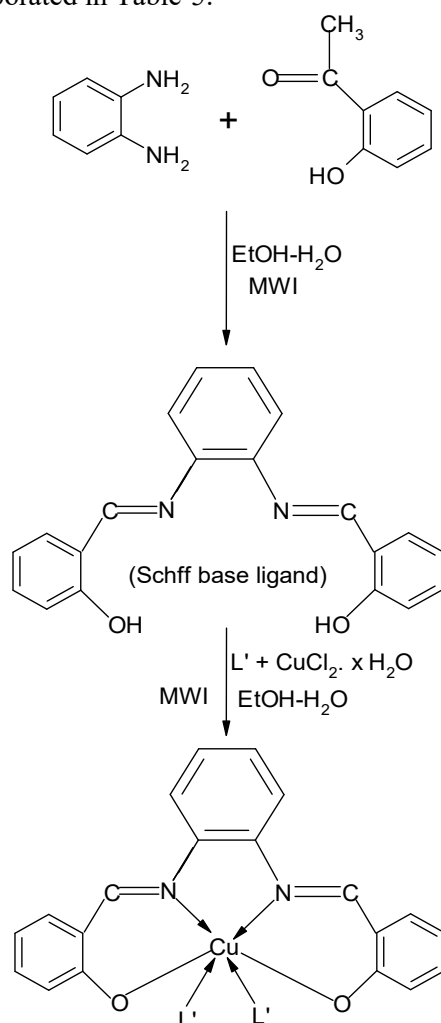
### Microwave-Promoted Synthesis of Cu (II) Complexes

A stoichiometric (1:2:1) molar mixture of a hot ethanolic solution of the synthesized Schiff base ligand (H<sub>2</sub>L), the secondary ligand (L') and an ethanolic solution CuCl<sub>2</sub>.2H<sub>2</sub>O was prepared to obtain the bivalent copper complexes. A few drops of triethylamine as catalyst were added to the reaction mixture, it was refluxed for 5-12 minutes in the microwave oven and then colored solids were obtained by filtration,

washing with ethanol and drying at room temperature. The complexes were obtained as colored solids on recrystallization with ethanol (Scheme-1).

### Antimicrobial Activity

The substantial antimicrobial activities (*in vitro*) of the investigated compounds were performed against four pathogenic bacterial strains and two fungal isolates. The bacterial strains were: two Gram-positive [*Staphylococcus aureus* (SA) and *Enterococcus faecalis* (EF)] and two Gram-negative [*Escherichia coli* (EC) and *Staphylococcus mutants* (SM)], but the fungal isolates were: *Candida albicans* (CA) and *Aspergillus niger* (AN). The synthesized green ligand and the bivalent copper complexes were dissolved separately in DMSO solvent (no inhibition activity) in a fixed concentration (1 µg per mL) per disc. We saturated the sterile discs with the solution of investigated compounds and then placed them in petri dishes containing culture medium (nutrient agar for bacterial strains and sabraoud dextrose sugar for fungal isolates). The petri dishes were incubated at 310 K and the zone of inhibition was measured after 24 hours for bacterial strains and after 48 hours for fungal isolates and compared with the standards (chloramphenicol for bacteria and griseofulvin for fungi). The results of antimicrobial activities of the undertaken compounds are incorporated in Table-5.



Scheme-1: Green synthesis of Schiff base and Cu(II) complexes using MWI

### RESULTS AND DISCUSSION

The reaction of 1:1:2 stoichiometric molar proportion of Schiff base (H<sub>2</sub>L), CuCl<sub>2</sub>·2H<sub>2</sub>O and monodentate ligand (L') in presence of triethylamine catalyst in an aqueous-ethanolic green solvent in a microwave oven resulted in the synthesis of [Cu L(L')<sub>2</sub>]. We observed that the synthetic reactions in MWI were completed in a relatively very short time (from 2-3 hours to 5-12 minutes) with higher yields (from 35-65

to 65-79%) compared to the conventional thermal method. Several physico-analytical and spectroscopic methods are used to characterize the environmentally benign synthesized bivalent copper complexes of the undertaken tetradentate Schiff base.

The present study revealed that all of the investigated compounds are non-hygroscopic, quite stable in air, and colored microcrystalline solids that do not have sharp melting points and decompose on heating at higher temperatures than the respective melting point range. All the synthesized compounds are commonly insoluble in water and common organic solvents. But these are readily soluble in DMF, DMSO, THF, acetonitrile and partially in hot ethanol. Table-1 comprises the results of elemental analysis and the physical properties like melting points, yields and molar conductance of the undertaken compounds. This table also presents the comparison between the results of the conventional method and the green synthetic approach related to this project.

The physico-analytical data shows that the undertaken coordination complexes correspond to  $[ML(L')_2]$  stoichiometry and mono-nuclear nature, where  $M = Cu^{2+}$ ,  $L =$  Schiff base ligand and  $L' =$  different monodentate secondary ligands. The very low values of experimental molar conductance (18.0 to 24.0  $ohm^{-1} cm^2 mol^{-1}$ ) of the complexes in the DMF ( $10^{-3}M$ ) solution indicate their non-electrolyte nature.<sup>24</sup>

Table-1: Comparison of the Conventional Method with MWI Synthetic Method and Physico-analytical Data of investigated Compounds

Compound (Color)	Reaction Time CM (MWI)	Yield (%) CM (MWI)	Mol. Mass	Melting Point (in K)	Elemental Analysis Calculated (Found)%				Molar Conductance ( $ohm^{-1} cm^2$ $mol^{-1}$ )
					C	H	N	Cu	
$C_{22}H_{20}N_2O_2$ ( $H_2L$ ) Pale Yellow	2 h. (6 min.)	65 (77)	344	521	76.74 (76.70)	5.81 (5.80)	8.13 (8.10)	--	20
$[CuL(H_2O)_2]$ Green	2.4 h. (5 min.)	60 (75)	445.45	561	59.79 (59.74)	4.98 (5.00)	6.34 (6.40)	14.38 (14.30)	22
$[CuL(NH_3)_2]$ Bright Green	2.5 h. (6 min.)	55 (77)	439.45	567	60.06 (59.98)	5.46 (5.50)	12.74 (12.70)	14.44 (14.40)	24
$[CuL(Py)_2]$ light green	2.5 h. (8 min.)	47 (79)	591.45	610	68.14 (68.20)	4.96 (4.90)	5.93 (5.90)	11.26 (11.20)	16
$[CuL(Q)_2]$ Deep Green	2.5 h. (10 min.)	45 (75)	563.45	589	72.39 (72.42)	4.82 (4.80)	8.44 (8.40)	9.75 (9.80)	18
$[CuL(PhI)_2]$ faint green	2.7 h. (9 min.)	52 (78)	611.45	594	70.64 (70.60)	4.57 (4.50)	9.15 (9.10)	10.38 (10.40)	20
$[CuL(\alpha-Pc)_2]$ Green	2.6 h. (12 min.)	35 (65)	519.45	584	68.57 (68.50)	5.40 (5.35)	9.46 (9.40)	10.70 (10.65)	19
$[CuL(\beta-Pc)_2]$ Light Green	2.6 h. (11 min.)	36 (67)	519.45	605	68.57 (68.50)	5.40 (5.35)	9.46 (9.40)	10.70 (10.65)	21
$[CuL(\lambda-Pc)_2]$ Deep Green	2.6 h. (10 min.)	35 (66)	519.45	595	68.57 (68.50)	5.40 (5.35)	9.46 (9.40)	10.70 (10.65)	20

CM = Conventional Method; MWI = Microwave Irradiation; L = Ligand ( $C_{22}H_{18}N_2O_2$ ); Py = Pyridine; Q = Quinoline; PhI = Phenyl isocyanide; Pc = Picoline

### Infrared Spectral Studies

To determine the coordination mode of ligands in the bivalent copper complexes, the infrared spectra of the Schiff base ligand and secondary ligands were compared with that of investigated metal complexes. Table-2 summarized the absorption bands of the Schiff base ligand and other secondary ligands and eight bivalent copper complexes. The appearance of the absorption band in infrared spectra of ligand ( $H_2L$ ) at  $1630 cm^{-1}$  and the characteristic ketonic carbonyl group ( $>C=O$ ) stretching band near  $1750 cm^{-1}$  in the infrared spectra of precursor o-hydroxy acetophenone, concluded the formation of Schiff base ligand.<sup>25</sup>

Appreciably varied frequencies shown because of the vibrations of different groups, e.g., -OH, C=N, C-N, etc., in the studied Schiff base ligand and secondary monodentate ligands during the complexation shown in the infrared spectra of investigated bivalent copper complexes. The strong band at 1610-1630  $\text{cm}^{-1}$  corresponding to the azomethine  $\nu(>\text{C}=\text{N}-)$  group ( $>\text{C}=\text{N}-$ ) of the free ligand ( $\text{H}_2\text{L}$ ) changes to lower wave numbers (bathochromic shift) by 20-35  $\text{cm}^{-1}$ , therefore the N atom of the azomethine group is coordinating with the  $\text{Cu}^{2+}$  ion in all complexes. This coordination mode was further confirmed by the presence of a band at 410-425  $\text{cm}^{-1}$  in complexes assigned to  $\nu(\text{M}-\text{N})$  vibrations. This revealed the involvement of the azomethine linkage in coordination<sup>26</sup>. A broad band appears near 3360  $\text{cm}^{-1}$  assigned to the free  $\nu(\text{O}-\text{H})$  stretching mode in the Schiff base spectra, but the disappearance of this band in the spectra of  $\text{Cu}(\text{II})$  complexes shows the deprotonation of the ligand and supports the coordination of the phenolic O atom with the copper ion. The appearance of  $\nu(\text{C}-\text{O})$  phenolic (CO) at lower frequencies (compared to 1450-1460  $\text{cm}^{-1}$  in the free ligand) in the range 1420-1430  $\text{cm}^{-1}$ , after complexation with  $\text{Cu}^{2+}$  ion in complexes.<sup>27</sup> This coordination mode was further supported by the appearance of a band at 450-470  $\text{cm}^{-1}$  in complexes assigned to  $\nu(\text{M}-\text{O})$  vibrations. Four coordination sites of the  $\text{Cu}^{2+}$  ion are fulfilled by two donor N atoms and two O atoms in the investigated complexes. Finally, a comparison of the IR spectra of the ligand ( $\text{H}_2\text{L}$ ) and the investigated complexes concludes the unsymmetrical nature of the dibasic tetradentate (ONNO donor). Since the (M-O) bond is more ionic than the (M-N) bond hence the frequency obtained due to the vibrations of (M-O) bond is greater than that of (M-N) bond.<sup>28</sup> An absorption band near 750  $\text{cm}^{-1}$  is attributed because of the rocking mode of coordinating water molecule with  $\text{Cu}^{2+}$  ion in the studied aqua complex, but no separate band appeared for coordinated water due to overlap of  $\nu(\text{O}-\text{H})$  and  $\nu(\text{N}-\text{H})$  vibrations.<sup>29</sup> The investigated amino complex of  $\text{Cu}^{2+}$  consists of one broad and strong band around 3460  $\text{cm}^{-1}$  assignable to  $\nu(\text{N}-\text{H})$  stretching vibration of coordinated  $\text{NH}_3$ . The coordination of the isocyanide group ( $-\text{N}=\text{C}$ ) in the investigated complex was confirmed by an increase in the  $\nu(\text{N}=\text{C})$  vibration of the isocyanide ligand from 2170  $\text{cm}^{-1}$  to 2220  $\text{cm}^{-1}$  in the phenyl isocyanide complex. The  $\text{Cu}(\text{II})$  complex with quinoline as secondary ligand exhibits a medium and broadband near 1680  $\text{cm}^{-1}$  because of the ring vibration showing the involvement of nitrogen atom of quinoline ring in the complexation. The absorption bands of the pyridine and picoline as secondary ligands are displayed in the fingerprint and far-infrared regions respectively.<sup>30</sup> The characteristic vibrations of the coordinated pyridine ligand appear as absorption bands near 1000-1100  $\text{cm}^{-1}$  in the investigated complex. The studied  $\text{Cu}(\text{II})$  complexes with Schiff base and different picoline molecules display infrared absorption bands at 790, 770 and 750  $\text{cm}^{-1}$  for  $\alpha$ -picoline,  $\beta$ -picoline and  $\gamma$ -picoline respectively.

Table-2: Observed IR Bands ( $\text{cm}^{-1}$ ) of the Ligand ( $\text{H}_2\text{L}$ ) and  $\text{Cu}(\text{II})$  Complexes

IR Bands ( $\text{cm}^{-1}$ ) OF $\text{H}_2\text{L}$	IR Bands of $\text{Cu}(\text{II})$ Complexes	Probable Assignment
--	3480	$\nu(\text{N}-\text{H})$
3360	--	$\nu(\text{O}-\text{H})$
1630	1580	$\nu(\text{C}=\text{N})$
1420	1450	$\nu(\text{O}-\text{H})_{\text{phenolic}}$
	1680	$\nu(\text{C}=\text{N})_{\text{quinoline}}$
	2220	$\nu(\text{NC})$
	1050	Due to pyridine ring
	790	$\nu(\alpha\text{-picoline})$
	750	$\nu(\beta\text{-picoline})$
	760	$\nu(\gamma\text{-picoline})$
	450-470	$\nu(\text{M}-\text{O})$
	410-425	$\nu(\text{M}-\text{N})$

### Electronic Spectral Studies

The electronic spectral study (UV-visible spectroscopy) is an important tool for coordination chemists to predict the plausible structure or geometry of the coordination compounds. The electronic spectral data,

magnetic moments and proposed geometry of the investigated complexes of the Cu(II) are incorporated in Table-3.

The electronic spectrum of the Schiff base ligand consists of an intense absorption band near 295 nm and another near 375 nm corresponding to  $\pi \rightarrow \pi^*$  and  $n \rightarrow \pi^*$  transitions that originated for the phenyl ring and the imine chromophore, respectively. The disappearance of the band of  $n \rightarrow \pi^*$  transition in the spectra of Cu (II) complexes confirms the coordination of the azomethine N atom with the central  $\text{Cu}^{2+}$  ion<sup>31-32</sup>. Further, the appearance of a low energy absorption band near 655-670 nm in the electronic spectra of the investigated complexes shows the d-d transition, which is consentaneous with the distorted octahedral geometry of the studied complexes.

Table-3: Electronic Spectral Bands, Magnetic Moments, and the proposed Geometry of the Cu (II) Complexes

Compound	$\lambda_{\text{max}}$ in nm		Magnetic Moments(BM)	Proposed Geometry
	Ligand Bands	d-d Bands		
$\text{C}_{22}\text{H}_{20}\text{N}_2\text{O}_2$ ( $\text{H}_2\text{L}$ )	295, 375	--	--	--
$[\text{CuL}(\text{H}_2\text{O})_2]$	295	655	1.96	Distorted Octahedral Geometry
$[\text{CuL}(\text{NH}_3)_2]$	295	665	1.92	Distorted Octahedral Geometry
$[\text{CuL}(\text{Py})_2]$	295	668	1.94	Distorted Octahedral Geometry
$[\text{CuL}(\text{Q})_2]$	295	675	1.90	Distorted Octahedral Geometry
$[\text{CuL}(\text{PhI})_2]$	295	672	1.82	Distorted Octahedral Geometry
$[\text{CuL}(\alpha\text{-Pc})_2]$	295	670	1.86	Distorted Octahedral Geometry
$[\text{CuL}(\beta\text{-Pc})_2]$	295	670	1.92	Distorted Octahedral Geometry
$[\text{CuL}(\lambda\text{-Pc})_2]$	295	670	1.88	Distorted Octahedral Geometry

### Magnetic Moment Studies

Further, Table-3 also comprises the experimentally determined magnetic moment values of the Cu(II) complexes at room temperature. The observed values of the magnetic moments (1.82 to 1.96 BM) of the complexes suggest their mononuclear spin-free distorted octahedral geometry.<sup>33</sup>

### ESR Studies

The information about the environment of the metal ion within the complexes can be ascertained by the study of the scanned polycrystalline state X-band ESR spectra of the complexes at room temperature without fine or hyperfine structure. The EPR parameters of the investigated Cu(II) complexes are presented in Table-4.

Table-4: EPR Parameters of the investigated Cu (II) Complexes

Compound	g Tensor Values			
	$g_{11}$	$g_{\perp}$	$g_{\text{av}}$	G
$[\text{CuL}(\text{H}_2\text{O})_2]$	2.300	2.021	2.110	2.68
$[\text{CuL}(\text{NH}_3)_2]$	2.240	2.024	2.082	3.05
$[\text{CuL}(\text{Py})_2]$	2.224	2.023	2.051	2.98
$[\text{CuL}(\text{Q})_2]$	2.232	2.023	2.024	2.84
$[\text{CuL}(\text{PhI})_2]$	2.214	2.026	2.028	3.09
$[\text{CuL}(\alpha\text{-Pc})_2]$	2.228	2.025	2.019	2.98
$[\text{CuL}(\beta\text{-Pc})_2]$	2.226	2.026	2.020	3.00
$[\text{CuL}(\lambda\text{-Pc})_2]$	2.227	2.027	2.011	3.02

The EPR parameters [ $g_{11} > g_{\perp} > 2.0023$ ] of the investigated Cu(II) complexes suggest that the ninth unpaired electron is in the orbital  $d_{x^2-y^2}$  (energy order:  $d_{x^2-y^2} > d_z^2$ ) orbital which is inconsistent with the

distorted octahedral geometry<sup>34</sup> due to rhombic distortion from the axial symmetry and covalent nature of the bonding between the  $\text{Cu}^{2+}$  ion and the ligand molecules.<sup>34,35</sup> Furthermore, it reveals that the 5<sup>th</sup> and 6<sup>th</sup> positions of the complexes are occupied by the same two monodentate ligands in the distorted octahedral structures.

### Proposed Structure of the Cu(II) Complexes

Based on the aforesaid experimental evidence, we tentatively suggest the distorted octahedral structure of the investigated complexes in which the macrocyclic Schiff base ligand acts as a dibasic tetradentate (ONNO donor) primary ligand. Figure-1 presents a common tentative geometry/structure of the complexes.

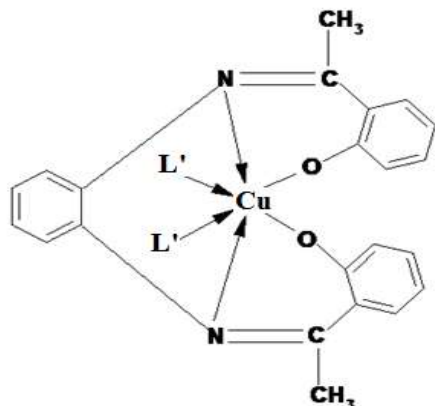


Fig.-1: Proposed distorted Octahedral Structure of Cu(II) Complexes

### Antimicrobial Activities

The experimental results of the antibacterial and antifungal activities of the investigated Schiff base ligand and the Cu(II) complexes are presented in Table-5.

Table-5: Antimicrobial Activity of the Schiff Base Ligand and Its Cu(II) Complexes

Compound	Bacteria				Fungi	
	Gram-positive		Gram-negative		CA	AN
	SA	EF	EC	SM		
$\text{C}_{22}\text{H}_{20}\text{N}_2\text{O}_2$	13	11	13	12	19	20
$[\text{CuL}(\text{H}_2\text{O})_2]$	9.3	6.5	10	7.4	16	13
$[\text{CuL}(\text{NH}_3)_2]$	7	7.3	12	9	12	16
$[\text{CuL}(\text{Py})_2]$	10	7	16	8	13	19
$[\text{CuL}(\text{Q})_2]$	4.9	6.5	5.4	7.6	9	14
$[\text{CuL}(\text{PhI})_2]$	6.5	5.6	7.3	6.7	11	12
$[\text{CuL}(\alpha\text{-Pc})_2]$	7.4	5.6	7.4	8.7	9	12
$[\text{CuL}(\beta\text{-Pc})_2]$	5.3	7.4	8.6	8.4	10	13
$[\text{CuL}(\gamma\text{-Pc})_2]$	6.5	7.9	6.7	5.3	9	12
Chloramphenicol	0.25	1.5	2.3	1.7	3.2	3.5
Griseofulvin	--	--	--	--	8	11

The experimental results revealed the moderate antimicrobial activities of the complexes which are quite anticipating and up to expectation. The bivalent copper complexes have slightly higher antimicrobial activities than the free Schiff base ligand; although the investigated compounds displayed poor activity than the used reference. We can explain the enhanced antimicrobial activities of copper complexes on the basis of Overtone's concept and Tweedy's chelation theory. The enhanced lipophilicity of the complexes is due to chelation due to the delocalization of  $\pi$ -electron over the whole chelate ring that controls their antimicrobial activities.<sup>36,37</sup>

### CONCLUSION

For the successful and efficient synthesis of unsymmetrical dibasic Schiff base (ONNO donor) tetradentate Schiff N, N'-bis (o-hydroxy acetophenone) -o-phenylenedimine, a primary ligand and its

eight Cu(II) complexes with some monodentate secondary ligands, we used an environmentally benign microwave irradiation protocol in a green solvent medium. The non-electrolyte and mononuclear nature of investigated complexes are determined by elemental analysis, molar conductance and magnetic moment measurements. The electronic, infrared, and ESR spectral observations characterized the dibasic tetradentate (ONNO donor) nature of the Schiff base ligand and distorted octahedral geometry of the complexes. The antimicrobial data suggests the moderate antimicrobial activities of complexes with greater activity than free ligand owing to enhanced lipophilicity by chelation.

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