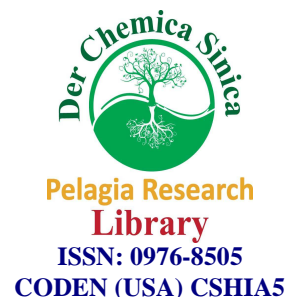




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### Synthesis, characterization and biocidal studies of Ni(II), Cu(II), Co(II), Fe(II), Mn(II) and Zn(II) complexes Containing tetra dentate and neutral bi dentate schiff base

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#### ABSTRACT

Tetra dentate schiff base *N,N'*-bis[2-hydroxyacetophenone]ethylenediamine (OAcPh-en) were formed by the 2:1 molar condensation of 2-hydroxyacetophenone with ethylenediamine. Their Ni(II), Cu(II), Co(II), Mn(II), Fe(II) and Zn(II) diaquo complexes (I) were synthesized. The neutral bidentate derived from benzaldehyde and ethylenediamine or *o*-phenylenediamine in 1:1 molar ratio in alcoholic solution yield bis[benzylidene]ethylenediamine (Ben-en) or bis[benzylidene]*o*-phenylenediamine (Ben-opd) (II). Further reaction between (I) and (II) in alcohol yielded mixed ligand complexes. The characterization of these newly synthesized mixed-ligand complexes were done by elemental analysis, magnetic measurements, Infrared spectra and uv-visible spectra, thermo gravimetric analysis, biocidal studies have also been under taken.

**Key words:** Schiff Bases, Mixed ligand complexes, Biocidal Activity.

#### INTRODUCTION

Ligand, a metal surrounded by a cluster of ions or molecule, is used for preparation of complex compounds named as Schiff bases [1]. Schiff bases have been widely used in many fields e.g., biological, inorganic, analytical and drug synthesis, as bidentate ligands in the field of coordination chemistry [2]. The chemistry of Schiff base metal complexes had been investigated extensively for the last several decades leading to new synthetic routes variety of structures and their potential biological applications [3-5]. Tetra dentate Schiff bases are well known for their coordination with various metal ions, forming stable compounds [6,7]. Many symmetrical bis tetradentate Schiff bases of 1, 2-diamines with *o*-hydroxy aldehydes/ketones have been prepared

and studied intensively. However, much less attention has been focused on unsymmetrical tetradentate Schiff bases derived from 1,2-diamines and different aldehydes/ketones. In particular those derived from aromatic 1,2-diamines have been under-investigated [6,8]. We prepared new mixed ligand complexes of the type  $[M(\text{OAcPh-en})(\text{Ben-en})]\text{H}_2\text{O}$  and  $[M(\text{OAcPh-en})(\text{Ben-opd})]\text{H}_2\text{O}$  where  $M = \text{Ni(II)}, \text{Cu(II)}, \text{Co(II)}, \text{Fe(II)}, \text{Mn(II)}$  and  $\text{Zn(II)}$ .  $\text{OAcPh-en} = \text{N,N}'\text{-bis[2-hydroxyacetophenone] ethylenediamine}$ ,  $\text{Ben-en} = \text{bis[benzaldehyde] ethylenediamine}$ ,  $\text{Ben-opd} = \text{bis[benzaldehyde]o-phenylenediamine}$ . The complexes were characterized on the basis of physical properties, elemental analysis, infrared and uv-visible spectra, thermo gravimetric analysis and antimicrobial activities.

## MATERIALS AND METHODS

### *Synthesis of N,N'-bis[2-hydroxyacetophenone]ethylenediamine (OAcPh-en):*

The ligand  $\text{OAcPh-en}$  was synthesized by refluxing an ethanolic solution of 2-hydroxyacetophenone ( $\text{OAcPh}$ ) and ethylenediamine ( $\text{en}$ ) (2:1 molar ratio) for one hour. The reaction mixture was cooled and a bright yellow crystalline solid of  $\text{N,N}'\text{-bis[2-hydroxyacetophenone]ethylenediamine}$  ( $\text{OAcPh-en}$ ) separated [10,11]. The solid was filtered off and recrystallized from ethanol to give bright yellow crystals.

### *Synthesis of bis[benzylidene]ethylenediamine (Ben-en):*

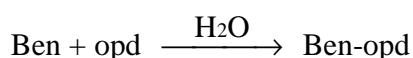
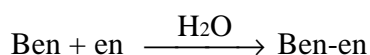
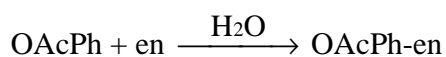
The ligand  $\text{Ben-en}$  was synthesized by refluxing an ethanoic solution of benzaldehyde ( $\text{Ben}$ ) and ethylenediamine ( $\text{en}$ ) (2:1 molar ratio) for one hour. The reaction mixture was cooled and a bright yellow crystalline solid of  $\text{bis[benzylidene]ethylenediamine}$  ( $\text{Ben-en}$ ) separated. The solid was filtered off and recrystallized from ethanol to give light yellow crystals.

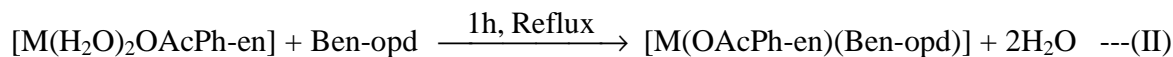
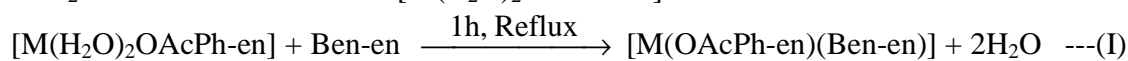
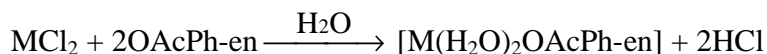
### *Synthesis of bis[benzylidene]o-phenylenediamine (Ben-opd):*

The ligand  $\text{Ben-opd}$  was synthesized by refluxing an ethanoic solution of benzaldehyde ( $\text{Ben}$ ) and o-phenylenediamine ( $\text{opd}$ ) (2:1 molar ratio) for one hour. The reaction mixture was cooled and a bright yellow crystalline solid of  $\text{bis[benzylidene]o-phenylenediamine}$  ( $\text{Ben-opd}$ ) separated. The solid was filtered off and recrystallized from ethanol to give yellow crystals.

### *Preparation of the complexes:*

The preparation of  $[\text{Ni}(\text{OAcPh-en})(\text{Ben-en})]\text{H}_2\text{O}$  was carried out by refluxing an ethanoic solution (250ml) of Nickel diaquo complex (0.01M) with Neutral bi dentate ligand  $\text{bis[benzylidene]ethylenediamine}$  ( $\text{Ben-en}$ ) (0.01M) for one hour. The solution was then concentrated and cooled in air to overnight. The formed crystals were collected and recrystallized, dry in air. The metal complexes of  $\text{Cu(II)}, \text{Co(II)}, \text{Fe(II)}, \text{Mn(II)}$ , and  $\text{Zn(II)}$  were prepared similarly. The mixed ligand complexes of  $[\text{M}(\text{OAcPh-en})(\text{Ben-opd})]\text{H}_2\text{O}$  [ $M = \text{Ni(II)}, \text{Cu(II)}, \text{Co(II)}, \text{Fe(II)}, \text{Mn(II)}$  and  $\text{Zn(II)}$ ] were also prepared similarly. The formation of the complexes may be represented by the following equations.





### Analytical procedures:

The complexes were analyzed for the metal contents by the EDTA-titration technique [11] after decomposing the organic matter first with a mixture of perchloric, sulphuric and nitric acid (1:1.5:2.5). The infrared spectra were recorded on spectrum GX FT-IR (Perkin Elmer, USA). The magnetic susceptibility of the chelates was determined by the Gouy method at room temperature. The uv-visible spectra were recorded on Lambda 19 (Perkin Elmer, USA). The thermo gravimetric analyses were recorded on Thermo gravimetric Analyzer (Perkin Elmer, USA). Elemental analyses were performed on a C, H, N analyzer (Perkin Elmer, USA). All melting points were recorded in open capillaries in a capillary melting point apparatus.

## RESULTS AND DISCUSSION

### Characterization of the complexes:

The given formulae (Table-1) are based on analytical data.

**Table-1: Analytical data of the complexes**

Complex	Formula Weight g/mole	Colour	M.P. °C	% Found. (Calc.)				% Yield	$\mu_{\text{eff}}$ (BM)
				C	H	N	M		
[Mn(OAcPh-en)(Ben-en)]H <sub>2</sub> O	584.94	Brown	>360	69.68 (69.74)	5.89 (5.81)	9.67 (9.57)	9.33 (9.39)	76.8	5.74
[Fe(OAcPh-en)(Ben-en)]H <sub>2</sub> O	585.85	Brown	>360	69.69 (69.64)	5.78 (5.80)	9.63 (9.55)	9.48 (9.53)	74.8	5.25
[Co(OAcPh-en)(Ben-en)]H <sub>2</sub> O	588.93	Yellowish Brown	284	69.33 (69.27)	5.82 (5.77)	9.42 (9.50)	10.09 (10.0)	70.4	4.00
[Ni(OAcPh-en)(Ben-en)]H <sub>2</sub> O	588.7	Yellowish Orange	288	69.24 (69.30)	5.82 (5.77)	9.40 (9.51)	10.03 (9.97)	71.7	2.71
[Cu(OAcPh-en)(Ben-en)]H <sub>2</sub> O	593.55	Yellowish Brown	296	68.82 (68.78)	5.66 (5.72)	9.52 (9.43)	10.62 (10.70)	73.3	1.80
[Zn(OAcPh-en)(Ben-en)]H <sub>2</sub> O	595.38	Yellow	290	68.47 (68.52)	5.78 (5.71)	9.48 (9.40)	10.91 (10.98)	80.1	---
[Mn(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	632.94	Brown	260	72.10 (72.04)	5.42 (5.37)	8.77 (8.84)	8.72 (8.68)	76.5	5.75
[Fe(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	633.85	Brown	>360	71.87 (71.94)	5.41 (5.36)	8.88 (8.83)	8.72 (8.81)	73.2	5.24
[Co(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	636.93	Brown	228	71.63 (71.59)	5.39 (5.33)	8.74 (8.79)	9.29 (9.25)	81.6	4.10
[Ni(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	636.7	Yellowish Orange	220	71.53 (71.61)	5.38 (5.34)	8.89 (8.79)	9.26 (9.21)	82.3	2.69
[Cu(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	641.55	Green	240	71.14 (71.07)	5.25 (5.29)	8.82 (8.72)	9.84 (9.90)	80.7	1.85
[Zn(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	643.38	Yellow	246	70.81 (70.87)	5.32 (5.28)	8.75 (8.70)	10.04 (10.16)	84.3	---

**Magnetic Measurements:**

The magnetic moment values are useful in the evaluation of results provided by the other techniques of the structural investigation [12]. The magnetic moment data are presented in table-1. The magnetic moment of the Cu(II) complex (1.80-1.85 B.M.) is very close to the spin-only value (1.73 B.M.) expected for the one unpaired electron which offers the possibility of an octahedral geometry [13-14]. The magnetic moment value for Fe(II) complex is 5.25-5.24 B.M. at room temperature and this value is characteristic of high spin octahedral geometry for this complex [14]. The magnetic moment values for Co(II) complexes are 4.00-4.10 B.M. corresponding to three unpaired electrons which suggests an octahedral geometry [15-16]. The magnetic moment of the nickel complex at room temperature was observed 2.71-2.69 B.M. These values are in tune with a high spin configuration and show the presence of an octahedral environment around the Ni(II) ion in the complex [13,17]. The Mn(II) complex show magnetic moments is 5.74-5.75 B.M. at room temperature corresponding to five unpaired electrons which suggest octahedral geometry [18]. Zn(II) complex is show diamagnetic nature and is found to be octahedral geometry [15, 19-21].

**IR Spectra:**

The infrared spectra of the complexes have been studied to characterize their structures. The IR spectra of the complexes register  $\nu(\text{C-O})$  at about  $1324\text{-}1340\text{ cm}^{-1}$  [22-23]. The IR spectra of Schiff base shows a sharp band near  $1610\text{ cm}^{-1}$  which may be due to azomethine linkage and shows lowering in frequency in metal complex indicating the coordination of metal ion through azomethine linkage [24-26]. The sharp bend in the range  $750\text{-}780\text{ cm}^{-1}$  and  $1525\text{-}1535\text{ cm}^{-1}$  are due to aromatic  $\nu(\text{C-H})$  [22,27] and  $\nu(\text{C=C})$  [22,28] respectively. The frequencies in the range  $1145\text{-}1165\text{ cm}^{-1}$  attributed to  $\nu(\text{C-N})$  stretching [29]. Conclusive evidence of the bonding is also shown by the observation that new bonds in the spectra of the metal complexes appears at  $455\text{-}460\text{ cm}^{-1}$  and  $514\text{-}525\text{ cm}^{-1}$  these are assigned to  $\nu(\text{M-O})$  and  $\nu(\text{M-N})$  stretching vibrations and are not observed in the spectra of the ligand [30-32].

**Table – II Infrared spectra ( $\text{cm}^{-1}$ )**

Complex	$\nu(\text{C-O})$	$\nu(\text{C=N})$	$\nu(\text{C-N})$	$\nu(\text{C=C})$	$\nu(\text{M-O})$	$\nu(\text{M-N})$
OAcPh-en	1330	1610	1143	1507	----	----
Ben-en	1324	1618	1150	1515	----	----
Ben-opd	1326	1620	1148	1514	----	----
[Mn(OAcPh-en)(Ben-en)]H <sub>2</sub> O	1342	1599	1141	1532	435	520
[Fe(OAcPh-en)(Ben-en)]H <sub>2</sub> O	1336	1596	1138	1529	440	512
[Co(OAcPh-en)(Ben-en)]H <sub>2</sub> O	1333	1598	1140	1528	434	515
[Ni(OAcPh-en)(Ben-en)]H <sub>2</sub> O	1340	1600	1142	1530	444	508
[Cu(OAcPh-en)(Ben-en)]H <sub>2</sub> O	1338	1602	1139	1533	438	510
[Zn(OAcPh-en)(Ben-en)]H <sub>2</sub> O	1335	1599	1140	1531	440	517
[Mn(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	1341	1598	1141	1532	450	519
[Fe(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	1342	1598	1140	1528	454	512
[Co(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	1335	1600	1138	1530	445	525
[Ni(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	1336	1605	1142	1535	448	520
[Cu(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	1338	1599	1141	1534	450	516
[Zn(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	1345	1602	1143	1533	452	522

The presence of sharp band corresponding to the remaining hydroxyl group at  $3400\text{ cm}^{-1}$  but it is obscured by the presence of water molecules bands. This was appeared for the most complexes and a very broad band at about  $3100\text{-}3500\text{ cm}^{-1}$  region, which was associated with coordinated or solvent water molecules [33].

### Electronic spectra:

The absorption bands for the complexes will help to give an idea of their structure [34]. Electronic spectrum of Co(II) complex exhibits absorption bands at  $12,920$ ,  $16,260$ , and  $23,450\text{ cm}^{-1}$ , which may be assigned to  ${}^4\text{T}_1(\text{F}) \rightarrow {}^4\text{T}_2(\text{F})$ ,  ${}^4\text{T}_1(\text{F}) \rightarrow {}^4\text{A}_2(\text{F})$  and  ${}^4\text{T}_1(\text{F}) \rightarrow {}^4\text{T}_1(\text{P})$  transitions respectively which are in support of octahedral arrangement of binding centers around the metal ion [35]. The electronic spectra of Mn(II) complexes show the absorption bands in the range  $16970\text{-}19540$ ,  $22280\text{-}24390$  and  $26109\text{-}27624\text{ cm}^{-1}$ . These absorption bands may be assigned to the  ${}^6\text{A}_{1g} \rightarrow {}^4\text{A}_{1g}$  ( ${}^4\text{G}$ ),  ${}^6\text{A}_{1g} \rightarrow {}^4\text{A}_{2g}$  ( ${}^4\text{G}$ ), and  ${}^6\text{A}_{1g} \rightarrow {}^4\text{E}_g$ ,  ${}^4\text{A}_{1g}$  ( ${}^4\text{G}$ ) transitions, respectively. These bands suggest that the complexes possess an octahedral geometry [36]. Bands at the regions  $675\text{ nm}$  ( $14815\text{ cm}^{-1}$ ) and  $535\text{ nm}$  ( $18690\text{ cm}^{-1}$ ) for Cu(II) complex were assigned to the  ${}^2\text{B}_{1g} \rightarrow {}^2\text{B}_{2g}$  and  ${}^2\text{B}_{2g} \rightarrow {}^2\text{E}_g$  transitions respectively, found for octahedral Cu(II) complexes [37]. The electronic spectrum of the Fe(II) complex exhibit a band at  $11,200\text{ cm}^{-1}$ , assigned to the  ${}^5\text{T}_{2g} \rightarrow {}^5\text{E}_g$  transition. A strong charge transfer band is observed at  $26,000\text{ cm}^{-1}$ . These data suggest an octahedral geometry [38]. The electronic spectra of the Ni(II) complex showed d-d bands in the region  $10115$  and  $26280\text{-}26410\text{ cm}^{-1}$   ${}^3\text{A}_{2g}(\text{F}) \rightarrow {}^3\text{T}_{2g}(\text{F})$  and  ${}^3\text{A}_{2g}(\text{F}) \rightarrow {}^3\text{T}_{2g}(\text{P})$  respectively, consistent with their well-defined octahedral configuration [39]. The Zn(II) complex has not shown any d-d absorptions [40].

### Thermo gravimetric Analysis:

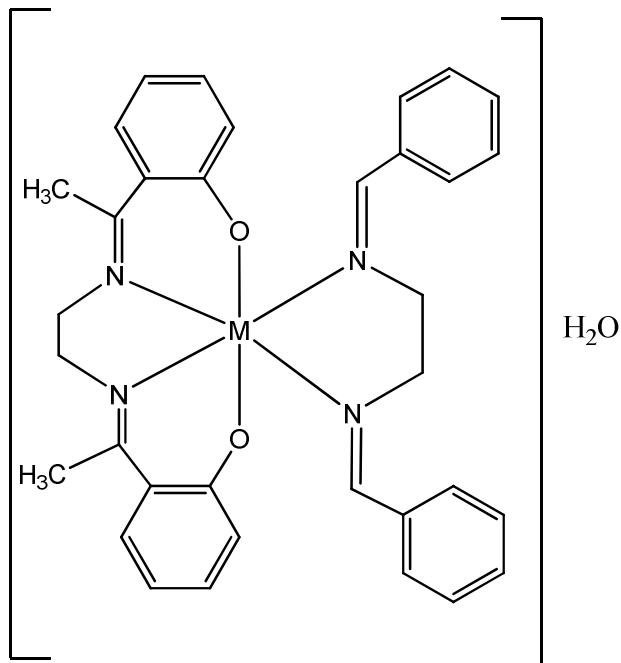
The analysis of the thermal curve of the complexes clearly indicated that the weight loss between  $35\text{-}105^\circ\text{C}$  corresponds to one water molecule for all complexes. Because of the low temperatures, this molecule may be consider as crystal water [41].

Table – III Antimicrobial activity

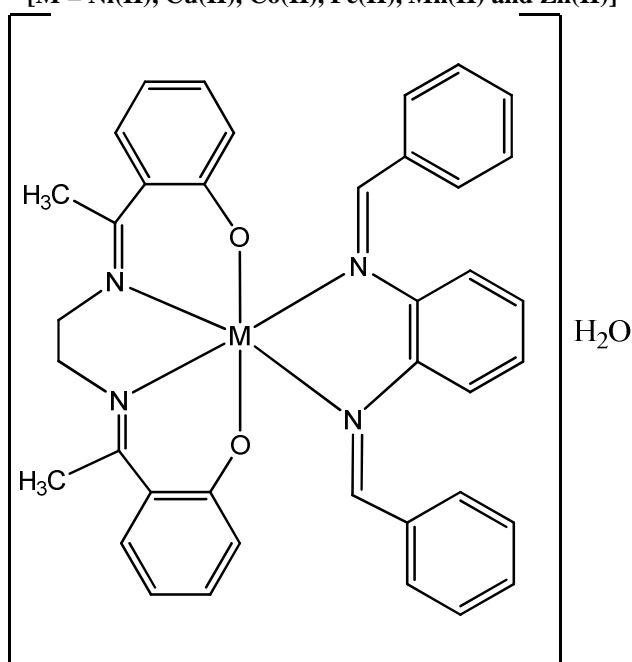
Complexes	Diameter of inhibition zone (mm)	
	<i>Bacillus Substlis</i>	<i>E.Coli</i>
[Mn(OAcPh-en)(Ben-en)]H <sub>2</sub> O	25	These metal complexes do not affect on <i>E.Coli</i> bacteria so there is no inhibition zone.
[Fe(OAcPh-en)(Ben-en)]H <sub>2</sub> O	24	
[Co(OAcPh-en)(Ben-en)]H <sub>2</sub> O	28	
[Ni(OAcPh-en)(Ben-en)]H <sub>2</sub> O	18	
[Cu(OAcPh-en)(Ben-en)]H <sub>2</sub> O	20	
[Zn(OAcPh-en)(Ben-en)]H <sub>2</sub> O	26	
[Mn(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	24	
[Fe(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	21	
[Co(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	24	
[Ni(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	17	
[Cu(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	18	
[Zn(OAcPh-en)(Ben-opd)]H <sub>2</sub> O	23	

The curves in the range between  $105\text{-}385^\circ\text{C}$  suggested that loss in weight for all complexes correspond to evaporation of bis[benzylidene]ethylenediamine (Ben-en) and bis[benzylidene]o-phenylenediamine (Ben-opd). The range above  $385^\circ\text{C}$  loss in weight correspond to remaining

organic ligand molecules. In all cases, final products are metal oxides. These results are in good accordance with the composition of the complexes.



**Figure 1:**  $[M(OAcPh-en)(Ben-en)]H_2O$   
[M = Ni(II), Cu(II), Co(II), Fe(II), Mn(II) and Zn(II)]



**Figure 2:**  $[M(OAcPh-en)(Ben-opd)]H_2O$   
[M = Ni(II), Cu(II), Co(II), Fe(II), Mn(II) and Zn(II)]

**Antimicrobial activity:**

The antimicrobial activity of the complexes against Gram positive bacteria *Bacillus Substlis* and Gram negative bacteria *E.Coli* have been done using the “Agar diffusion method” [42]. The diameter of inhibition zone of the various compounds is recorded in table III. All the compounds have significant antibacterial activity at  $1.0 \times 10^4 \mu\text{gml}^{-1}$  against bacteria. The compounds [M(OAcPh-en)(Ben-en)].H<sub>2</sub>O, [M = Ni(II), Cu(II), Co(II), Fe(II), Mn(II) and Zn(II)] are more active as compared to the compounds [M(OAcPh-en)(Ben-opd)].H<sub>2</sub>O, [M = Ni(II), Cu(II), Co(II), Fe(II), Mn(II) and Zn(II)] against bacteria. All the compounds screened are more active against gram positive bacteria *Bacillus Substlis* then gram negative bacteria *E.Coli*.

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