



ISSN No: 0975-7384  
CODEN(USA): JCPRC5

*J. Chem. Pharm. Res.*, 2010, 2(5):518-525

---

**Synthesis, characterization and biocidal studies of some transition metal complexes containing tetra dentate and neutral bi dentate schiff base**

**Y.J.Thakor, S.G.Patel and K.N.Patel\***

*Department of Chemical Sciences, N. V. Patel College of Pure and Applied Sciences,  
Vallabh Vidyanagar, Gujarat, India*

---

**ABSTRACT**

*New mixed-ligand complexes of the transition metal ions Ni(II), Cu(II), Co(II), Fe(II), Mn(II), and Zn(II) with N,N'-bis[2-hydroxyacetophenone]ethylenediamine (OAcPh-en) and bis[4-methoxybenzylidene]ethylenediamine (MeBen-en) or bis[4-methoxybenzylidene]o-phenylenediamine (MeBen-opd) have been synthesized. 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**

In coordination chemistry, Schiff bases have a significant role as ligands still a century after their discovery [1]. Schiff base derived from the reaction of aromatic aldehydes and aliphatic or aromatic amines represent an important series of widely studied organic ligands. Schiff base and the relevant transition metal complexes are still found to be of great interest in inorganic chemistry although this subject has been studied extensively [2-4]. The chelating abilities and analytical and biological applications of these compounds have attracted remarkable attention [5]. The complexes containing O, N donor atoms are very important owing to their significant antibacterial and anticancer activity [6,7]. The Schiff base complexes have been used in catalytic reactions and as models for biological systems and also have been used as fine chemicals and medical substrates [8]. It is well known that some drugs have increased activity when administered as metal complexes then as free organic compounds [6,9]. We prepared new mixed ligand complexes of the type  $[M(OAcPh-en)(MeBen-en)] \cdot H_2O$  and  $[M(OAcPh-en)(MeBen-opd)] \cdot H_2O$  where M = Ni(II), Cu(II), Co(II), Fe(II), Mn(II) and Zn(II). OAcPh-en = N,N'-bis[2-hydroxyacetophenone]ethylenediamine, MeBen-en = bis[4-methoxy benzylidene] ethylene diamine, MeBen-opd = bis[4-methoxy benzylidene] -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.

## EXPERIMENTAL SECTION

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

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

### *Synthesis of bis[4-methoxybenzylidene]ethylenediamine (MeBen-en):*

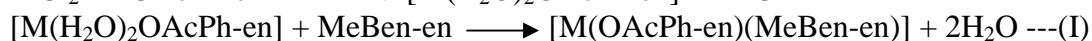
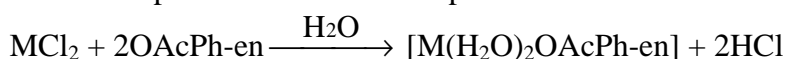
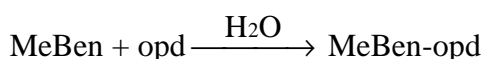
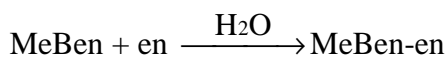
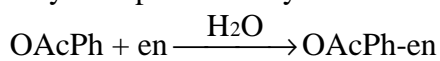
The ligand MeBen-en was synthesized by refluxing an ethanoic solution of 4-methoxybenzaldehyde and ethylenediamine (2:1 molar ratio) for one hour. The reaction mixture was cooled and a bright yellow crystalline solid of bis[4-methoxybenzylidene]ethylenediamine (MeBen-en) separated. The solid was filtered off and recrystallized from ethanol to give bright yellow crystals.

### *Synthesis of bis[4-methoxybenzylidene]o-phenylenediamine (MeBen-opd):*

The ligand MeBen-opd was synthesized by refluxing an ethanoic solution of 4-methoxybenzaldehyde and o-phenylenediamine (2:1 molar ratio) for one hour. The reaction mixture was cooled and a bright yellow crystalline solid of bis[4-methoxybenzylidene]o-phenylenediamine (MeBen-opd) separated. The solid was filtered off and recrystallized from ethanol to give yellow crystals.

### *Preparation of the complexes:*

The preparation of [Ni(OAcPh-en)(MeBen-en)].H<sub>2</sub>O was carried out by refluxing an ethanoic solution (250ml) of Nickel diaquo complex (0.01M) with Neutral bi dentate ligand bis[4-methoxybenzylidene]ethylenediamine (MeBen-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 Cu(II), Co(II), Fe(II), Mn(II), and Zn(II) were prepared similarly. The mixed ligand complexes of [M(OAcPh-en)(MeBen-opd)].H<sub>2</sub>O [M = Ni(II), Cu(II), Co(II), Fe(II), Mn(II) and 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 [12] 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)(MeBen-en)].H <sub>2</sub> O	644.94	Brown	>360	66.91 (66.98)	5.96 (5.89)	8.73 (8.68)	8.40 (8.51)	65.5	5.88
[Fe(OAcPh-en)(MeBen-en)].H <sub>2</sub> O	645.85	Brown	253	66.94 (66.88)	5.81 (5.88)	8.62 (8.67)	8.76 (8.64)	78.5	5.32
[Co(OAcPh-en)(MeBen-en)].H <sub>2</sub> O	648.93	Yellow	231	66.48 (66.57)	5.94 (5.85)	8.56 (8.62)	9.17 (9.08)	72.1	4.38
[Ni(OAcPh-en)(MeBen-en)].H <sub>2</sub> O	648.70	Yellowish Orange	282	66.64 (66.59)	5.78 (5.85)	8.60 (8.63)	9.12 (9.04)	68.9	2.95
[Cu(OAcPh-en)(MeBen-en)].H <sub>2</sub> O	653.55	Brown	248	66.02 (66.10)	5.87 (5.81)	8.68 (8.56)	9.64 (9.72)	75.3	1.84
[Zn(OAcPh-en)(MeBen-en)].H <sub>2</sub> O	655.38	Yellow	200	65.98 (65.91)	5.68 (5.79)	8.61 (8.54)	9.94 (9.97)	64.2	—
[Mn(OAcPh-en)(MeBen-opd)].H <sub>2</sub> O	692.94	Brown	212	69.32 (69.27)	5.52 (5.48)	8.01 (8.08)	7.96 (7.92)	63.70	5.92
[Fe(OAcPh-en)(MeBen-opd)].H <sub>2</sub> O	693.85	Brown	220	69.14 (69.14)	5.44 (5.47)	8.02 (8.07)	8.09 (8.04)	72.52	5.40
[Co(OAcPh-en)(MeBen-opd)].H <sub>2</sub> O	696.93	Yellowish Brown	236	68.82 (68.87)	5.49 (5.45)	8.08 (8.03)	8.50 (8.45)	81.6	4.31
[Ni(OAcPh-en)(MeBen-opd)].H <sub>2</sub> O	696.70	Yellowish Orange	240	68.84 (68.87)	5.50 (5.45)	8.10 (8.03)	8.40 (8.42)	76.2	2.85
[Cu(OAcPh-en)(MeBen-opd)].H <sub>2</sub> O	701.55	Yellowish Brown	220	68.46 (68.41)	5.49 (5.41)	7.91 (7.98)	9.00 (9.05)	73.5	1.85
[Zn(OAcPh-en)(MeBen-opd)].H <sub>2</sub> O	703.38	Yellow	290	68.29 (68.24)	5.44 (5.40)	7.91 (7.96)	9.32 (9.29)	79.5	—

**Table – II Infrared spectra (cm<sup>-1</sup>)**

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	----	----
Meben-en	1332	1615	1147	1510	----	----
Meben-opd	1328	1618	1145	1502	----	----
[Mn(OAcPh-en)(MeBen-en)].H <sub>2</sub> O	1342	1599	1140	1531	459	520
[Fe(OAcPh-en)(MeBen-en)].H <sub>2</sub> O	1330	1595	1150	1528	460	520
[Co(OAcPh-en)(MeBen-en)].H <sub>2</sub> O	1335	1600	1145	1535	455	515
[Ni(OAcPh-en)(MeBen-en)].H <sub>2</sub> O	1340	1605	1152	1532	455	520
[Cu(OAcPh-en)(MeBen-en)].H <sub>2</sub> O	1325	1598	1150	1529	450	518
[Zn(OAcPh-en)(MeBen-en)].H <sub>2</sub> O	1330	1593	1142	1535	453	515
[Mn(OAcPh-en)(MeBen-opd)].H <sub>2</sub> O	1341	1599	1140	1531	445	515
[Fe(OAcPh-en)(MeBen-opd)].H <sub>2</sub> O	1330	1600	1147	1528	450	513
[Co(OAcPh-en)(MeBen-opd)].H <sub>2</sub> O	1325	1598	1145	1535	450	510
[Ni(OAcPh-en)(MeBen-opd)].H <sub>2</sub> O	1325	1600	1142	1531	445	513
[Cu(OAcPh-en)(MeBen-opd)].H <sub>2</sub> O	1329	1601	1150	1528	452	510
[Zn(OAcPh-en)(MeBen-opd)].H <sub>2</sub> O	1333	1598	1152	1534	450	510

**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  $1324\text{-}1340\text{ cm}^{-1}$  [6,13-14]. In the Schiff base  $\nu(\text{C=N})$  stretching band at  $1610\text{ cm}^{-1}$ . This band shifts to lower energy by 10 to  $30\text{ cm}^{-1}$  in chelates indicating co-ordination through the azomethine nitrogen [15-22]. The sharp bend in the range  $750\text{-}780\text{ cm}^{-1}$  and  $1525\text{-}1535\text{ cm}^{-1}$  are due to aromatic  $\nu(\text{C-H})$  [6,23] and  $\nu(\text{C=C})$  [6,24] respectively. The frequencies in the range  $1145\text{-}1165\text{ cm}^{-1}$  attributed to  $\nu(\text{C-N})$  stretching [25]. 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 [26-30]. 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 [31].

**Magnetic Measurements:**

The magnetic moment values are useful in the evaluation of results provided by the other techniques of the structural investigation [31]. The magnetic moment data are presented in table-1. The magnetic moment of the Cu(II) complex (1.84-1.85 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 [32-33]. The magnetic moment values for Co(II) complexes is 4.31-4.38 B.M. corresponding to three unpaired electrons which suggests an octahedral geometry [34-35]. The magnetic moment of the nickel complex at room temperature was observed 2.84-2.95 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 [32,36]. The magnetic moment value for Fe(II) complex is 5.32-5.40 B.M. at room temperature and this value is characteristic of high spin octahedral geometry for this complex [33]. The Mn(II) complex show magnetic moments is 5.88-5.92 B.M. at room temperature corresponding to five unpaired electrons which suggest octahedral geometry [37]. Zn(II) complex is show diamagnetic nature and is found to be octahedral geometry [34, 38-40].

**Electronic spectra:**

The absorption bands for the complexes will help to give an idea of their structure [41]. Electronic spectrum of Co(II) complex shows bands, 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 centres around the metal ion [42]. The electronic spectrum of the Cu(II) complex recorded at room temperature, in DMF solution, shows broad band absorption at  $13,120$ ,  $15,340$  and  $24,480\text{ cm}^{-1}$ , which may be assign to  ${}^2\text{B}_{1g} \rightarrow {}^2\text{A}_{1g}$ , ( $d_{x^2-y^2} \rightarrow d_z^2$ )(v1),  ${}^2\text{B}_{1g} \rightarrow {}^2\text{B}_{2g}$ , ( $d_{x^2-y^2} \rightarrow d_{zy}$ )(v2), and  ${}^2\text{B}_{1g} \rightarrow {}^2\text{E}_g$ , ( $d_{x^2-y^2} \rightarrow d_{zy}$ ,  $d_{yz}$ )(v3) transition and it is in conformity with octahedral geometry [43]. 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 [44]. 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 [45]. 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 [46]. The Zn(II) complex has not shown any d-d absorptions [47].

**Thermo gravimetric Analysis:**

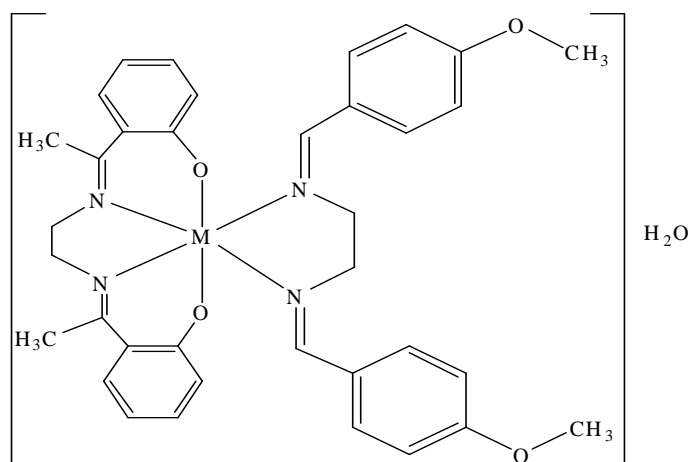
The analysis of the thermal curve of  $[M(\text{OAcPh-en})(\text{MeBen-en})].\text{H}_2\text{O}$  and  $[M(\text{OAcPh-en})(\text{MeBen-opd})].\text{H}_2\text{O}$  clearly indicated that the weight loss between 45-105°C corresponds to one water molecule for all complexes. Because of the low temperatures, this molecule may be considered as crystal water [48]. The curves in the range between 105-305°C suggested that loss in weight for all complexes correspond to evaporation of bis[4-methoxybenzylidene]ethylenediamine (MeBen-en) and bis[4-methoxybenzylidene]o-phenylenediamine (MeBen-opd). The range above 305°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.

**Antimicrobial activity:**

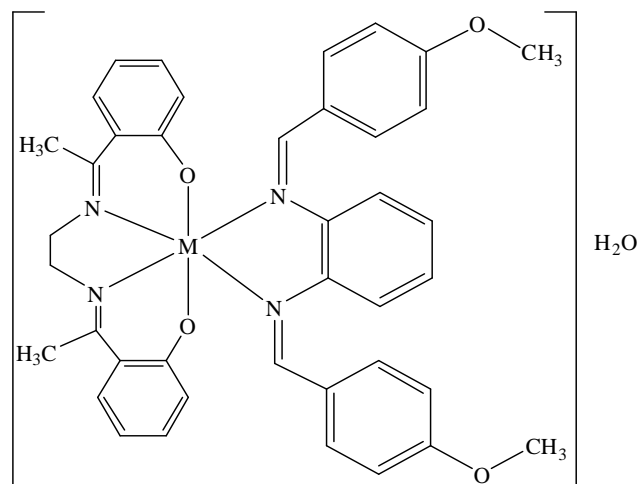
The antimicrobial activity of the complexes against Gram positive bacteria *Bacillus Substlis* and Gram negative bacteria *E.Coli* have been done using the "Agar diffusion method" [49]. The diameter of inhibition zone of the various compounds is recorded in table III. All the compounds have significant antibacterial activity at  $1.0 \times 10^4 \mu\text{gml}^{-1}$  against bacteria. The antimicrobial activity of the complexes has been found to decrease in the following order,  $\text{Ni} > \text{Cu} > \text{Mn} > \text{Zn} > \text{Fe} > \text{Co}$ . All the compounds screened are more active against gram positive bacteria *Bacillus Substlis* than gram negative bacteria *E.Coli*.

**Table – III Antimicrobial activity**

Complexes	Diameter of inhibition zone (mm)	
	<i>Bacillus Substlis</i>	<i>E.Coli</i>
$[\text{Mn}(\text{OAcPh-en})(\text{MeBen-en})].\text{H}_2\text{O}$	16	These metal complexes do not affect on <i>E.Coli</i> bacteria so there is no inhibition zone.
$[\text{Fe}(\text{OAcPh-en})(\text{MeBen-en})].\text{H}_2\text{O}$	20	
$[\text{Co}(\text{OAcPh-en})(\text{MeBen-en})].\text{H}_2\text{O}$	21	
$[\text{Ni}(\text{OAcPh-en})(\text{MeBen-en})].\text{H}_2\text{O}$	14	
$[\text{Cu}(\text{OAcPh-en})(\text{MeBen-en})].\text{H}_2\text{O}$	15	
$[\text{Zn}(\text{OAcPh-en})(\text{MeBen-en})].\text{H}_2\text{O}$	18	
$[\text{Mn}(\text{OAcPh-en})(\text{MeBen-opd})].\text{H}_2\text{O}$	20	
$[\text{Fe}(\text{OAcPh-en})(\text{MeBen-opd})].\text{H}_2\text{O}$	22	
$[\text{Co}(\text{OAcPh-en})(\text{MeBen-opd})].\text{H}_2\text{O}$	24	
$[\text{Ni}(\text{OAcPh-en})(\text{MeBen-opd})].\text{H}_2\text{O}$	17	
$[\text{Cu}(\text{OAcPh-en})(\text{MeBen-opd})].\text{H}_2\text{O}$	18	
$[\text{Zn}(\text{OAcPh-en})(\text{MeBen-opd})].\text{H}_2\text{O}$	21	



**Figure 1:  $[\text{M}(\text{OAcPh-en})(\text{MeBen-en})].\text{H}_2\text{O}$**   
 $[\text{M} = \text{Ni}(\text{II}), \text{Cu}(\text{II}), \text{Co}(\text{II}), \text{Fe}(\text{II}), \text{Mn}(\text{II}) \text{ and } \text{Zn}(\text{II})]$



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

### Acknowledgements

The authors are thankful to Dr. Vasishta Bhatt and Dr. Akshaya Gupte, Heads, Department of Chemical Science and Microbiology respectively, Natubhai V. Patel College of Pure and Applied Sciences, Vallabh Vidyanagar, Gujarat, India for providing the necessary laboratory facilities and SICART, Vallabh Vidyanagar Gujarat, India, for providing analytical support.

### REFERENCES

- [1] Demir Mulazimoglu Aysen; Ozkalp Birol and Mercimek Bedrettin. *Int. J. Drug Dev. & Res.*, **2010**, 2(1), 102-107
- [2] N.H. Patel; H.M. Parekh and M.N. Patel. *Transition Metal Chem.*, **2005**, 30, 13-17
- [3] M. Sekerci. *XIV<sup>th</sup> National chemistry congress*, Diyarbakir, Turkey, **2000**, 414.
- [4] M. Sekerci; C. Alkan; A. Cukurovali and Sayadam, *XIII<sup>th</sup> National chemistry congress*, Samsun, Turkey, **1999**, 182.
- [5] A.A. Jarrahpour; M. Motamedifar; K. Pakshir; N. Hadi and M. Zarei. *Journal of Molecules*, **2004**, 9, 815-824.
- [6] M.N. Patel; N.H. Patel; P.K. Panchal and D.H. Patel. *synth. React. Inorg. Met.-Org. Chem.*, **2004**, 34(5), 873-882.
- [7] A. Saxena; J.K. Koacher; J.P. Tandon. *Inorg. Nucl. Chem. Lett.*, **1981**, 17(7-8), 229-233.
- [8] Sheikh Aadil Abbas; Muhammad Munir; Annum Fatima; Sumera Naheed and Zeeshan Ilyas. *The BIOL (E-Journal of Life Sciences)*, **2010**, 1(2), 37-40.
- [9] A. Gerli; K.S. Hogen; L.G. Marzilli. *Inorg. Chem.*, **1991**, 30(24), 4673-4676.
- [10] R.M. Ramadan; M.S.A. Hamza and S.A. Ali. *J. Coordi. chem.*, **1998**, 43, 31-39.
- [11] S.A. Ali; A.A. Solliman; M.M. Aboaly and R.M. Ramadan. *J. Coord. Chem.*, **2002**, 55(10), 1161-1170
- [12] A.I. Vogel, "A Text Book of Quantitative Inorganic Analysis", Longmans green, London **1962**.
- [13] A.A. Ahmed; S.A. BenGuzzi and A.O. Agoob Rasayan. *J. chem.*, **2009**, 2(2), 271-275.
- [14] S.J. Gruber; C.M. Harris; E. Sinn. *J. Inorg. Nucl. Chem.*, **1968**, 30, 1805-1830.
- [15] Rekha Shankar Hunoor; Basavaraj Rudragouda Patil; Dayananda Shivappa Badiger; Ramesh Shivananadappa Vadavi; Kalagouda Basappa Gudasi; Chandrashekhar Venkaraddy Magannavar and Iranna Sangappa Muchandi. *Chem. Pharm. Bull.*, **2010**, 58(5) 712-716.
- [16] Sarika R. Yaul; Amit R. Yaul; Gaurav B. Pethe and Anand S. Aswar. *American-Eurasian Journal of Scientific Research*, **2009**, 4(4), 229-234.

- [17] K. Shanker; M. Ashok; P. Muralidhar Reddy; R. Rohini and V. Ravinder. *International Journal of ChemTech Research*, **2009**, 1(3), 777-783.
- [18] I. Demir; M. Akkaya; M. Bayrakci and A. I. Pekacar. *Asian J. Chem.*, **2007**, 19, 3954.
- [19] Byeong-Goo Jeong; Chae-Pyeong Rim; Seong-Keum Kook; Ki-Hyung Chjo and Yong-Kook Choi. *Bull. Korean Chem. Soc.*, **1996**, 17(2), 173.
- [20] Wan Nazihah Wan Ibrahim; Mustaffa Shamsuddin and Bohari M. Yamin; *The Malaysian Journal of Anal. Sci.*, **2007**, 11(1), 98-104.
- [21] L. Demir and A. I. Pecakar. *Synth. React. Inorg. Met. Org. Nano-Met. Chem.*, **2005**, 35, 825.
- [22] T. Daniel Thangadurai and Son- Ki Ihm. *J. Ind. Eng. Chem.*, **2003**, 9(5), 569-575.
- [23] H.H. Freedman; *J. Am. Chem. Soc.*, **1961**, 83, 2900-2905.
- [24] B.D. Sarma; C. Bailar. *J. Am. Chem. Soc.*, **1955**, 77, 5476.
- [25] K.N. Patel; K.M. Patel; N.H. Patel; C.P. Patel and M.N. Patel. *Synth. React. Inorg. Met. Org. Chem.*, **2000**, 30(8), 1617-1627.
- [26] Netra Pal Singh; Vaibhav Pratap Tyagi and Bindiya Ratnam. *J. Chem. Pharm. Res.*, **2010**, 2(1), 473-477.
- [27] Shayma A. Shaker; Yang Farina; Abbas A. Salleh. *European Journal of Scientific Research* **2009**, 33(4), 702-709.
- [28] Hamdi Temel; Berrin Ziyadanogullari; Isil Aydin and Firat Aydin. *J. Coord. Chem.*, **2005**, 58(14), 1177-1185.
- [29] Hamdi Temel; Umit Cakir; Veysel Tolan; Birolc Otludin and H. Ibrahim Ugras. *J. Coord. Chem.*, **2004**, 57(7), 571-581.
- [30] Hamdi Temel; Umit Cakir; H. Ibrahim Ugras and Memet Sekerci. *J. Coord. Chem.*, **2003**, 56(11), 943-951.
- [31] Abdul hakim; A. Ahmed; Salima A. BenGuzzi. *Journal of Science and Its Applications*, **2008**, 2(1), 83-90.
- [32] Dharpal Singh; Krishan Kumar; Ramesh Kumar and Jitender Singh. *J. Serb. Chem. Soc.*, **2010**, 75(2), 217-228.
- [33] Shayma A. Shaker; Yang Farina; Abbas A. Salleh. *European Journal of Scientific Research*, **2009**, 33(4), 702-709.
- [34] R. Shakru; N.J.P. Subhashini; K. Sathish Kumar; Shivaraj. *J. Chem. Pharm. Res.*, **2010**, 2(1), 38-46.
- [35] N. Mcmdal, O.K. Dcy; S. Mitra and K.M. Abdul Malik. *Polyhedron.* , **2000**, 19, 2707.
- [36] M S Surendra Babu; P G Krishna; K Hussain Reddy & G H Philip. *Indian Journal of Chemistry*, **2008**, 47A, 1661-1665.
- [37] S. Chandra; M. Tyagi and K. Sharma. *J. Iran. Chem. Soc.*, **2009**, 6(2), 310-316.
- [38] V.P. Krzyminiewska; H. Litkowska and W.R. Paryzek. *Monatsh. Chem.*, **1999**, 130, 243.
- [39] Smartin; M.R. Bermejo; A.M.G. Deibe; M. Manetro; C. Lage and A.J.C.Filho. *Polyhedron.* , **2000**, 19, 185.
- [40] K. Bertonecelio; G.D. Fallon; K.S. Mury and E.R.T. Tiekine, *Inorg Chem.*, **1999**, 130, 243.
- [41] Dr.Matheel D. Ai-Sabti; dr.Mohmad J.Al-Jeboori & Kanan M. Tawfiq. *Eng. & Tech. Journal*, **2010**, 28(1).
- [42] Pulimamidi Saritha Reddy; P.V.Anantha Lakshmi and V.Jayatyaga raju. *International Journal of Chem Tech Research.* , **2010**, 2(3), 1494-1500.
- [43] Rayees Ahmad Sheikh; Sheikh Shreaz; Luqman Ahmad Khan and Athar Adil Hashmi. *J. Chem. Pharm. Res.*, **2010**, 2(3), 274-286.
- [44] N.Turan; N.Colak; M. Sekerci. *International Journal of Natural and Engineering Sciences*, **2008**, 2(3), 27-32.
- [45] Sulekh Chandra and Amit Kumar Sharma. *Research Letters in Inorganic Chemistry*, **2009**, Article ID 945670.
- [46] K.N. Patel; N.H. Patel; K.M. Patel and M.N. Patel. *Synth. React. Inorg. Met.-Org. Chem.*, **2000**, 30(5), 829-841.

---

[47] Rekha Shankar Hunoor; Basavaraj Rudragouda Patil; Dayananda Shivappa Badiger; Ramesh Shivananadappa Vadavi; Kalagouda Basappa Gudasi; Chandrashekhhar Venkaraddy Magannavar and Iranna Sangappa Muchandi. *Chem. Pharm. Bull.*, **2010**, 58(5), 712-716.

[48] K.N. Patel; N.H. Patel; K.M. Patel and M.N. Patel. *Synth. React. Inorg. Met.-Org. Chem.*, **2000**, 30(5), 921-930.

[49] British pharmacopeia, vol.2, Her Majesties stationary Office, London, A112(**1980**) and British pharmacopeia, Pharmaceutical Press, London, p.796, **1953**.