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Synthesis, Characterization and Antimicrobial activity of Mixed Ligand Complexes of Mn(II) and Zn(II) with Phthalic Acid or Succinic Acid and Heterocyclic Amines

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ABSTRACT

Mixed ligand complexes of Mn(II) and Zn(II) in the presence of phthalic acid or succinic acid and heterocyclic amines were synthesized and characterized. The structure of the synthesized complex was determined by elemental analysis, magnetic moment measurements, conductometric, FTIR spectral studies, and electronic spectra analysis. The complexes were showed colored, non-electrolytic in nature. Complexes of Mn(II) were paramagnetic and Zn(II) were diamagnetic. Our results indicated that the Mn(II) complexes were octahedral, while Zn(II) complexes were of tetrahedral and octahedral structure. The synthesized complexes showed both antifungal and antibacterial activity.

Keywords: Mixed ligand complexes, Phthalic acid, Succinic acid, Heterocyclic amines, Electronic spectra, Antimicrobial activity

INTRODUCTION

For the last two decades, the synthesis of coordination compound with transition metals and their applications have become an attractive field because of the excellent properties such as catalysis, ion exchange, microelectronics, nonlinear optics, porous materials, etc. [1-7]. Due to the smaller size, and coordination numbers (4 or 6) of first row transition metal ions are easily coordinated with nitrogen and oxygen atoms and that's why their coordination geometric structures are simple [8-14]. The mixed ligands complexes of transition metals are very important in different field of chemistry like photochemistry, analytical chemistry, magneto chemistry etc. [15].

The coordination chemistry of Mn has studied in inorganic biochemistry [16] as an interested field. The magnetic properties, diverse catalytic activity of such compounds is now being explored [17-19] for its biological importance. Recently a few reports on the antimicrobial activity of Mn(II) complexes were studied [20-24]. The complexes of Zn(II) were observed good biological and exhibit enhanced activities as compared to their parental ligands [25]. The complexes of Zn(II) showed good bacterial and fungicidal effects [26].

The synthesis of one, two or three dimensional networks [27-29] with metal complexes [30-32] succinate ($C_4H_4O_4$)₂ anions were used as a polydentate ligands. Phthalate acid is used as a bridging ligand to connect metal ions through the four oxygen atoms of its two carboxylate groups [33-35]. Pyridine ring containing organic compounds play an important role in many biological reactions [36].

In the present work, we describe the synthesis, characterization and antimicrobial activity of mixed ligand complexes of Mn(II) and Zn(II) with phthalic acid or succinic acid act as an primary ligands and heterocyclic amines that played as a secondary ligand. The structures of our used ligands were shown in **Figure 1**. The prepared complexes have also been tested in-vitro to assess their antibacterial and antifungal activities against some common reference bacteria and fungi.

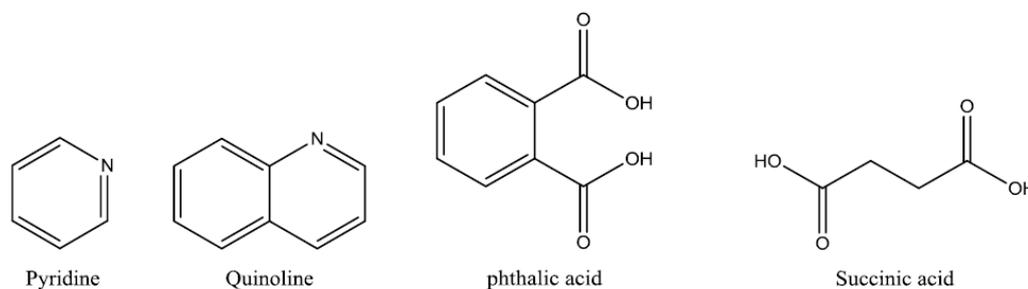


Figure 1: Structures of the ligands.

MATERIALS AND METHODS

Reagents and Chemicals

Manganese(II) chloride, $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$ (pure), zinc (II) chloride, $\text{ZnCl}_2 \cdot \text{H}_2\text{O}$ (97%) were purchased from May and Baker (England). The ligands succinic acid (97%), quinoline (pure) from BDH (England) and phthalic acid (pure), pyridine (pure) obtained from Thomas and Baker (India). Solvents were purified and dried according to standard procedures.

Physical Measurements

The melting or decomposition temperatures of all the synthesized metal complexes were detected in an electro thermal melting point apparatus model SMP30. It was not possible to measure the melting points beyond 390°C . We used SHERWOOD SCIENTIFIC Magnetic Susceptibility Balance for investigating our present study. Infrared spectra (KBr) were recorded in a SHIMADZU FTIR-8400 (Japan) spectrophotometer in the range of $4000\text{--}400\text{ cm}^{-1}$. The absorbances of the complexes were noted on SHIMADZU spectrophotometer (model UV-1800).

General Method for the Preparation of the Complexes

An ethanolic solution of metal chloride (1 mM) and deprotonated phthalic acid (1 mM) or succinic acid (1 mM) was mixed as a first ligand with constant stirring but no precipitate was observed. Ethanolic solution was used for dissolving the reactants. Then 25 mL an ethanolic potassium hydroxide solution of secondary ligand (pyridine and quinoline) in calculated ratio was mixed to the resulting mixture and heat on a magnetic regulator hot plate with constant stirring. We wait until the volume of the solution was decies to one half and allowed to cool. The precipitate was filtered, washed several times with ethanol and then dried in desiccators over anhydrous CaCl_2 .

Antibacterial and antifungal screening

The antibacterial and antifungal activities of the complexes were investigated against the tested bacterial species: Gram positive (*Bacillus cereus* and *Staphylococcus aureus*) and Gram negative (*Escherichia coli* and *Shigella sonnei*) and the fungal species: *Candida albicans*, *Saccharomyces cerevisiae* (Human Pathogens) and *Aspergillus niger* (Plant Pathogens). Ciprofloxacin and fluconazole were used as the standard antibacterial and antifungal agents. For this study diffusion method [37,38] was used. On the agar medium inoculated, the well was made with microorganisms. By using micropipette the well was filled with the test solution and the plate was incubated at 37°C for 16 h for bacteria and fungi. The test solution diffused and the growth of the inoculated microorganisms was affected during this incubated period. The zone of inhibition developed on the plate was measured. All the prepared complexes dissolved in DMSO were used for study.

RESULTS AND DISCUSSION

Elemental analysis and conductivity measurement

The physical properties and analytical data of the Mn(II) and Zn(II) complexes are tabulated in **Table 1**. Elemental data of the prepared complexes were represented in **Table 2**. Their structures have been proposed on the basis of conductivity and magnetic moment measurements. The molar conductance's of $1 \times 10^{-3}\text{ M}$ solution of the complexes in DMSO were measured at 30°C . The molar conductance values ranged from 55.3 to $99.9\text{ ohm}^{-1}\text{ cm}^2\text{ mol}^{-1}$ indicates that the compounds are non-electrolytic in nature [39,40].

Table 1: Physical properties and Analytical data of the complexes

No.	Complex	Color	Decomposition temp. ($\pm 5^\circ\text{C}$)	Molar Conductance ($\text{ohm}^{-1}\text{cm}^2\text{mol}^{-1}$)	μ_{eff} (B.M.)
1	[Mn(DPA)(Py) ₄]	Brown	384	74.8	1.3
2	[Mn(DPA)(Q) ₄]	Black	>385	94.5	2.1
3	[Mn(Suc)(Py) ₄]	Deep Brown	270	89.2	1.5
4	[Mn(Suc)(Q) ₄]	Black	>385	91.4	1.9
5	[Zn(DPA)(Py) ₄]	White	380	99.9	Dia
6	[Zn(DPA)(Q) ₄]	Off White	200	97.6	Dia
7	[Zn(DPA)(Py) ₂]	White	382	55.3	Dia
8	[Zn(DPA)(Q) ₂]	White	335	82.1	Dia

Where, Py=Pyridine, Q=Quinoline; DPA=Deprotonated phthalic acid, Succ=Deprotonated Succinic acid

Table 2: Elemental analysis data of the complexes

No.	Complex	Molecular Weight	%Metal	%C	%H	%N	%O	
1	[Mn(DPA)(Py) ₄]	Calculate	535.45	8.5	63.27	4.52	10.47	11.95
		Found	535.48	8.53	63.3	4.54	10.51	11.99
2	[Mn(DPA)(Q) ₄]	Calculate	735.69	6.19	72.37	4.38	7.62	8.7
		Found	735.73	6.23	72.41	4.42	7.65	8.74
3	[Mn(Suc)(Py) ₄]	Calculate	537.77	8.47	54	4.5	10.42	11.9
		Found	537.8	8.51	54.04	4.54	10.46	11.93
4	[Mn(Suc)(Q) ₄]	Calculate	688.01	6.62	70.35	4.69	9.3	8.14
		Found	688.05	6.63	70.38	4.72	9.34	8.18
5	[Zn(DPA)(Py) ₄]	Calculate	544.9	12	62.18	4.44	10.28	11.74
		Found	544.93	12.04	62.21	4.48	10.3	11.76
6	[Zn(DPA)(Q) ₄]	Calculate	746.14	8.76	71.35	4.32	7.51	8.58
		Found	746.18	8.79	71.37	4.35	7.53	8.6
7	[Zn(DPA)(Py) ₂]	Calculate	387.70	16.86	56.18	3.64	7.23	16.51
		Found	387.75	16.88	56.22	3.67	7.25	16.54
8	[Zn(DPA)(Q) ₂]	Calculate	487.82	13.40	64.49	3.72	5.74	13.12
		Found	487.85	13.45	64.54	3.76	5.80	13.16

Magnetic moment and electronic spectra

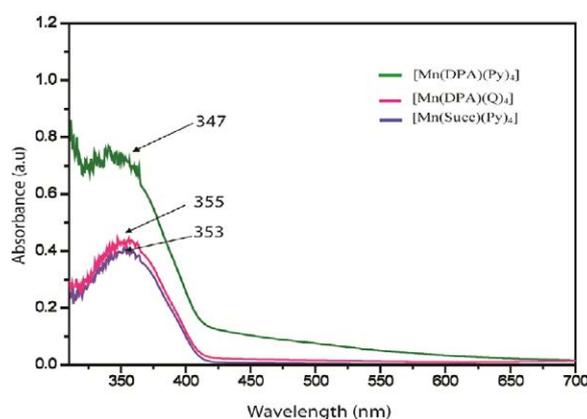
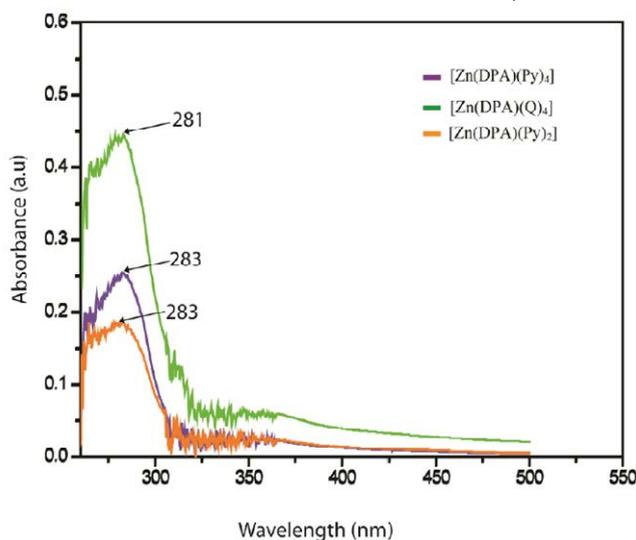
The observed values of effective moment (μ_{eff}) of the Mn(II) and Zn(II) complexes at room temperature are given in **Table 1**. Mn(II) complexes have μ_{eff} values (1.3-2.1 B.M.) shows the presence of one unpaired electron. So the compounds of Mn(II), under investigation are found to be paramagnetic [41]. But Zn(II) complexes have no unpaired electron. All the compounds of Zn(II), under investigation were found to be diamagnetic [42-44]. The electronic spectra of all complexes were recorded in 10^{-3} M DMSO at 30°C . The electronic spectra of [Mn(DPA)(Py)₄], [Mn(DPA)(Q)₄], [Mn(Suc)(Py)₄], and [Mn(Suc)(Q)₄] were shown 347, 355, 353 and 354 nm respectively, indicate octahedral geometry for the Mn(II) complex [45,46]. In Zn(II) complexes, d₁₀ orbital are completely filled hence it does not show any d-d electronic transition but exhibit charge transfer spectra [47]. The [Zn(DPA)(Py)₄], [Zn(DPA)(Q)₄] and [Zn(DPA)(Py)₂], [Zn(DPA)(Q)₂] complexes showed bands at 283, 281 and 283, 271 nm due to the L \rightarrow M charge transfer transition that correspond to octahedral [48] and tetrahedral structure [49,50]. The all data of electronic spectra of Mn(II) and Zn(II) complexes as shown in **Table 3** and **Figure 2 and 3**.

FTIR studies

FTIR spectra of the Mn(II) and Zn(II) complexes were shown in **Table 4** and **Figure 4**. The complexes display bands in the regions (1520-1600) and (1280-1390) cm^{-1} due to $\nu(\text{C}=\text{O})$ and $\nu(\text{C}-\text{O})$ respectively, significantly lower than that of free ligand indicating the coordination of metal ion through its carboxylate anion. The band observed in the (1395-1605) cm^{-1} region due to $\nu(\text{C}=\text{N})$ vibrations. The in-plane and out-of-plane ring deformation modes of heterocyclic amines observed at 680 and 620 cm^{-1} respectively undergo a positive shift in mixed ligand complexes confirming their coordination through nitrogen. The presence of metal nitrogen bonding in the complexes is evident from the appearance at 456-690 cm^{-1} region in the spectra of the complexes of $\nu(\text{M}-\text{N})$ modes [51] and $\nu(\text{M}-\text{O})$ appearance at (543-690) cm^{-1} .

Table 3: Data for the determination of UV-visible spectral bands **Antibacterial and antifungal screening**

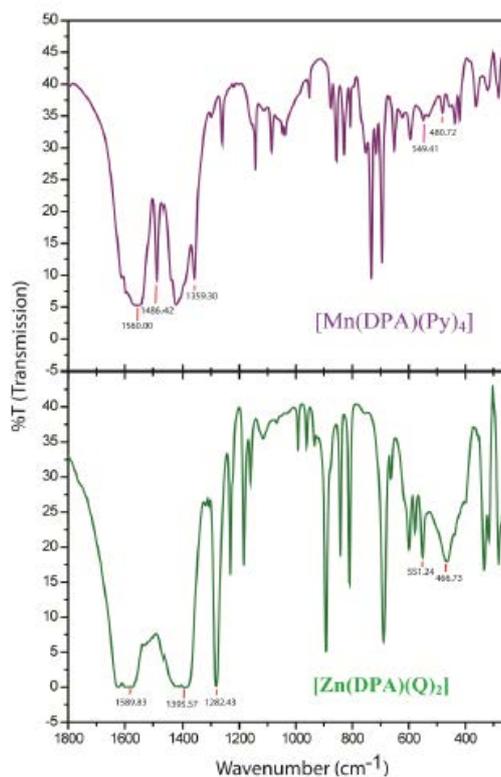
No.	Complex	λ_{\max} (nm)
1	[Mn(DPA)(Py) ₄]	347
2	[Mn(DPA)(Q) ₄]	355
3	[Mn(Suc)(Py) ₄]	353
4	[Mn(Suc)(Q) ₄]	354
5	[Zn(DPA)(Py) ₄]	283
6	[Zn(DPA)(Q) ₄]	281
7	[Zn(DPA)(Py) ₂]	283
8	[Zn(DPA)(Q) ₂]	271

**Figure 2:** UV/VIS absorption spectra achieved from the complexes of [Mn(DPA)(Py)₄], [Mn(DPA)(Q)₄], and [Mn(Suc)(Py)₄]**Figure 3:** UV/VIS absorption spectra achieved from the complexes of [Zn(DPA)(Py)₄], [Zn(DPA)(Q)₄], and [Zn(DPA)(Py)₂]

Metal complexes play an important role in regulating biological activities. The antimicrobial activity is depending on the cell membrane of the microorganisms and nature of metal ions [52]. The uses of metals for preventing or reducing growth of bacterial and fungal in medical treatment have been reported [53]. The antibacterial and antifungal potentiality of all the tested compounds against the four chosen bacteria and three fungi were shown in **Tables 5 and 6**. The zone of inhibition (mm) around the discs was measured. Results are expressed as zone of inhibition. The results of the antibacterial and antifungal screening indicated that all the compounds exhibit broad spectra against the reference bacteria and fungi.

Table 4: Data for the determination of IR Spectroscopy in cm^{-1}

No.	Complex	$\nu(\text{C}=\text{O})$	$\nu(\text{C}-\text{O})$	$\nu(\text{C}=\text{N})$	$\nu(\text{M}-\text{O})$	$\nu(\text{M}-\text{N})$
1	$[\text{Mn}(\text{DPA})(\text{Py})_4]$	1560.00	1359.30	1486.42	549.41	480.72
2	$[\text{Mn}(\text{DPA})(\text{Q})_4]$	1530.54	1298.56	1498.67	654.12	499.32
3	$[\text{Mn}(\text{Suc})(\text{Py})_4]$	1598.98	1380.45	1545.23	543.98	460.32
4	$[\text{Mn}(\text{Suc})(\text{Q})_4]$	1576.87	1298.65	1467.78	687.34	466.12
5	$[\text{Zn}(\text{DPA})(\text{Py})_4]$	1575.23	1308.87	1604.4	632.23	498.23
6	$[\text{Zn}(\text{DPA})(\text{Q})_4]$	1588.34	1390.65	1585.8	609.12	456.12
7	$[\text{Zn}(\text{DPA})(\text{Py})_2]$	1521.12	1376.45	1578.2	543.43	502.76
8	$[\text{Zn}(\text{DPA})(\text{Q})_2]$	1589.83	1282.43	1395.57	551.24	466.73

**Figure 4:** FTIR spectra of the complexes $[\text{Mn}(\text{DPA})(\text{Py})_4]$, and $[\text{Zn}(\text{DPA})(\text{Q})_2]$.**Table 5:** Antibacterial activity of the synthesized complexes

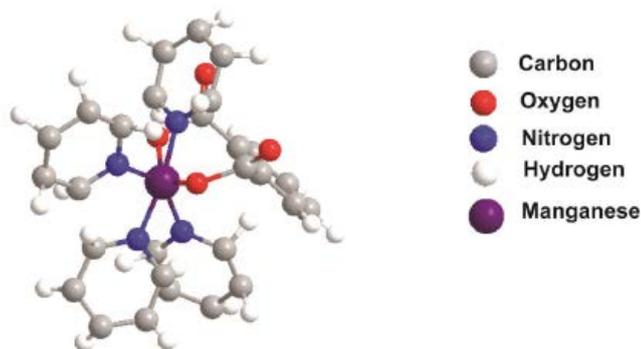
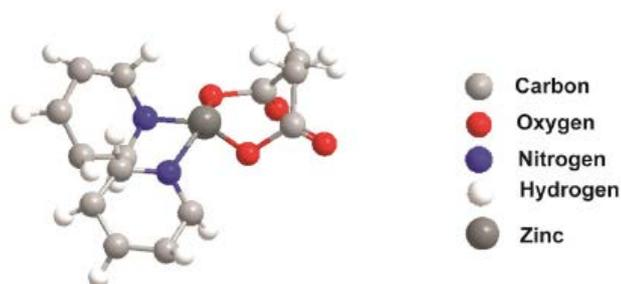
Complex	Zone of inhibition, diameter in mm			
	<i>Bacillus cereus</i>	<i>Staphylococcus aureus</i>	<i>Escherichia coli</i>	<i>Shigella sonnei</i>
$[\text{Mn}(\text{DPA})(\text{Q})_4]$	12	6	10	10
$[\text{Mn}(\text{DPA})(\text{Py})_4]$	7	6	10	8
$[\text{Zn}(\text{DPA})(\text{Py})_4]$	18	12	12	8
$[\text{Zn}(\text{DPA})(\text{Q})_2]$	6	6	11	6
Ciprofloxacin	30	40	30	40

Table 6: Antifungal activity of the synthesized complexes

Complex	Zone of inhibition, diameter in mm		
	<i>Candida albicans</i>	<i>Aspergillus niger</i>	<i>Saccharomyces cerevaceae</i>
[Mn(DPA)(Q) ₄]	8	16	14
[Mn(DPA)(Py) ₄]	12	12	12
[Zn(DPA)(Q) ₂]	20	16	12
[Zn(DPA)(Py) ₄]	12	15	18
Fluconazole	12	12	12

CONCLUSION

On the basis of the above analysis, we have proposed octahedral geometry for Mn(II) complex but tetrahedral, and octahedral geometry both for Zn(II) complexes. The proposed structures of [Mn(DPA)(Py)₄], and [Zn(Succ)(Py)₂] have presented in **Figure 5** and **Figure 6**. All the prepared complexes observed good antimicrobial activity. The tested mixed-ligand complexes showed higher activities against fungi compared to bacteria.

**Figure 5:** Probable 3-D structure of the complex. [Mn(DPA)(Py)₄].**Figure 6:** Probable 3-D structure of the complex [Zn(Succ)(Py)₂].

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