

**MIXED LIGAND COMPLEXES OF Ni(II) AND Cd(II) WITH PHTHALIC ACID OR SUCCINIC ACID AND HETEROCYCLIC AMINES: SYNTHESIS AND CHARACTERIZATION WITH ANTIMICROBIAL STUDY**

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**ABSTRACT**

Mixed ligand complexes of Ni(II) and Cd(II) with phthalic acid or succinic acid and heterocyclic amines have been synthesized. The synthesized complexes were characterized by using elemental analysis, conductometric, magnetic moment measurements, electronic spectra and FTIR spectral studies. The complexes were found to be colored, non-electrolytic in nature. The results showed that the Ni(II) complexes were octahedral, while Cd(II) complexes were tetrahedral structure. Antibacterial and antifungal activities of synthesized mixed ligand complexes were studied.

**KEYWORDS:** Mixed ligand complexes, FTIR, Electronic spectra, Antimicrobial activity.

**INTRODUCTION**

Last many decades, coordination compounds play a vital role in the research field of chemistry. Biologically active ligands are very much important for coordination compound.<sup>[1-3]</sup> Mixed ligands complexes also play an important role in magneto chemistry, analytical chemistry and photochemistry.<sup>[4]</sup> These complexes are given more important information about the explanation of enzymatic processes of biological applicability.<sup>[5,6]</sup> Also, these complexes are very much biologically active against pathogenic microorganisms.<sup>[7,8]</sup> Antimicrobial activities of Ni(II) and Cd(II) ions have been described.<sup>[9,10]</sup> The preparation of one, two or three-dimensional linkages<sup>[11-13]</sup> with metal centers<sup>[14-16]</sup> succinate ( $C_4H_4O_4$ )<sup>2-</sup> anions plays an important role because it used as a polydentate ligands. Phthalic acid, which is a bridging ligand containing two carboxylic groups in ortho position form coordination compound with a different metal.<sup>[17]</sup>

O and N donor atoms containing d-group metal complexes have biologically active.<sup>[18-21]</sup> Recently, one of the challenging areas of antibacterial investigation is to an improvement of new complexes to contract with resistant bacteria.<sup>[22,23]</sup> The production of new metal-

based compounds of antibacterial and antifungal is the part of metallo-organic chemistry.<sup>[24,25]</sup> Organic compounds with Pyridine ring play a vital role in numerous biological reactions.<sup>[26]</sup>

Here, we present the synthesis strategy, the characterization with antimicrobial activity of mixed ligand complexes of Ni(II) and Cd(II) via phthalic acid or succinic acid (primary ligand) and heterocyclic amines (secondary ligand). "Fig. 1" denotes the corresponding structure of used four ligands. Also, we broadly studied the antibacterial and antifungal activities of obtained complexes against different kind of common reference bacteria and fungi.

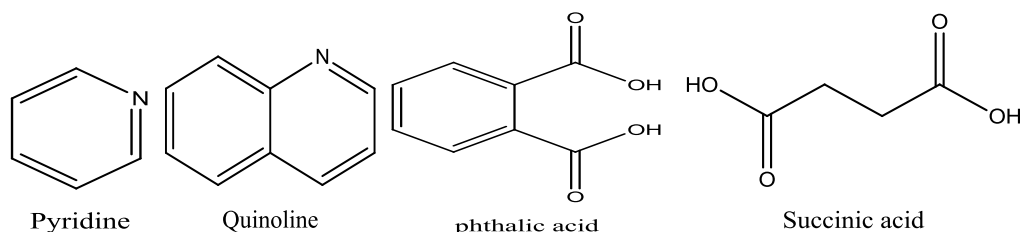


Fig. 1: Structures of the ligands.

## MATERIALS AND METHODS

### 2.1 Reagents and chemicals

Nickel chloride,  $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$  (Thomas & Baker,  $\geq 97\%$ ), Cadmium chloride,  $\text{CdCl}_2 \cdot \text{H}_2\text{O}$  (May & Baker, England), Succinic acid and Quinoline (BDH, England, 98%), Phthalic acid and Pyridine (Thomas & Baker, India). Solvents were purified and dried according to standard procedures.

### 2.2. General method for the preparation of the complexes

An ethanolic solution of Ni(II) or Cd(II) chloride (1 mM) and deprotonated phthalic acid or succinic acid (1 mM) was mixed as the first ligand with constant stirring but no precipitate was observed. Ethanolic solution was used to dissolve the reactants.<sup>[27]</sup> Then 25 mL an ethanolic KOH solution of secondary ligand (pyridine or quinoline) in calculated ratio was added to the resulting mixture and heat on a magnetic regulator hot plate with constant stirring. The volume of the solution was reduced to one half and allowed to cool. The precipitate was filtered, washed several times with ethanol and then dried in desiccators over anhydrous  $\text{CaCl}_2$ .

### 2.3. Physical measurements

The synthesized metal complexes were examined in an electro thermal melting point apparatus model SMP30 for the purpose of melting or decomposition temperature. Above  $390^\circ\text{C}$  temperature was impossible to measure the melting points. The magnetic moment data was investigated with the help of SHERWOOD SCIENTIFIC Magnetic Susceptibility Balance. SHIMADZU FTIR-8400 (Japan) spectrophotometer in the range of  $4000\text{--}400\text{ cm}^{-1}$  we used for FT-IR. The absorbances of all complexes were detected on SHIMADZU spectrophotometer model UV-1800.

### 2.4. Antibacterial and antifungal screening

The complexes were investigated against the tested four bacterial species among them two are gram-positive (*Bacillus cereus* and *Staphylococcus aureus*) and two are gram-negative (*Escherichia coli* and *Shigella sonnei*). We also observed three fungal species where two are human pathogens (*Candida albicans* and *Saccharomyces cerevisiae*) and one is planted pathogens (*Aspergillus niger*). Biological activities are tested with the help of

reference antibacterial (ciprofloxacin) and antifungal (fluconazole) agents respectively. The paper disc diffusion method<sup>[28-33]</sup> was used to test the antibacterial and antifungal activity of the complexes whose concentration  $40\text{ }\mu\text{g}/0.01\text{ mL}$  in DMSO solution. 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  $37^\circ\text{C}$  for 16 h for bacteria and fungi. The test solution diffused and the growth of the inoculated microorganisms was influenced during this incubated period. The zone of inhibition developed on the plate was measured.

## RESULTS AND DISCUSSION

### 3.1. Physical properties and elemental analysis

The physical properties and analytical data of the Ni(II) and Cd(II) complexes are tabulated in **Table 1**. At room temperature, the molar conductance's of  $1 \times 10^{-3}\text{ M}$  solution of the complexes in DMSO were investigated. The molar conductance values ranged from  $39.7$  to  $68.3\text{ ohm}^{-1}\text{ cm}^2\text{ mol}^{-1}$  indicates that the compounds are non-electrolytic in nature.<sup>[34,35]</sup> The elemental analysis data (obtained and calculated) of all the prepared complexes have presented in **Table 2**. It can be observed from this **Table 2** that the values of Metal, C, H, N, and O are consistent with their proposed molecular formula.

**Table 1: Physical properties and analytical data of the complexes.**

No.	Complex	Color	Decomposition temp. ( $\pm 5^{\circ}\text{C}$ )	Molar conductance ( $\text{ohm}^{-1} \text{cm}^2 \text{mol}^{-1}$ )	$\mu_{\text{eff}}$ (B.M.)
1	[Ni(DPA)(Py) <sub>4</sub> ]	Lemon	290	62.9	2.97
2	[Ni(DPA)(Q) <sub>4</sub> ]	Ash	381	68.3	3.13
3	[Ni(Suc)(Py) <sub>4</sub> ]	Lemon	270	55.8	2.95
4	[Ni(Suc)(Q) <sub>4</sub> ]	Light Brown	200	66.9	3.15
5	[Cd(DPA)(Py) <sub>2</sub> ]	White	206	64.1	Dia
6	[Cd(DPA)(Q) <sub>2</sub> ]	Off White	214	54.6	Dia
7	[Cd(Suc)(Py) <sub>2</sub> ]	White	213	39.7	Dia

Where,

Py = Pyridine, Q = Quinoline

DPA = Deprotonated phthalic acid, Succ = Deprotonated Succinic acid

**Table 2: Elemental analysis data of the complexes.**

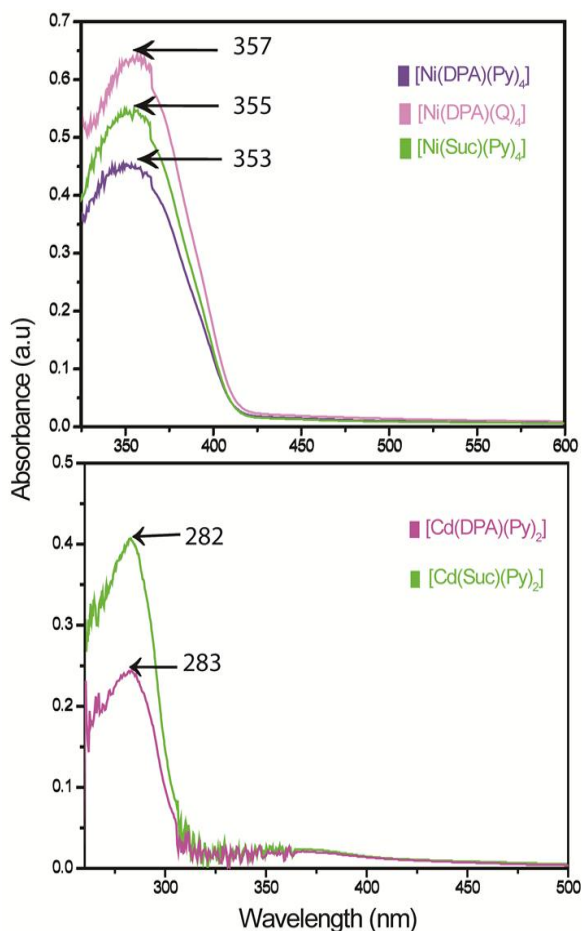
No.	Complex	Molecular Weight	% Metal	% C	% H	% N	% O	
1	[Ni(DPA)(Py) <sub>4</sub> ]	Calculate	539.20	10.88	62.83	4.49	10.39	11.87
		Found	539.24	10.92	62.86	4.55	10.43	11.92
2	[Ni(DPA)(Q) <sub>4</sub> ]	Calculate	739.44	7.94	72.00	4.36	7.58	8.66
		Found	739.51	7.97	72.04	4.40	7.63	8.69
3	[Ni(Suc)(Py) <sub>4</sub> ]	Calculate	491.52	11.94	59.08	4.92	11.40	13.02
		Found	491.57	11.97	59.18	4.95	11.46	13.08
4	[Ni(Suc)(Q) <sub>4</sub> ]	Calculate	691.76	8.48	69.97	4.67	8.10	9.25
		Found	691.79	8.53	70.03	4.72	8.14	9.29
5	[Cd(DPA)(Py) <sub>2</sub> ]	Calculate	434.72	25.85	50.10	3.25	6.44	14.72
		Found	434.76	25.87	50.14	3.28	6.47	14.75
6	[Cd(DPA)(Q) <sub>2</sub> ]	Calculate	534.85	21.02	58.38	3.39	5.24	11.96
		Found	534.87	21.05	58.42	3.44	5.26	11.99
7	[Cd(Suc)(Py) <sub>2</sub> ]	Calculate	387.04	29.04	43.77	3.66	7.24	16.54
		Found	387.08	29.08	43.80	3.71	7.29	16.58

### 3.2. Magnetic moment and electronic spectra

**Table 1**, represents the observed effective moment ( $\mu_{\text{eff}}$ ) values of the Ni(II) and Cd(II) complexes at room temperature. From the value of effective magnetic moments,  $\mu_{\text{eff}}$ , it can be concluded that the Ni(II) complexes are paramagnetic whereas Cd complexes are diamagnetic.<sup>[36-38]</sup> The electronic spectra of all complexes were noted at room temperature with the concentration of DMSO solution was  $10^{-3}$  M. The electronic spectra of [Ni(DPA)(Py)<sub>4</sub>], [Ni(DPA)(Q)<sub>4</sub>], [Ni(Suc)(Py)<sub>4</sub>], and [Ni(Suc)(Q)<sub>4</sub>] complexes were shown 353, 357, 355, and 350 nm respectively, and these are suggestive of octahedral geometry around Ni(II) ion.<sup>[39-41]</sup> In Cd (II) complexes,  $d^{10}$  orbital is completely filled that's why it does not show any d-d electronic transition but exhibit charge transfer spectra.<sup>[42]</sup> The [Cd(DPA)(Py)<sub>2</sub>], [Cd(DPA)(Q)<sub>2</sub>], and [Cd(Suc)(Py)<sub>2</sub>] complexes were shown 282, 281, and 283 nm due to the L→M charge transfer transition that corresponds to tetrahedral structure.<sup>[38,43]</sup> All data of electronic spectra of Ni(II) and Cd(II) complexes as shown in **Table 3** and "**Fig. 2**".

**Table 3: Data for the determination of UV-visible spectral bands.**

No.	Complex	$\lambda_{\text{max}}$ (nm)
1	[Ni(DPA)(Py) <sub>4</sub> ]	353
2	[Ni(DPA)(Q) <sub>4</sub> ]	357
3	[Ni(Suc)(Py) <sub>4</sub> ]	355
4	[Ni(Suc)(Q) <sub>4</sub> ]	350
5	[Cd(DPA)(Py) <sub>2</sub> ]	282
6	[Cd(DPA)(Q) <sub>2</sub> ]	281
7	[Cd(Suc)(Py) <sub>2</sub> ]	283



**Fig. 2:** UV/VIS absorption spectra achieved from the complexes of  $[\text{Ni}(\text{DPA})(\text{Py})_4]$ ,  $[\text{Ni}(\text{DPA})(\text{Q})_4]$ ,  $[\text{Ni}(\text{Suc})(\text{Py})_4]$ ,  $[\text{Cd}(\text{DPA})(\text{Py})_2]$ , and  $[\text{Cd}(\text{Suc})(\text{Py})_2]$ .

### 3.3. FTIR studies

FTIR spectra of the Ni(II) and Cd(II) complexes were represented in **Table 4** and “**Fig. 3**”. The complexes display bands in the regions (1550-1675) and (1150-1395)  $\text{cm}^{-1}$  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 (1395-1395)  $\text{cm}^{-1}$  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  $\text{cm}^{-1}$  respectively undergo a positive shift in mixed ligand complexes confirming their coordination through nitrogen. The low intensity in the ranges 360-495 and 500-580  $\text{cm}^{-1}$  can be allotted to stretching vibrations  $\nu(\text{M}-\text{N})$  and  $\nu(\text{M}-\text{O})$ , respectively.<sup>[44,45]</sup>

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

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	$[\text{Ni}(\text{DPA})(\text{Py})_4]$	1656.40	1306.25	1530.18	501.13	490.28
2	$[\text{Ni}(\text{DPA})(\text{Q})_4]$	1585.28	1359.75	1463.21	532.90	488.57
3	$[\text{Ni}(\text{Suc})(\text{Py})_4]$	1567.66	1296.78	1398.88	519.28	361.05
4	$[\text{Ni}(\text{Suc})(\text{Q})_4]$	1668.95	1390.40	1440.02	521.14	483.45
5	$[\text{Cd}(\text{DPA})(\text{Py})_4]$	1656.40	1298.96	1423.12	509.40	415.34
6	$[\text{Cd}(\text{DPA})(\text{Py})_2]$	1554.50	1154.14	1414.96	533.83	445.08
7	$[\text{Cd}(\text{Suc})(\text{Py})_2]$	1672.80	1387.56	1504.87	574.54	423.43

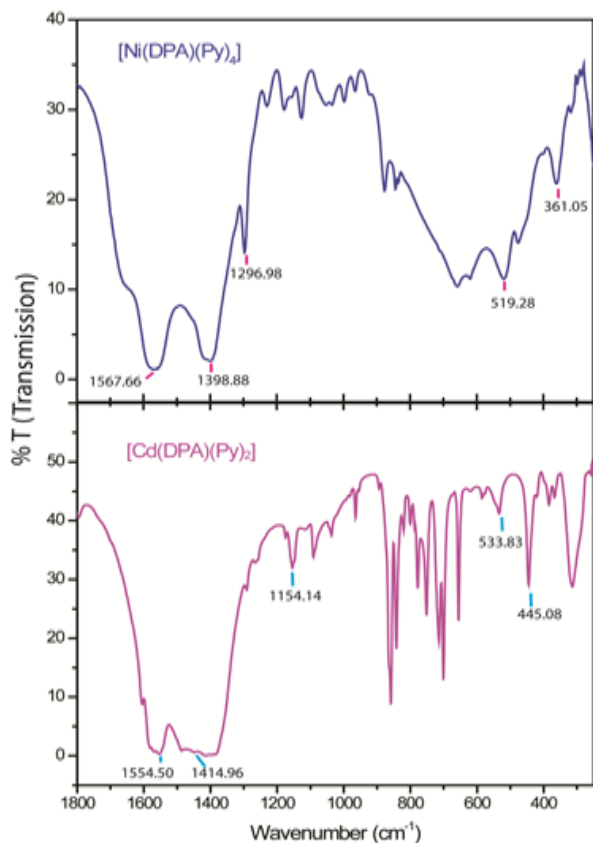


Fig. 3: FTIR spectra of the complexes  $[\text{Ni}(\text{DPA})(\text{Py})_4]$ , and  $[\text{Cd}(\text{DPA})(\text{Py})_2]$ .

### 3.4. Antibacterial and antifungal screening

Metal complexes show a significant character in regulating biological activities. The antimicrobial activity is subjected to the cell membrane of the

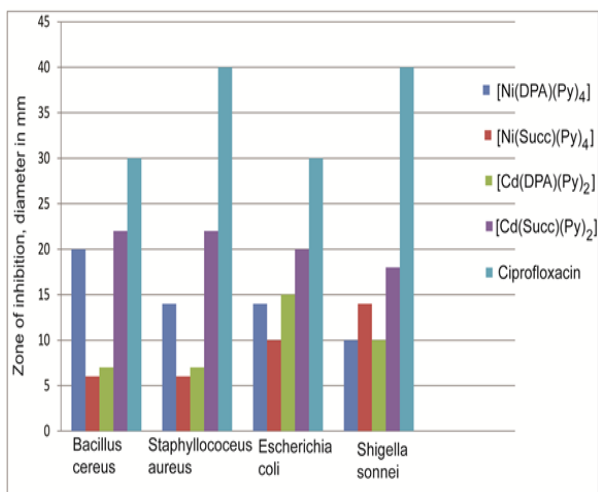
microorganisms and nature of metal ions.<sup>[46]</sup> The uses of metals for stopping or decreasing growth of bacterial and fungal in medical treatment have been reported.<sup>[47]</sup> The antibacterial and antifungal potentiality of all the tested compounds against the four chosen bacteria and three fungi were presented in **Table 5**, **Table 6** and “**Fig. 4**”, “**Fig. 5**” respectively. The zone of inhibition (mm) around the discs was measured. Results are spoken as the zone of inhibition. The results of the antibacterial and antifungal screening have shown that all the compounds display broad spectra against the reference bacteria and fungi.

Table 5: Antibacterial activity of the synthesized complexes

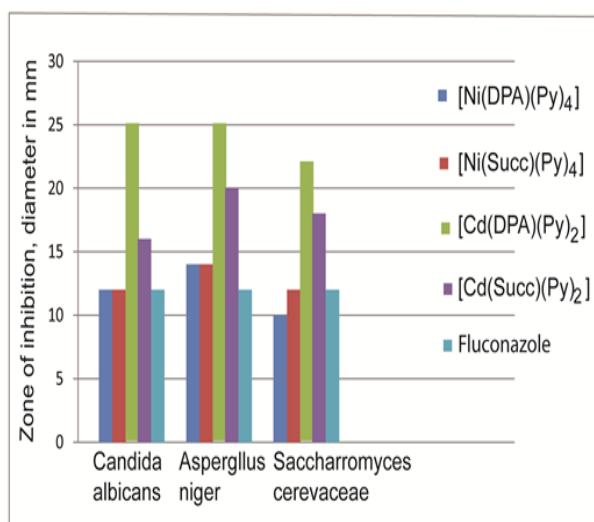
Complex	Zone of inhibition, diameter in mm			
	<i>Bacillus cereus</i>	<i>Staphylococcus aureus</i>	<i>Escherichia coli</i>	<i>Shigella sonnei</i>
$[\text{Ni}(\text{DPA})(\text{Py})_4]$	20	14	14	10
$[\text{Ni}(\text{Succ})(\text{Py})_4]$	6	6	10	14
$[\text{Cd}(\text{DPA})(\text{Py})_2]$	7	7	15	10
$[\text{Cd}(\text{Succ})(\text{Py})_2]$	22	22	20	18
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>
$[\text{Ni}(\text{DPA})(\text{Py})_4]$	12	14	10
$[\text{Ni}(\text{Succ})(\text{Py})_4]$	12	14	12
$[\text{Cd}(\text{DPA})(\text{Py})_2]$	25	25	22
$[\text{Cd}(\text{Succ})(\text{Py})_2]$	16	20	18
Fluconazole	12	12	12



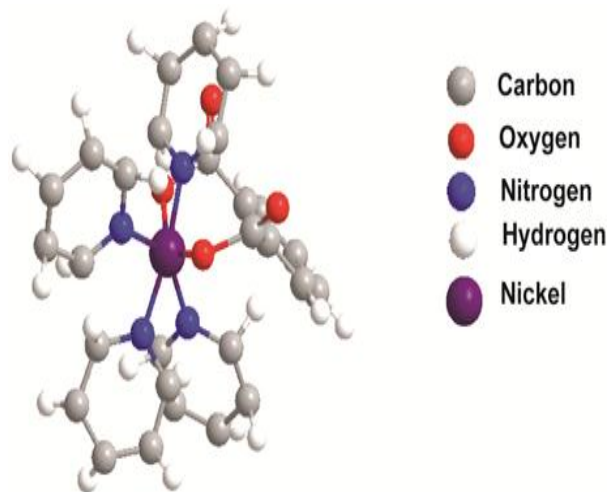
**Fig. 4:** Antibacterial activity of the synthesized complexes.



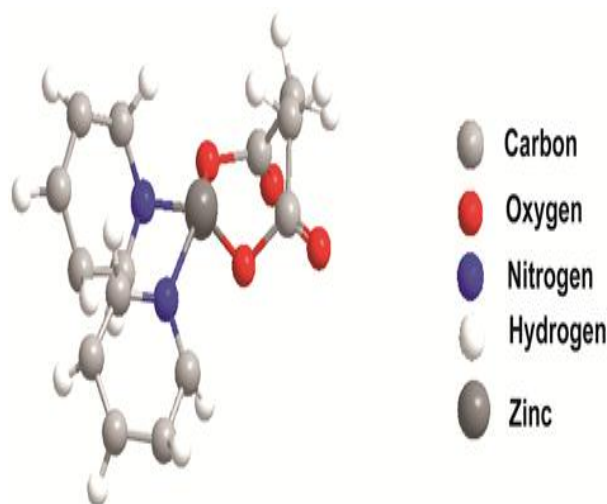
**Fig. 5:** Antifungal activity of the synthesized complexes.

## CONCLUSION

On the basis of all the above experimental measurement, we have offered octahedral geometry for Ni(II) compound but tetrahedral geometry for Cd(II) compound. The proposed structures of [Ni(DPA)(Py)<sub>4</sub>], and [Cd(DPA)(Py)<sub>2</sub>] have revealed in “Fig. 6” and “Fig. 7”. All the prepared complexes detected good antimicrobial activity. The tested mixed-ligand complexes displayed higher activities against fungi compared to bacteria.



**Fig. 6:** Probable 3-D structure of the complex [Ni(DPA)(Py)<sub>4</sub>].



**Fig. 7:** Probable 3-D structure of the complex [Cd(DPA)(Py)<sub>2</sub>].

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