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Synthesis and Reactivity in Inorganic and Metal-Organic Chemistry

Publication details, including instructions for authors and subscription information: <u>http://www.tandfonline.com/loi/lsrt19</u>

Synthesis, Structural Characterization, and Antibacterial Studies of Some Mixed-Ligand First Row d-Transition Metal Complexes

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To cite this article: Pragnesh K. Panchal & M. N. Patel (2004) Synthesis, Structural Characterization, and Antibacterial Studies of Some Mixed-Ligand First Row d-Transition Metal Complexes, Synthesis and Reactivity in Inorganic and Metal-Organic Chemistry, 34:7, 1277-1289, DOI: <u>10.1081/SIM-120039271</u>

To link to this article: http://dx.doi.org/10.1081/SIM-120039271

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SYNTHESIS AND REACTIVITY IN INORGANIC AND METAL-ORGANIC CHEMISTRY Vol. 34, No. 7, pp. 1277–1289, 2004

Synthesis, Structural Characterization, and Antibacterial Studies of Some Mixed-Ligand First Row d-Transition Metal Complexes

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ABSTRACT

Some mixed-ligand complexes of the transition metal ions Mn(II), Co(II), Ni(II), Cu(II), Zn(II), and Cd(II) with the potassium salt of salicylideneglycine and 2,2'-bipyridylamine or di(benzylidene)-1,8-diaminonaphthalene have been prepared. The coordination behavior of the mixed-ligand complexes was characterized on the basis of elemental analyses, IR, and electronic spectra. The magnetic measurements have been obtained by a vibrating sample magnetometer. It is observed that the mixed-ligand complexes of Mn(II), Co(II), Ni(II), Cu(II) are paramagnetic, while the Zn(II) and Cd(II) complexes are diamagnetic. All of these mixed-ligand complexes are colored crystalline solids. Octahedral geometries have been assigned to all of the prepared mixed-ligand complexes. The antibacterial activities of the ligands, the metal chlorides, the mixed-ligand complexes,

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DOI: 10.1081/SIM-120039271 Copyright © 2004 by Marcel Dekker, Inc. 0094-5714 (Print); 1532-2440 (Online) www.dekker.com

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the standard drug (tetracycline), and control (dimethyl sulfoxide) were tested on the pathogenic bacteria *S. typhi, E. coli*, and *Serratia marcescens* using the "disc diffusion method," and the results are discussed.

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Key Words: Mixed-ligand; Disc diffusion method; Schiff bases; 2,2'-Bipyridylamine.

INTRODUCTION

Schiff bases and their metal complexes have shown biological activity as carcinogenic^[1] and antitumor^[2] because of their specific structures. Amino acid Schiff base complexes derived from aldehydes are involved in a variety of biological processes such as catalysis of transamination, racemization, and carboxylation reactions.^[3,4] In continuation of earlier work,^[5] we prepared some new mixed-ligand complexes of the type [M(L)(bipy-amine)(H₂O)] or [M(L)(bendan)(H₂O)], where, M = Mn(II), Co(II), Ni(II), Cu(II), Zn(II), and Cd(II), KHL = potassium salt of salicylideneglycine and bipy-amine = 2,2'-bipyridylamine and bendan = *di*(benzylidene)-1,8-diamino-naphthalene. The physical properties were determined and are discussed herein. The suggested structure of the ligands are shown in Fig. 1.



Figure 1. Structures of the organic ligands used in this study.



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EXPERIMENTAL

Reagent and Solvents

All the chemicals used were of analytical grade. 2,2'-Bipyridylamine and 1,8-diaminonaphthalene were purchased from the Eastgate, White Lund, Morecambe, Lancaster, England. Salicylaldehyde, glycine, benzaldehyde, and metal chlorides were purchased from E. Merck (India) Limited, Mumbai. The organic solvents were purified by standard methods.^[6]

Preparation of Schiff Bases

Potassium Salt of Salicylideneglycine (KHL)

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A solution of potassium hydroxide (10 mmol, 0.56 g) in 30 mL absolute alcohol was added to glycine (10 mmol, 0.75 g). The mixture was stirred until all dissolved in the ethanol–KOH solution. Then an ethanolic solution (100 mL) of salicylaldehyde (10 mmol, 1.22 g) was quickly added. The reaction was allowed to complete by stirring at 50 °C for an hour. The product was crystallized by diffusion of excess absolute diethyl ether into the solution. The product was filtered, and dried in a vacuum desiccator. Yield: 1.59 g (63%), m.p.: 207 °C.

Di(benzylidene)-1,8-diaminonaphthalene (Bendan)

An ethanolic solution (100 mL) of 1,8-diaminonaphthalene (10 mmol, 1.58 g) and an ethanolic solution (100 mL) of benzaldehyde (20 mmol, 2.12 g) in the mole ratio 1:2 were mixed with constant stirring. Refluxing was carried out for 5 hr and then the solution was cooled in a refrigerator to 0-2 °C overnight. A fine-crystalline yellow product was obtained. The product was filtered and dried in air. Yield: 2.51 g (68%), m.p.: 235 °C.

Synthesis of the Mixed-Ligand Complexes

The preparation of the mixed-ligand complexes was carried out by mixing an aqueous (100 mL) solution of metal chloride (10 mmol) with a hot methanolic (100 mL) solution of the potassium salt of salicylideneglycine (10 mmol, 2.17 g) and 2,2'-bipyridylamine (10 mmol, 1.71 g) or di(benzylidene)-1,8-diaminonaphthalene (10 mmol, 3.34 g) in 1:1:1 mole ratio. The mixed-ligand complexes were formed upon heating the mixtures in a water bath for 2 hr

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at 50 °C. The mixtures were kept overnight at room temperature. The obtained crystals were removed by filtration, washed with water, ethanol, and dried in air. Elemental analyses, formula weight, colors, melting points and percentage yields data are presented in Table 1.

Physical Measurements

The magnetic moments have been obtained by a model 7304, vibrating sample magnetometer,^[7] Lake Shore, USA. The vibrating sample magnetometer reports the total magnetic moment, m, of a sample in emu. However, the end goal of the magnetic measurement is not the moment in emu, but to get the effective magnetic moment. Therefore, the magnetic moment can be converted to susceptibility units by realizing that 1 emu = 1 gauss cm³. The susceptibility of a sample has units of volume and is defined for paramagnetic material by the equation:

$$\chi(\text{cm}^3) = \frac{m(\text{emu})}{H(\text{oersted})}$$

The gram susceptibility χ_g , was calculated using the expression. Gram susceptibility:

$$\chi_{\rm g} = \frac{\chi({\rm cm}^3)}{{
m mass}}$$

The gram susceptibility was multiplied by the molecular weight of the sample to obtain the molar susceptibility, $\chi_{\rm M}$. A correction was applied for the diamagnetism of the ligands to obtain the corrected molar susceptibility $\chi'_{\rm M}$. The effective magnetic moment was calculated from the expression.

 $\mu_{\rm eff} = 2.84 (\chi'_{\rm M} \times T)^{1/2}$, where $T = {\rm absolute \ temperature \ (K)}$

Infrared spectra were recorded on a FT–IR Nicolet 400D spectrophotometer in KBr pellets. Carbon, hydrogen, and nitrogen were analyzed with a model 240 Perkin–Elmer elemental analyzer. The metal contents of the mixedligand complexes were analyzed by EDTA titration^[8] after decomposing the organic matter with a mixture of perchloric, sulfuric, and nitric acids (1:1.5:2.5). Themogravimetric analyses have been obtained by a model 5000\2960 SDTA, TA Instruments, USA. The TGA curves were obtained at a heating rate of 10 °C/min in N₂ atmosphere over the temperature range of 50–800 °C. The reflectance spectra of the mixed-ligand complexes were recorded in the range 1700–350 nm (as MgO discs) on a Beckman DK-2A spectrophotometer.



	T_{6}	ble I. Analyt	ical data of 1	the mixed-li	gand compl	exes. ^a			
				% Found	l (calcd.)			$\mu_{ m eff}$	
Compounds, empirical formula	Formula weight	Color	C	Н	z	Metal	M.p. (°C)	(B.M.) (calcd.)	Yield (%)
KHL, C ₉ H ₈ NO ₃ K	217.18	Orange	49.78	3.65	6.43	Ι	207	Ι	63
		yellow	(49.76)	(3.68)	(6.44)				
bendan, $C_{24}H_{18}N_2$	334.24	Yellowish	86.20	5.40	8.42	I	235	I	68
IO HVanima mid/ DaM	C1 1CV		(07.00)	(00.0) 131	(10.0)	12.07	~360	6 01	22
C.oH.oPy-annovar2001, C.oH.oMnN.O.	71.174	1 CITOM	(54.18)	(22.7)	(13.29)	(13.04)		(5.92)	C C
$[Co(L)(bipy-amine)(H_2O)],$	425.12	Brown	53.69	4.20	13.15	13.82	>360	4.03	35
$C_{19}H_{18}CoN_4O_4$			(53.67)	(4.23)	(13.17)	(13.86)		(3.87)	
$[Ni(L)(bipy-amine)(H_2O)],$	425.88	Yellowish	53.67	4.27	13.21	13.82	333	2.98	32
$C_{19}H_{18}N_4NiO_4$		green	(53.70)	(4.23)	(13.18)	(13.81)		(2.83)	
[Cu(L)(bipy-amine)(H ₂ O)],	429.73	Green	53.13	4.12	13.00	14.82	190	1.84	40
$C_{19}H_{18}CuN_4O_4$			(53.10)	(4.18)	(13.03)	(14.78)		(1.73)	
$[Zn(L)(bipy-amine)(H_2O)],$	431.58	Yellow	52.83	4.21	12.95	15.18	>360		48
$C_{19}H_{18}N_4O_4Zn$			(52.87)	(4.17)	(12.97)	(15.15)			
[Cd(L)(bipy-amine)(H ₂ O)],	478.60	Yellow	47.72	3.75	11.73	23.51	280		52
$C_{19}H_{18}CdN_4O_4$			(47.67)	(3.76)	(11.70)	(23.48)			
$[Mn(L)(bendan)(H_2O)],$	594.26	Brown	66.73	4.50	7.08	9.42	>360	6.03	43
$\mathrm{C}_{33}\mathrm{H}_{27}\mathrm{MnN}_{3}\mathrm{O}_{4}$			(66.69)	(4.54)	(1.06)	(9.40)		(5.92)	
[Co(L)(bendan)(H ₂ O)],	588.26	Brown	67.31	4.63	7.10	10.10	>360	4.02	32
$C_{33}H_{27}CoN_3O_4$			(67.37)	(4.59)	(7.14)	(10.01)		(3.87)	
$[Ni(L)(bendan)(H_2O)],$	588.02	Yellowish	67.34	4.50	7.19	9.93	340	3.01	35
$C_{33}H_{27}N_3NiO_4$		green	(67.40)	(4.59)	(7.14)	(86.98)		(2.83)	
								(con	tinued)

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				% Found	l (calcd.)			$\mu_{ m eff}$	
Compounds, empirical formula	Formula weight	Color	U	Н	z	Metal	M.p. (°C)	(B.M.) (calcd.)	Yield (%)
$[Cu(L)(bendan)(H_2O)],$	592.87	Green	06.99	4.61	7.07	10.75	198	1.82	38
$C_{33}H_{27}CuN_3O_4$			(66.85)	(4.55)	(7.08)	(10.70)		(1.73)	
$[Zn(L)(bendan)(H_2O)],$	594.72	Yellow	66.61	4.60	7.09	11.02	>360		40
$C_{33}H_{27}N_3O_4Zn$			(66.64)	(4.54)	(1.06)	(10.99)			
$[Cd(L)(bendan)(H_2O)],$	641.74	Yellow	61.79	4.24	6.59	17.50	300		45
$C_{33}H_{27}CdN_3O_4$			(61.75)	(4.20)	(6.54)	(17.52)			

Panchal and Patel



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RESULTS AND DISCUSSION

The formation of mixed-ligand complexes having 1:1:1 metal-ligand mole ratio is facilitated by loss of two protons from the salicylideneglycinate ligand. All the mixed-ligand complexes are insoluble in water, methanol, and dimethyl formamide, but are soluble in dimethyl sulfoxide. They possess high melting points, are colored, and are stable in air. The general equations for the formation of the mixed-ligand complexes are shown below.

$$\begin{split} \text{MCl}_2 \cdot n\text{H}_2\text{O} + \text{KHL} + \text{bipy-amine} &\longrightarrow [\text{M}(\text{L})(\text{bipy-amine})(\text{H}_2\text{O})] \\ &+ \text{KCl} + \text{HCl} + (n-1)\text{H}_2\text{O} \end{split} \tag{1} \end{split} \tag{1} \\ \\ \text{MCl}_2 \cdot n\text{H}_2\text{O} + \text{KHL} + \text{bendan} &\longrightarrow [\text{M}(\text{L})(\text{bendan})(\text{H}_2\text{O})] + \text{KCl} \\ &+ \text{HCl} + (n-1)\text{H}_2\text{O} \end{aligned}$$

IR Spectra

The spectra of the mixed-ligand complexes exhibit a broad band in the region 3200-3300 cm⁻¹ due to ν (O-H) and other bands at ~850 cm⁻¹ and \sim 715 cm⁻¹ due to the rocking and wagging mode of the OH group. The presence of the latter band indicates the coordination nature of the water molecule.^[9] The ν (C=N) frequency of 2,2'-bipyridylamine occurs at 1585 cm^{-1} . This band shifts to higher energy by $\sim 35-45 \text{ cm}^{-1}$ in the mixed-ligand complexes indicating the bidentate (N,N-pyridyl nitrogen) coordination of this heterocyclic ligand.^[10] In the Schiff base the strong-intensity band due to ν (C=N) at 1645 cm⁻¹ is shifted to lower frequency by about $10-20 \text{ cm}^{-1}$ in the case of the mixed-ligand complexes indicating the coordination of the azomethine^[11] group through its nitrogen. The mode of coordination is further supported by the presence of a new band in the \sim 530 cm⁻¹ region due to ν (M–N) vibrations.^[12] The mixed-ligand complexes exhibit two characteristic strong and broad bands in the $1400-1600 \text{ cm}^{-1}$ region, attributed to the $v_{as}(\text{COO}^{-})$ (~1585 cm⁻¹) and $v_{\rm s}({\rm COO}^{-})$ (~1390 cm⁻¹) stretching vibrations of the coordinated carboxylate groups.^[13] The mode of coordination is further supported by the presence of a new band in the $\sim 420 \,\mathrm{cm}^{-1}$ region due to the $\nu(\mathrm{M-O})_{\mathrm{carbo}}$ vibration.^[12] The ν (O–H) band identified in the free ligand was absent in the IR spectra of all the mixed-ligand complexes. This is attributed to the deprotonation of the phenolic group on complexation.^[14] Further, a shift in the ν (C–O) band by $10-20 \text{ cm}^{-1}$ to higher frequency confirms coordination through the phenolic oxygen in the ligand.^[15] In the mixed-ligand complexes a new band



observed in the $\sim 470 \text{ cm}^{-1}$ region is due to the $\nu(M-O)_{\text{phenolic}}$ vibration.^[12] The sharp bands in the range 750–780 cm⁻¹ and 1525–1535 cm⁻¹ are due to aromatic $\nu(C-H)$ and $\nu(C=C)$, respectively. The absorption in the 1160–1170 cm⁻¹ range is attributed to $\nu(C-N)$.

Magnetic Moments and Electronic Spectra

The information regarding geometry of the mixed-ligand complexes is obtained from their electronic spectral data and magnetic moments.

The Co(II) mixed-ligand complexes show magnetic moment values of 4.03 and 4.01 B.M., respectively, at room temperature. These high values of the magnetic moments and the stoichiometries suggest a coordination number of six for the central Co(II) ion and an octahedral geometry. The electronic spectra of the mixed-ligand complexes are also consistent with their octahedral environment around the Co(II) ion. The Co(II) mixed-ligand complexes exhibit three bands at ~9,200 (ν_1), ~18,000 (ν_2), and ~19,000 (ν_3) cm⁻¹, which may be assigned to the transitions ${}^{4}T_{1g}(F) \rightarrow {}^{4}T_{2g}(F)$, ${}^{4}T_{1g}(F) \rightarrow {}^{4}A_{2g}(F)$, and ${}^{4}T_{1g}(F) \rightarrow {}^{4}T_{1g}(P)$ expected for an octahedral structure. ture.^[16] The electronic spectra of the Ni(II) mixed-ligand complexes exhibit absorption bands at ~10,500 (ν_1), ~17,500 (ν_2), and ~24,200 (ν_3) cm⁻¹ attributed to ${}^{3}A_{2g}(F) \rightarrow {}^{3}T_{2g}(F)$, ${}^{3}A_{2g}(F) \rightarrow {}^{3}T_{1g}(F)$, and ${}^{3}A_{2g}(F) \rightarrow {}^{3}T_{1g}(P)$ transitions, respectively, in an octahedral geometry. The values of the ligand field parameters for the Co(II) and Ni(II) complexes are in the range reported for an octahedral structure^[17] and are presented in Table 2. The values of the</sup> magnetic moment (2.98 and 3.01 B.M.) may be taken as additional evidence for their octahedral structure.^[18] The Cu(II) mixed-ligand complexes exhibit magnetic moments of 1.84 and 1.82 B.M., respectively. These values are close to the spin-allowed values expected for a S = 1/2 system (1.73 B.M.) and may be indicative of an octahedral geometry around the Cu(II) ions. The Cu(II) mixed-ligand complexes display a broad band at $\sim 15,000 \text{ cm}^{-1}$ due to the ${}^{2}E_{g} \rightarrow {}^{2}T_{2g}$ transition.^[19] The electronic spectra of the Mn(II) mixed-ligand complexes exhibit three weak absorption bands for the transitions ${}^{6}A_{1g} \rightarrow {}^{4}T_{1g}$ (~15,000 cm⁻¹) (ν_{1}), ${}^{6}A_{1g} \rightarrow {}^{4}T_{2g}$ (~20,000 cm⁻¹) (ν_{2}), and ${}^{6}A_{1g} \rightarrow {}^{4}E_{g}$, ${}^{4}A_{1g}$ (~25,000 cm⁻¹) (ν_{3}). These absorptions are consistent with an octahedral geometry around Mn(II) in all of the mixedligand complexes. The magnetic moment values of the Mn(II) mixed-ligand complexes are 6.01 and 6.03 B.M., suggesting an octahedral geometry.^[20] The Zn(II) and Cd(II) mixed-ligand complexes are diamagnetic as expected for d¹⁰ systems. The electronic spectra of the mixed-ligand complexes were recorded in the solid state. Therefore, the ε values of all the mixed-ligand complexes were not determined.

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β

В

 ν_2/ν_1

 v_3

 \mathbf{V}_2

 ν_1

Mixed-ligand complexes

Mixed Ligand Transition Metal Complexes

Table 2. Electronic parameters of the Co(II) and Ni(II) complexes.^a

Observed bands (cm⁻¹)

the Co(II) and Ni(II) complexes have been calculated using the secular equations given by E. König. For Co(II) complexes: 10 Dq = $1/2[(2\nu_1 - \nu_3) + (\nu_3^2 + \nu_1\nu_3 - \nu_1^2)^{1/2}]$; 15B = $\nu_3 - 2\nu_1 + 10$ Dq; $\beta = B/B0$ [Bo(free ion) = 971]; $\beta^0 = (1 - \beta) \times 100$. For Ni(II) *Note:* The ligand field splitting energy (10 Dq), interelectronic repulsion parameter (*B*), and covalency factor (nephelauxetic ratio) (β) for $\begin{array}{c} 0.79 \\ 0.75 \\ 0.77 \\ 0.76 \end{array}$ complexes: 10 Dq = ν_1 ; 15B = $(\nu_2 + \nu_3) - 3\nu_1$; $\beta = B/B_0$ [Bo(free ion) = 1030]; $\beta^0 = (1 - \beta) \times 100$. 7770 751 791 2.00 1.70 2.00 1.72 19,050 24,975 18,920 25,040 18,150 17,425 18,220 17,700 9,075 10,250 9,110 10,290 [Co(L)(bipy-amine)(H₂O)] [Ni(L)(bipy-amine)(H₂O)] $[Co(L)(bendan)(H_2O)]$ [Ni(L)(bendan)(H₂O)]

^aBipy-amine = 2,2'- bipyridylamine; bendan = di(benzylidene)-1,8-diaminonaphthalene.





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Thermogravimetric Analyses

Thermogravimetric analyses studies were carried out on the mixed-ligand complexes in the temperature range of 50-800 °C. All of the mixed-ligand complexes show a loss in weight corresponding to one water molecule in the range $\sim 150-180$ °C indicating that these water molecules are coordinated to the metal ion.^[21] In the temperature range 180-800 °C, the ligand molecules are lost. In all cases, the final products are metal oxides. These results are in good accordance with the proposed mixed-ligand complex compositions. From the above discussion, suggested structures for the mixed-ligand complexes are shown in Fig. 2.

Antibacterial Activity

The ligands, the metal chlorides, and the mixed-ligand complexes have been tested for their effects on the growth of the microbial cultures of *S. typhi, E. coli, Serratia marcescens* using the disc diffusion method.^[22,23] A stock solution of 250 µg/mL was made by dissolving 25 mg of each compound in dimethyl sulfoxide (100 mL). Filter paper, Whatmann No. 4 discs of 6 mm diameter, were soaked in solutions (20 µL) of the test compounds and after drying off the solvent were placed on a nutrient agar plate containing lawn cultures of different bacteria. The plates were incubated at an optimum growth temperature of 37 °C. The zone of inhibition (in mm) around the discs was measured after 24 hr. All experiments were performed in triplicate and the average of the three trials was considered as the zone of inhibition. Tetracycline was used as a standard drug. The mixed-ligand complexes are soluble in dimethyl sulfoxide. Therefore, the growth was compared with dimethyl



Figure 2. Proposed structures of the mixed-ligand complexes.

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sulfoxide as the control. For the control experiments equivalent volumes of solvents without added test compounds were applied to the paper discs and the zone of inhibition (in mm) measured. Results are expressed as zone of inhibition and percentage inhibition vs. control. The results are presented in Table 3. From this data it is observed that the mixed-ligand complexes show higher activities against bacteria compared to the control (blank), the ligands, and metal salts. The mixed-ligand complexes show moderate activities as compared to the standard drug (tetracycline). The results suggest that the changes in structure due to the coordination in the mixed-ligand complexes decrease the growth of microorganisms relative to the metal ion,

Table 3. Antibacterial activities of the mixed-ligand complexes.^a

	Zone of inhibition in mm (% inhibition)			
Compounds	S. typhi	E. coli	Serratia marcesens	
DMSO (control)	9	12	15	
Tetracycline	18 (100)	21 (75)	26 (73)	
KHL	10 (11)	13 (08)	17 (13)	
bipy-amine	10 (11)	15 (25)	16 (06)	
bendan	11 (22)	14 (17)	17 (13)	
$MnCl_2 \cdot 4H_2O$	10 (11)	13 (08)	16 (06)	
$CoCl_2 \cdot 6H_2O$	10 (11)	13 (08)	17 (13)	
$NiCl_2 \cdot 6H_2O$	11 (22)	14 (17)	16 (06)	
$CuCl_2 \cdot 2H_2O$	11 (22)	13 (08)	17 (13)	
ZnCl ₂	10 (11)	14 (17)	17 (13)	
$CdCl_2 \cdot 2H_2O$	11 (22)	14 (17)	17 (13)	
[Mn(L)(bipy-amine)(H ₂ O)]	12 (33)	16 (33)	19 (27)	
[Co(L)(bipy-amine)(H ₂ O)]	12 (33)	16 (33)	23 (53)	
[Ni (L)(bipy-amine)(H ₂ O)]	13 (44)	17 (42)	21 (40)	
[Cu(L)(bipy-amine)(H ₂ O)]	13 (44)	18 (50)	23 (53)	
[Zn(L)(bipy-amine)(H ₂ O)]	12 (33)	17 (42)	22 (47)	
[Cd(L)(bipy-amine)(H ₂ O)]	13 (44)	17 (42)	21 (40)	
[Mn(L)(bendan)(H ₂ O)]	13 (44)	17 (42)	20 (33)	
$[Co(L)(bendan)(H_2O)]$	12 (33)	17 (42)	22 (47)	
[Ni(L)(bendan)(H ₂ O)]	12 (33)	16 (33)	23 (53)	
$[Cu(L)(bendan)(H_2O)]$	13 (44)	18 (50)	21 (40)	
$[Zn(L)(bendan)(H_2O)]$	13 (44)	18 (50)	21 (40)	
[Cd(L)(bendan)(H ₂ O)]	12 (33)	16 (33)	23 (53)	

^aKHL, potassium salt of salicylideneglycine; bipy-amine, 2,2'-bipyridylamine; bendan, *bis*(benzylidene)-1,8-diaminonaphthalene.



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the ligands, and control. Hence, the complexation increases the antibacterial activity. The patterns of activity are complex, and no clear trend can be ascertained.

ACKNOWLEDGMENTS

We wish to express our sincere thanks to Prof. R. M. Patel, Head, Department of Chemistry and Prof. M. Dutta, Head, Department of Biosciences, Sardar Patel University, Vallabh Vidyanagar, India, for providing the necessary laboratory facilities.

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Received December 15, 2003 Accepted March 23, 2004

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