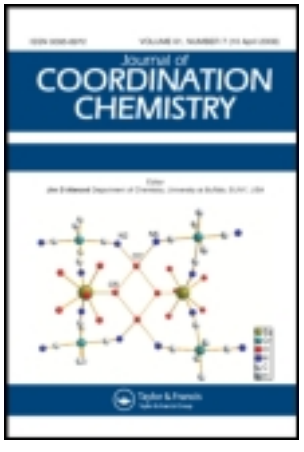

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Synthesis, characterization, and biological activity of complexes derived from E-N′-(3,4,5-trimethoxybenzylidene)benzofuran-2-carbohydrazide and ortho-phenylenediamine/ 2,6-diaminopyridine

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Synthesis, characterization, and biological activity of complexes derived from E-*N*′-(3,4,5-trimethoxybenzylidene)benzofuran-2-carbohydrazide and *ortho*-phenylenediamine/ 2,6-diaminopyridine

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Mixed ligand complexes of the type $MLL'Cl_2$ (where M = Co(II), Ni(II), Cu(II), Zn(II), Cd(II), and Hg(II), L = ligand derived from the reaction between benzofuran-2-carbohydrazide and 3,4,5-trimethoxybenzaldehyde (TMeOBFC), and L′ = *ortho*-phenylenediamine (opd)/2,6-diaminopyridine (2,6-dap)) have been synthesized. The complexes have been characterized by analytical data, IR, UV-Vis, ^{1}H NMR, DART-MS and ESR spectral data, and magnetic studies. Molar conductance values indicate that the complexes are non-electrolytic in DMF. Antibacterial and antifungal activities of the ligands and complexes have been screened against bacteria *Escherichia coli* and *Staphylococcus aureus* and against fungi *Aspergillus niger* and *Aspergillus flavus*.

Keywords: Benzofuran Schiff bases; Mixed ligand complexes; Spectral studies; Biological activity

1. Introduction

Schiff bases have many applications in food industry, dye industry, analytical chemistry, catalysis, fungicidal, agrochemical, and biological activities. Schiff bases exhibit a broad spectrum of pharmacological and biological properties, such as analgesics, anticancer agents, anti-inflammatory, etc., which may be due to the azomethine linkage [1]. Schiff-base complexes containing nitrogen and oxygen donors play an important role in biological systems and represent models for metalloproteins and metalloenzymes that catalyze the reduction of nitrogen and oxygen [2]. Schiff bases show the ability to complex with many metal ions [3] and such complexes play an important role in coordination chemistry, enzyme reactions, and molecular architectures [4, 5]. Metal complexes play an essential role in agriculture, pharmaceutical, and industrial chemistry [6]. Many drugs possess modified toxicological and pharmacological properties when administered as metal complexes [7, 8]. Metal complexes with

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mixed ligands provide new materials with useful properties such as magnetic exchange [9], electrical conductivity [10], non-linear optical activity [11], and many other activities [12]. Mixed ligand complexes are involved in the exchange and transport mechanism of trace metal ions in the human body [13].

Benzofuran compounds are abundant in nature, particularly among plants. Natural products possessing benzofuran have useful pharmacological properties [14]. Baker's yeast contains a benzofuran derivative which acts as an antioxidant preventing hemorrhaging liver necrosis in rats and hemolysis of red cells during vitamin-E deficiency [15]. Some benzofuran derivatives such as 2-acetylbenzofuran and 2-nitrobenzofuran are well-known biodynamic agents possessing various pharmacological properties [16–18]. Amiodarone hydrochloride, used as an antiarrhythmic drug [19], contains a 2,3-disubstituted benzofuran.

This work deals with the synthesis and characterization of Co(II), Ni(II), Cu(II), Zn(II), Cd(II), and Hg(II) complexes with primary ligand derived from benzofuran-2-carbohydrazide and 3,4,5-trimethoxybenzaldehyde (TMeOBFC) and *ortho*-phenylenediamine (opd) or 2,6-diaminopyridine (2,6-dap). The Schiff base and metal complexes are screened for antibacterial and antifungal activities.

2. Experimental

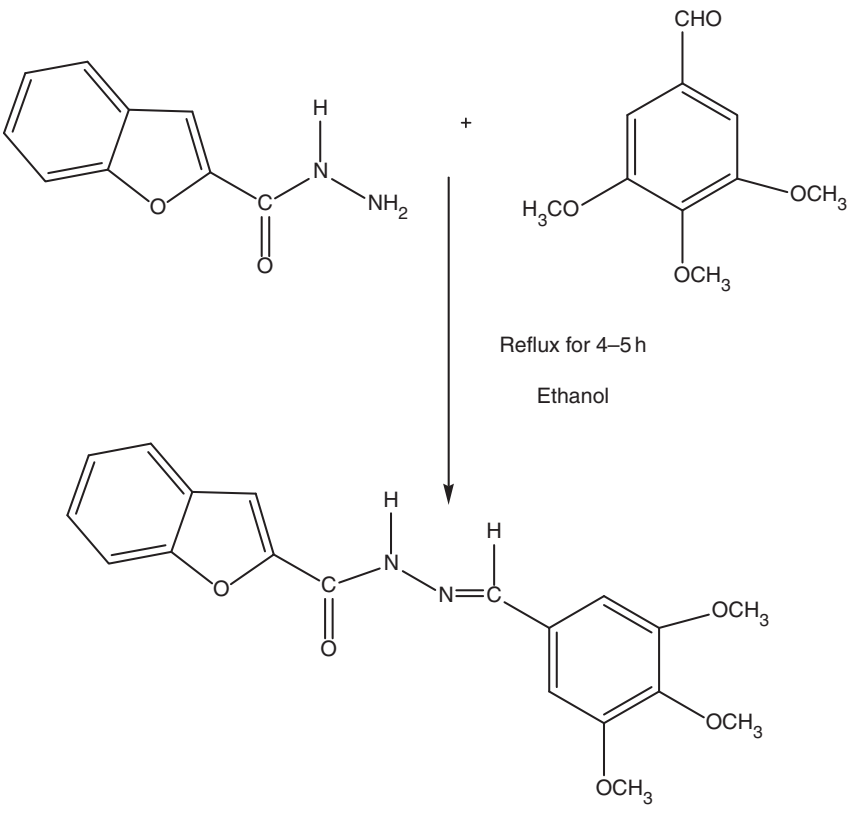
Chemicals used in this work were of analytical reagent (AR) grade and of highest purity available. Benzofuran-2-carbohydrazide was synthesized according to the literature procedure [20]. Metal and chloride contents were determined as per Vogel's procedure [21].

2.1. *Synthesis of (E)-N′-(3,4,5-trimethoxybenzylidene)benzofuran-2-carbohydrazide [TMeOBFC]*

A solution of benzofuran-2-carbohydrazide (1.76 g, 0.01 mol) in ethanol (25 mL) was added to 3,4,5-trimethoxybenzaldehyde (1.96 g, 0.01 mol) in ethanol (20 mL) and refluxed on a water bath for 4–5 h. On the partial removal of solvent and cooling to room temperature, the Schiff base was separated as yellowish crystalline solid, which was then filtered, washed with ethanol, and recrystallized from ethanol. The synthesis of the Schiff base is shown in scheme 1.

2.2. *Synthesis of metal(II) mixed ligand complexes*

An ethanolic solution (20 mL) of 0.01 mol of the appropriate metal chloride, MCl_2 (M = Co, 2.378 g; M = Ni, 2.377 g; M = Cu, 1.705 g; M = Zn, 1.363 g; M = Cd, 2.013 g; M = Hg, 2.715 g), was added to an ethanolic solution (30 mL) of the Schiff base, TMeOBFC (3.54 g, 0.01 mol). An ethanolic solution (20 mL) of 0.01 mol of *ortho*-phenylenediamine (1.08 g) or 2,6-diaminopyridine (1.09 g) was added to the previous solution and the reaction mixture was refluxed for 3 h on a water bath, and then aqueous alcoholic solution of sodium acetate was added to adjust the pH to 6–7 and further refluxed for an hour. The precipitated complexes were filtered and washed with



Scheme 1. Synthesis of TMeOBFC ($C_{19}H_{18}O_5N_2$ [TMeOBFC]: mol. wt = 354, m.p. = 233°C, yield = 72%).

distilled water, then with alcohol, and finally dried in vacuum over fused calcium chloride (yield = 55–60%).

2.3. *Physical measurements*

Carbon, hydrogen, and nitrogen analyses were carried out microanalytically on a Perkin-Elmer 240C model at the Central Drug Research Institute (CDRI), Lucknow. IR spectra of the Schiff base and complexes were recorded in KBr pellets from 4000 to 350 cm^{-1} on a Perkin-Elmer 783 FT-IR spectrophotometer. Electronic spectra of the Co(II), Ni(II), and Cu(II) complexes were recorded on an Elico-SL-164 double beam UV-Vis spectrophotometer from 200 to 1100 nm in DMF (10^{-3} mol L^{-1}). ^{1}H NMR spectra were recorded in DMSO-d_6 on a Bruker 300 MHz spectrophotometer using TMS as an internal standard. The ESR spectrum of the Cu(II) complex in polycrystalline state was recorded on a Varian-E-4X band EPR spectrophotometer using TCNE as "*g*" marker ($g = 2.00277$) at room temperature. Direct analysis in real time-mass spectra (DART-MS) were recorded on a JEOL-AccuTOF JMS-T100LC mass spectrometer having a DART source. Dry helium was used with 4 L min^{-1} flow rate for ionization at 350°C, the orifice 1 set at 28 V. Molar conductivity measurements

were recorded on an Elico CM-180 conductivity bridge in DMF (10^{-3} mol L^{-1}) using a dip-type conductivity cell fitted with a platinum electrode and magnetic susceptibility measurements were made at room temperature on a Gouy balance using $Hg[Co(NCS)_4]$ as the calibrant.

2.4. *Biological activities*

The Schiff base and complexes have been studied for antibacterial and antifungal activities by agar diffusion in DMF against *Escherichia coli* and *Staphylococcus aureus* bacterial and *Aspergillus niger* and *Aspergillus flavus* fungi species [22, 23].

2.5. *Antibacterial screening*

Media used: peptone 10 g, NaCl 10 g, yeast extract 5 g, and agar 20 g in 1000 mL of distilled water.

Initially, stock cultures of *E. coli* and *S. aureus* were revived by inoculating in broth media and grown at 37°C for 18 h. The agar plates of the above media were prepared and wells were made in the plate. Each plate was inoculated with 18-h-old cultures and spread evenly on the plate. After 20 min, the wells were filled with test solutions. Gentamycin was used as the standard antibacterial drug. All the plates were incubated at 37°C for 24 h and the diameters of inhibition zones were noted.

2.6. *Antifungal screening*

Media used: potato dextrose agar (PDA). 250 g of peeled potatoes were boiled for 20 min and squeezed and filtered. 20 g of dextrose was added to this filtrate and the volume was made up to 1000 mL by distilled water.

Initially, stock cultures of *A. flavus* and *A. niger* were revived by inoculating in broth media and grown at 37°C for 48 h. The agar plates of the above media were prepared and wells were made in the plate. Each plate was inoculated with 18-h-old cultures and spread evenly on the plate. After 20 min, the wells were filled with test solutions. Fluconazole was used as the standard antifungal drug. All the plates were incubated at 37°C for 48 h and the diameters of inhibition zones were noted.

3. Results and discussion

Analytical data show that all the complexes have 1 : 1 : 1 (metal : primary ligand : secondary ligand) stoichiometry (table 1). The molar conductance values (11.74–20.23 $Ohm^{-1}\,cm^2\,mol^{-1}$) are too low to account for the dissociation of the complexes in DMF, indicating the presence of non-electrolytes [24]. All the complexes are light in color, stable, non-hygroscopic, and possess high melting points. The complexes are insoluble in common organic solvents but soluble in DMF and DMSO.

Table 1. Elemental analysis, molar conductance, and magnetic susceptibility data for TMeOBFC and complexes.

Molecular formula of Schiff base/complexes	Mol. weight	C%		H%		N%		M%		Cl%		Λ_M[a]	μ_{eff} (BM)
		Found	Calcd	Found	Calcd	Found	Calcd	Found	Calcd	Found	Calcd		
$C_{19}H_{18}O_5N_2$ TMeOBFC (L)	354	64.21	64.41	4.82	5.08	7.76	7.91	–	–	–	–	–	–
[CoL(opd)Cl_2]	592.34	50.33	50.69	4.23	4.42	9.22	9.46	9.72	9.95	11.78	11.97	14.21	4.81
[NiL(opd)Cl_2]	592.09	50.56	50.71	4.28	4.43	9.18	9.46	9.68	9.91	11.65	11.98	20.23	2.98
[CuL(opd)Cl_2]	596.95	50.09	50.30	4.18	4.39	9.06	9.39	10.23	10.65	11.56	11.88	18.16	1.78
[ZnL(opd)Cl_2]	598.79	49.92	50.15	4.16	4.38	9.14	9.36	10.66	10.92	11.68	11.84	16.09	–
[CdL(opd)Cl_2]	645.81	46.23	46.49	3.88	4.06	8.46	8.68	17.16	17.41	10.75	10.98	14.18	–
[HgL(opd)Cl_2]	733.99	40.72	40.91	3.23	3.57	7.41	7.63	27.09	27.33	9.41	9.66	17.13	–
[CoL(2,6-dap)Cl_2]	593.32	48.29	48.58	4.06	4.25	11.62	11.80	9.72	9.93	11.68	11.95	15.28	4.92
[NiL(2,6-dap)Cl_2]	593.08	48.35	48.60	4.03	4.25	11.56	11.81	9.68	9.90	11.72	11.96	14.65	3.11
[CuL(2,6-dap)Cl_2]	597.94	48.05	48.21	3.89	4.21	11.48	11.71	10.48	10.63	11.65	11.86	13.86	2.01
[ZnL(2,6-dap)Cl_2]	599.78	47.89	48.06	3.92	4.20	11.42	11.68	10.72	10.90	11.59	11.82	12.82	–
[CdL(2,6-dap)Cl_2]	646.80	44.21	44.57	3.63	3.90	10.65	10.83	17.05	17.38	10.71	10.96	11.74	–
[HgL(2,6-dap)Cl_2]	734.98	39.06	39.22	3.21	3.43	9.21	9.53	27.11	27.29	9.34	9.65	12.24	–

[a]Molar conductance values are given in $Ohm^{-1} cm^2 mol^{-1}$.

3.1. *Magnetic properties*

Magnetic susceptibility measurements at room temperature exhibit paramagnetism for Co(II), Ni(II), and Cu(II) complexes; μ_{eff} values are listed in table 1. The Co(II) complexes exhibit magnetic moments of 4.81 and 4.92 BM, suggesting an octahedral geometry [25, 26]. The Ni(II) complexes show magnetic moment values of 2.98 and 3.11 BM, slightly higher than the spin only value (2.83 BM), indicating an octahedral environment around Ni(II) [27, 28]. The observed magnetic moments for the Cu(II) complexes are 1.78 and 2.01 BM, suggesting a distorted octahedral geometry for Cu(II) complexes [29]. The primary and secondary ligands are bidentate in all complexes.

3.2. *Electronic spectral studies*

Electronic spectra of the Co(II), Ni(II), and Cu(II) complexes were recorded for freshly prepared solution in DMF (10^{-3} mol L^{-1}) at room temperature (table 2). Spectra of Co(II) complexes show two bands at 15,314, 19,763 cm^{-1} and at 16,155, 20,450 cm^{-1}, assignable to $^4T_{1g}\,(F) \rightarrow {}^4A_{2g}\,(F)$ (ν_2) and $^4T_{1g}\,(F) \rightarrow {}^4T_{2g}\,(P)$ (ν_3) transitions in an octahedral environment [30]. The ν_1 band could not be observed; however, ν_1 was calculated using an equation suggested by Underhill and Billing [31].

Ni(II) complexes exhibit two bands at 15,983, 26,523 cm^{-1} and at 15,423, 26,423 cm^{-1} assignable to $^3A_{2g}\,(F) \rightarrow {}^3T_{1g}\,(F)$ (ν_2) and $^3A_{2g}\,(F) \rightarrow {}^3T_{1g}\,(P)$ (ν_3) transitions, respectively, in an octahedral environment. The ν_1 could not be observed due to limited range of the instrument used. However, it is calculated by using band-fitting procedure [31].

The Cu(II) complexes exhibit a single broad asymmetric band at 13,141–17,560 cm^{-1}. The broadness of the band indicates that the three expected transitions $^2B_{1g} \rightarrow {}^2A_{1g}$ (ν_1), $^2B_{1g} \rightarrow {}^2B_{2g}$ (ν_2), and $^2B_{1g} \rightarrow {}^2E_g$ (ν_3) are similar in energy and give rise to only one broad band. The broadness of the band may be due to dynamic Jahn–Teller distortion. All these data suggest a distorted octahedral geometry around Cu(II).

The octahedral geometry [32] is further supported by the values of ligand field parameters such as Racah inter-electronic repulsion parameter (B'), ligand-field splitting energy ($10\,D_q$), covalency factor (β), and ligand-field stabilization energy (LFSE) [33]. The B' values for the complexes were lower than the free ion values,

Table 2. Electronic spectral bands and ligand field parameters of the Co(II), Ni(II), and Cu(II) complexes in DMF (10^{-3} mol L^{-1}).

Complexes	Transitions in cm^{-1}			D_q (cm^{-1})	B' (cm^{-1})	β	β%	ν_2/ν_1	LFSE (kcal)
	ν_1[a]	$\nu_{2(\varepsilon)}$[b]	$\nu_{3(\varepsilon)}$[b]						
[CoL(opd)Cl$_2$]	7114	15314 (260)	19763 (440)	820	915	0.942	5.767	2.152	15.685
[NiL(opd)Cl$_2$]	9860	15983 (250)	26523 (580)	986	860	0.827	17.308	1.621	33.806
[CuL(opd)Cl$_2$]		13223–16821 (310)		1502	–	–	–	–	25.749
[CoL(2,6-dap)Cl$_2$]	7515	16155 (360)	20450 (480)	864	938	0.966	3.399	2.150	14.811
[NiL(2,6-dap)Cl$_2$]	9390	15423 (270)	26423 (560)	939	911	0.876	12.404	1.642	32.194
[CuL(2,6-dap)Cl$_2$]		13141–17560 (340)		1535	–	–	–	–	26.314

[a]Calculated values.
[b]Molar extinction coefficient values are given in parenthesis in units of L mol^{-1} cm^{-1}.

indicating orbital overlap and delocalization of d-orbitals. The β values obtained are less than unity, suggesting considerable covalent character for the metal–ligand bonds. The β value for Ni(II) complexes are less than the Co(II) complexes, indicating more covalency of M–L bonds [34].

3.3. *IR spectral studies*

IR spectra of the ligand and complexes are presented in table 3. The spectra are complex and it is not possible to assign all the bands.

TMeOBFC shows medium broad bands at 3448 and 3216 cm^{-1} due to ν(NH) of hydrazine. These bands are assigned to ν_{asym} and ν_{sym} stretching vibrations of secondary amide and shift to a higher wavenumber by 5–15 cm^{-1} in the complexes, showing non-involvement of "N" of NH in bonding with metal ions [35]. The strong intensity bands at 1695 and 1626 cm^{-1} are assigned to ν(C=O) and ν(C=N), respectively, in free ligand. These bands shift to a lower wavenumber by about 20–45 cm^{-1} in the complexes, indicating coordination to metal through the oxygen of C=O and nitrogen of –N=CH– [36, 37]. The band at 1031 cm^{-1} is assigned to ν(N–N) of hydrazine, which shifts to a higher wavenumber by 10–15 cm^{-1} in metal complexes, confirming bonding through one nitrogen of hydrazine [38].

ortho-Phenylenediamine shows broad bands at 3365 and 3360 cm^{-1} due to ν(NH) of primary amine [39]. In all metal complexes the band shifts to a lower wavenumber, 3335–3295 cm^{-1}, indicating the involvement of nitrogen of amine in bonding with metal ion.

The 2,6-diaminopyridine shows broad bands at 3392 and 3386 cm^{-1} due to ν(NH) of primary amine. In metal complexes the band shifts to a lower wavenumber, 3362–3331 cm^{-1}, indicating the involvement of amino nitrogen in bonding with metal. An absorption at 1578 cm^{-1} can be assigned to ν(C=N) of pyridine in 2,6-diaminopyridine. There is not much change in the position of this band in the metal complexes, indicating non-involvement of pyridine nitrogen in bonding with metal [40].

Metal–ligand vibrations are difficult to assign on an empirical basis since they are sensitive to both ligand and metal. However, non-ligand new bands at 530–515 and 408–460 cm^{-1} are assigned to ν(M–O) and ν(M–N) [41, 42]. The weak intensity bands at 340–305 cm^{-1} are assigned to ν(M–Cl) vibrations.

Table 3. IR frequencies (in cm^{-1}) of TMeOBFC and metal complexes.

Schiff base/complexes	ν(C=O)	ν(C=N)	ν(N–N)	ν(M–O)	ν(M–N)	ν(M–Cl)
TMeOBFC (L)	1695	1626	1031	–	–	–
[CoL(opd)Cl_2]	1672	1598	1042	526	432	320
[NiL(opd)Cl_2]	1675	1589	1046	519	416	326
[CuL(opd)Cl_2]	1666	1606	1041	530	408	318
[ZnL(opd)Cl_2]	1658	1601	1043	523	445	340
[CdL(opd)Cl_2]	1667	1592	1045	516	450	332
[HgL(opd)Cl_2]	1656	1594	1044	528	428	315
[CoL(2,6-dap)Cl_2]	1670	1590	1043	524	456	336
[NiL(2,6-dap)Cl_2]	1659	1581	1042	518	441	323
[CuL(2,6-dap)Cl_2]	1662	1604	1046	515	460	305
[ZnL(2,6-dap)Cl_2]	1650	1596	1041	527	423	328
[CdL(2,6-dap)Cl_2]	1671	1587	1044	520	438	316
[HgL(2,6-dap)Cl_2]	1674	1592	1045	517	421	321

Table 4. ^{1}H NMR spectral data of TMeOBFC and Zn(II) and Cd(II) complexes [chemical shifts in δ (ppm)].

Ligands/complexes	–CONH–	–N=CH–	Aromatic protons	Methoxy protons
[TMeOBFC]	12.13 (s, 1H)	8.44 (s, 1H)	6.94–7.83 (m, 7H)	3.29, 3.67, 3.91 (s, 9H)
[ZnL(opd)Cl_2]	12.60 (s, 1H)	8.74 (s, 1H)	6.33–7.93 (m, 15H)	3.32, 3.70, 3.90 (s, 9H)
[CdL(opd)Cl_2]	12.51 (s, 1H)	8.71 (s, 1H)	6.35–7.89 (m, 15H)	3.31, 3.68, 3.89 (s, 9H)
[ZnL(2,6-dap)Cl_2]	12.47 (s, 1H)	8.64 (s, 1H)	7.01–7.88 (m, 14H)	3.31, 3.72, 3.89 (s, 9H)
[CdL(2,6-dap)Cl_2]	12.38 (s, 1H)	8.68 (s, 1H)	7.03–7.86 (m, 14H)	3.29, 3.69, 3.90 (s, 9H)

3.4. *^{1}H NMR spectral studies*

^{1}H NMR spectra of [TMeOBFC] and the Zn(II) and Cd(II) complexes were recorded in DMSO-d_6 (table 4).

The signal at δ (12.13) (s, 1H) is assigned to amide proton (–CONH–) and the signal at δ (8.44) (s, 1H) is assigned to azomethine proton (–N=CH–) in [TMeOBFC]. Signals due to (−CONH–) are shifted downfield in the spectra of Zn(II) δ (12.60, 12.47) (s, 1H) and Cd(II) δ (12.51, 12.38) (s, 1H) complexes, indicating the coordination of the oxygen of –CONH– with metal. The azomethine protons shift downfield δ (8.64–8.74) (s, 1H), supporting the coordination of nitrogen of –N=CH– with metal. The aromatic protons at 6.94–7.83 shift downfield in the complexes. Signals at δ 3.29 (s, 3H, –OCH_3), δ 3.67 (s, 3H, –OCH_3), and δ 3.91 (s, 3H, –OCH_3) are due to the protons of three –OCH_3 groups present on the phenyl ring. Thus, ^{1}H NMR spectra supplement the assigned geometry.

3.5. *Mass spectral studies*

The DART-MS of TMeOBFC shows a molecular ion peak at m/z 355, one mass unit more than the molecular weight of the Schiff base. The DART-MS of [Co(TMeOBFC)(opd)Cl_2] shows a molecular ion peak at m/z 592, which is same as that of the molecular weight of the complex. The DART-MS of [Ni(TMeOBFC)(2,6-dap)Cl_2] shows a molecular ion peak at m/z 593, the same as the molecular weight of the complex. Mass spectral studies support the proposed composition.

3.6. *ESR spectra of Cu(II) complexes*

The ESR spectra of copper complexes in a polycrystalline state have been recorded at room temperature. The $g_{||}$ and $g_{\perp}$ values are 2.358, 2.339 and 2.089, 2.074, respectively. The g_{av} was calculated to be 2.182 and 2.166. The spectra have asymmetric bands with $g_{||} > g_{\perp} > 2.00277$ (TCNE) observed, indicating that the unpaired electron lies predominantly in the $d_{x^2-y^2}$ orbital with possible mixing of d_{z^2} because of low symmetry [43]. The axial symmetry parameter "G" is determined as $G = (g_{||} - 2.00277)/(g_{\perp} - 2.00277) = 4.123, 4.709$, respectively, more than 4 [44].

Table 5. Antimicrobial activity of the ligands and complexes.

	Zone of inhibition (mm)			
	Bacteria		Fungi	
Schiff base/complexes	*E. coli*	*S. aureus*	*A. niger*	*A. flavus*
TMeOBFC (L)	11	12	14	16
opd	14	13	17	15
$[CuL(opd)Cl_2]$	16	15	18	18
$[NiL(opd)Cl_2]$	15	14	18	17
$[CoL(opd)Cl_2]$	17	16	19	19
$[ZnL(opd)Cl_2]$	18	20	21	20
$[CdL(opd)Cl_2]$	19	17	18	19
$[HgL(opd)Cl_2]$	17	19	19	18
2,6-dap	13	15	14	13
$[CuL(2,6\text{-dap})Cl_2]$	14	16	17	18
$[NiL(2,6\text{-dap})Cl_2]$	14	17	16	19
$[CoL(2,6\text{-dap})Cl_2]$	16	16	18	17
$[ZnL(2,6\text{-dap})Cl_2]$	19	21	20	21
$[CdL(2,6\text{-dap})Cl_2]$	17	18	18	19
$[HgL(2,6\text{-dap})Cl_2]$	18	20	19	20
Gentamycine	20	22	–	–
Fluconazole	–	–	22	23

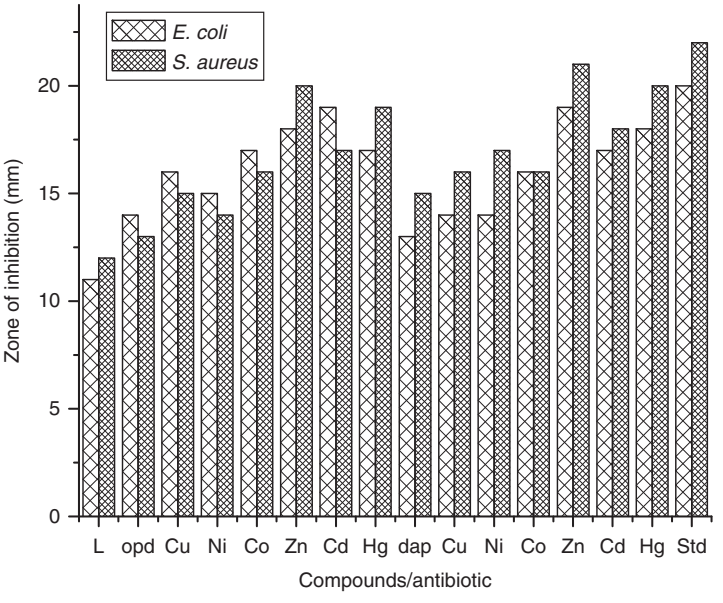


Figure 1. Antibacterial results of TMeOBFC, *ortho*-phenylenediamine (opd), 2,6-diaminopyridine (dap), metal(II) complexes, and Gentamycin (Std) at $1\,mg\,mL^{-1}$.

3.7. *Antibacterial and antifungal activities*

The Antibacterial and antifungal activities of the ligands and complexes have been tested by agar diffusion method for their activity against two bacteria *E. coli* and *S. aureus* and two fungi *A. niger* and *A. flavus*.

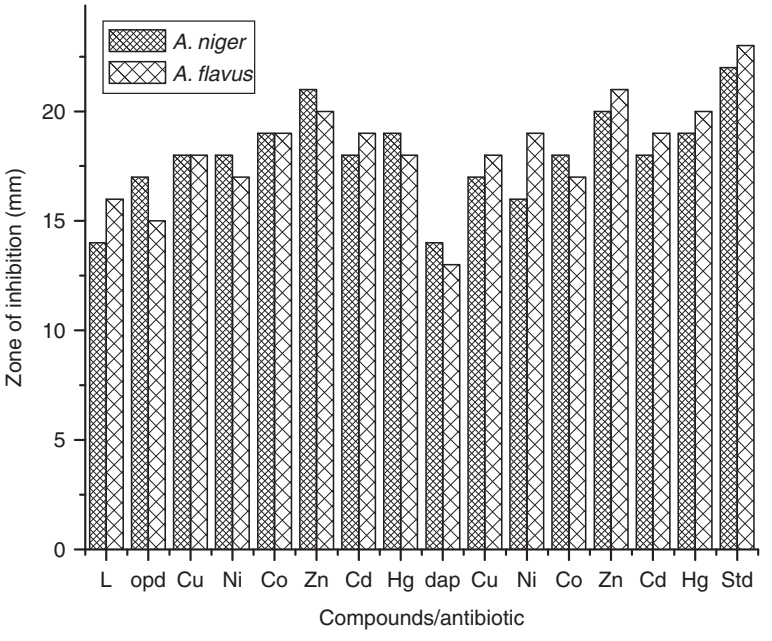


Figure 2. Antifungal results of TMeOBFC, opd, 2,6-dap, metal(II) complexes, and Fluconazole (Std) at 1 mg mL^{-1}.

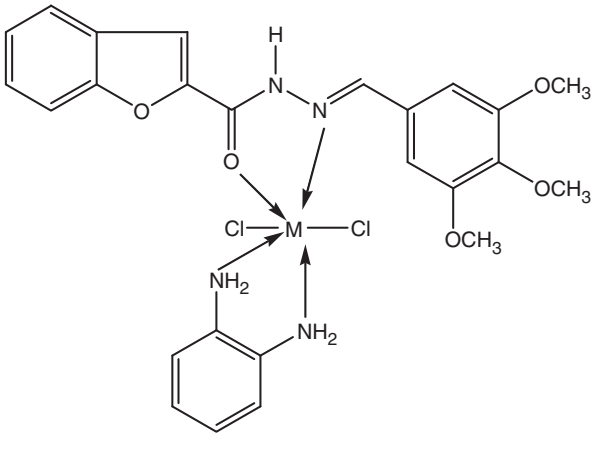


Figure 3. Suggested structures of Co(II), Ni(II), Cu(II), Zn(II), Cd(II) and Hg(II) complexes, TMeOBFC, and opd (M = Co(II), Ni(II), Cu(II), Zn(II), Cd(II), or Hg(II)).

The activities of TMeOBFC, opd, and 2,6-dap are shown in table 5 along with standards. From table 5 it is clear that Zn(II), Cd(II), and Hg(II) complexes show good activity against both bacteria and fungi, having higher activity than ligands against the same microorganisms under the same experimental conditions [45, 46] (figures 1 and 2).

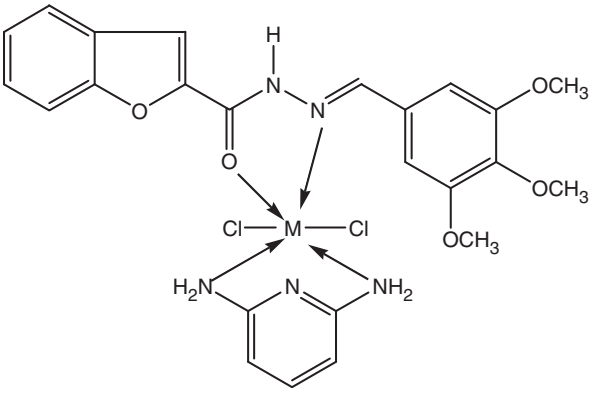


Figure 4. Suggested structures of metal complexes of TMeOBFC and 2,6-dap (M = Co(II), Ni(II), Cu(II), Zn(II), Cd(II), or Hg(II)).

4. Conclusion

We report new complexes of the type $MLL'Cl_2$, where M = Co(II), Ni(II), Cu(II), Zn(II), Cd(II), and Hg(II), L = TMeOBFC and L′ = opd or 2,6-dap. TMeOBFC is neutral bidentate, coordinating through the azomethine nitrogen and oxygen of –CONH–, whereas opd or 2,6-dap coordinate through amino groups. Based on analytical data and spectral studies, we propose octahedral structures to all the complexes (figures 3 and 4). Biological activity results show that metal complexes are more active compared to the parent ligands.

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