Original Article

8

Synthesis, Spectral and Biological Studies of Co(II), Fe(II), Ni(II), Cu(II), Pd(II), Mn(II), Hg(II), Cd(II), and Zn(II) Complexes Derived from Benzohydrazide Schiff Base

Debaraj B. Pallai^a | Raj R. Badekar^b | Kalimoddin I. Momin^c | Abhay S. Bondge^d | Ganpat R. Nagargoje^e | Prasad D. Kadam^f | Sharad P. Panchgalle^g | Vijaykumar S. More^{h,*}

^aDelicare Life Sciences Badlapur - 421503, Dist. Thane, (M.S.), India

^bRiva Industries, Karjat - 410201, Dist. Raigad, (M.S.), India

^cDepartment of Chemistry, Rajarshi Shahu Mahavidyalaya, Latur - 413512, Dist. Latur, (M.S.), India

^dDepartment of Chemistry, Shivaneri Mahavidyalaya Shirur Anantpal – 413544, Dist. Latur, (M.S.), India

^eDepartment of Chemistry, Shivaji Mahavidyalaya, Renapur - 413527, Dist. Latur, (M.S.), India

^fDepartment of Chemistry, Shri Kumarswami Mahavidyalaya, Ausa – 413520, Dist. Latur, (M.S.), India

^gDepartment of Chemistry, K. M. C. College, Khopoli -410203, Dist. Raigad, (M.S.), India

^hDepartment of Chemistry, Kai. Rasika Mahavidyalaya Deoni - 413519, Dist. Latur, Maharashtra, India



Citation D. Pallai, R. Badekar, K.I. Momin, A. Bondge, G.R. Nagargoje, P.D. Kadam, S.P. Panchgalle, V.S. More **Synthesis, Spectral and Biological Studies of Co(II), Fe(II), Ni(II), Cu(II), Pd(II), Mn(II), Hg(II), Cd(II), and Zn(II) Complexes Derived from Benzohydrazide Schiff Base.** *J. Appl. Organomet. Chem.*, **2024**, *4*(1), 76-87.

doihttps://doi.org/10.48309/JAOC.2024.434283.1156



Article info: Submitted: 6 January 2024 Revised: 5 February 2024 Accepted: 12 February 2024

ID: JAOC-2401-1156 Checked for Plagiarism: Yes Language Editor Checked: Yes

Key	w	or	ds:
-----	---	----	-----

Antimicrobial	activity,	4-
Chlorobenzohydrazide,	2,	6-
Dihydroxybenzaldehyd	e, Oct	ahedral
geometry		

<u>ABSTRACT</u>

Electronic absorption spectra, infrared spectroscopy, nuclear magnetic resonance, molar conductance, and elemental analysis are some of the physicochemical methods used to characterize the 4-chloro-N'-[(E)-(2,6dihydroxy phenyl) methylidene] benzohydrazide (HBHDH) molecule that has been synthesized. The results of the analysis show that the metal and ligand formed a 1:2 ratio. According to the spectra, the ligand HBHDH forms tridentate coordination spheres with metal ions using an ONO bond. Complexes with Fe(II), Co(II), Ni(II), Cu(II), and Mn(II) have an octahedral shape because of this. In contrast to the tetrahedral structure of the complexes created with Zn(II), Cd(II), and Hg(II), the Pd(II) complex takes on a square planar layout. According to the molar conductance values in DMF, the complexes do not undergo electrolysis. It has been determined that the metal complexes and Schiff's base HBHDH ligand have antibacterial action. The antibacterial efficiency of each combination and ligand against E. coli bacteria is higher than that of regular streptomycin.Candida albicans (MCC 1439) and Saccharomyces cerevisiae (MCC 1033) have significant antifungal effectiveness, with their growth being inhibited by over 92% in the presence of Mn(II) and Fe(II) complexes, respectively.

*Corresponding Authors: Vijaykumar S. More (vijaymore@gmail.com)

Introduction

n particular, hydrazones -NH-N=CRR' (where R and R' = H, alkyl, aryl) have attracted a lot of attention as a family of ligands due to their ability to create stable complexes with most metal ions through novel coordination mechanisms [1-4]. Catalytic processes and simulations of biological systems have made use of hydrazone Schiff base complexes. Schiff base metal complexes with nitrogen and other donors have recently been the subject of a great deal of chemical investigation. Their biological activity and stability, like their anti-convulsant and antitumor properties, mav be related to osteoporosis. Oxidation catalysis and electrochemistry are two of the many possible uses for hydrazones [5,6]. Hydrazones have multiple uses, including as growth regulators, nematocides, rodenticides, and herbicides [7]. According to literature [8-10], much of the research has focused on hydrazone complexes of transition metal ions.

Hydrazine Schiff base metal complexes prepared from 4-chlorobenzohydrazide and 2, 4-dihydroxybenzaldehyde have not yet been documented in any study. The objective of this study is to create and characterize complexes of 4-chloro-N'-[(E)-(2,6-

dihydroxyphenyl)methylidene]benzohydrazide (HBHDH) with several metals. Spectroscopic methods (electronic absorption, infrared, ¹H nuclear magnetic resonance) and magnetic moments are used to investigate the complexes' shape, bond types, and probable chelation mechanism. Antimicrobial and antifungal activity testing was performed on all of the synthesized compounds.

Experimental

The analytical grade (AR) chemicals and solvents used to synthesize the HBHDH ligand and its related metal complexes were supplied by S.D. Fine. To prepare metal complexes, certain metal salts are required, such as FeSO₄,

 $CoCl_2 \cdot 6H_2O$, $NiCl_2 \cdot 7H_2O$, $PdCl_2$, $CuCl_2 \cdot 5H_2O$, ZnCl₂·7H₂O, CdCl₂, HgCl₂, and MnCl₂·2H₂O. You can buy them and use them just the way they are. The production of 4-chlorobenzohydrazide [11] followed the steps outlined in the published literature. We found the amounts of metals and chlorides using Vogel's approach [12]. The ligand and its metal complexes were studied by measuring their infrared spectra with KBr pellets using a Brucker infrared spectrophotometer. А JASCO UV-Visible spectrophotometer (V-650) was used to detect the electronic absorption spectra of the metal complexes within the 1000 to 200 nm wavelength range. Researchers recorded the ¹H-NMR spectra of the ligand and its metal complexes in DMSO-d6 using a Bruker 400 MHz spectrophotometer, with TMS acting as an internal reference. The method developed by Rast was used to determine molecular weights [13-17].

At room temperature, the metal complexes' magnetic susceptibilities were measured using Gouy's method, calibrated with Hg [Co(SCN)₄], which stands for mercury (II) tetra thiocyanate cobalt (II). A solution of nitrobenzene (10⁻³ M) was used to determine the molar conductivity at room temperature.

Preparation of HBHDH ligand

In **Scheme 1**, a hot 4-chlorobenzohydrazide (**1**) was mixed with a hot ethanolic solution of 2, 6dihydroxybenzaldehyde (**2**) dropwise while being constantly agitated. An oil bath could heat the reaction mixture for three to four hours. We filtered the yellow PPT, washed it with ethanol, and dried it under vacuum after it cooled to room temperature. Colour; yellow. M.W.; 290.70 g/mol. % yield; 69.07. M.P.; 183. % composition: C, 57.84; H, 3.81; N, 9.64; O, 16.51; S, 12.20. UVvis (λ nm); 335, 295, and 335. FT(IR) (cm⁻¹); 3257, 2926, 3190, 2937, 1629, 1605, and 1295. ¹H-NMR (δ); 12.01 (-OH, C2), 11.46 (-OH, C6), 10.02 (-NH), 8.55 (-CH=), and 6.36-7.99 (Ar. protons).



Scheme 1. Preparation of HBHDH ligand

Preparation of metal complexes

The appropriate metal salt (0.05 mol) was slowly added to a 15 mL container of hot ethanolic HBHDH ligand (0.1 mol) solution while stirring continuously. Hot ethanol was used to carry out the reaction. The inclusion of 0.1 N NaOH brought the reaction mixture's pH up to 7. The mixture was heated in an oil bath and refluxed for five to eight hours. Cooling the solution after digestion reduced its volume by half and allowed it to solidify. Following a period of drying at room temperature, the product was rinsed with ethanol. Results should range from 69 to 73%.

$[Fe(BHDH)_2]$

Colour: Blue. M.W.: 635.24 g/mol. % yield: 73.46. M.P.: 200. % composition: C, 52.89; H, 3.75; N, 8.82; O, 15.11; S, 11.20; Fe, 8.79. UV-Vis (λ_{nm}): 613 and 503. FT(IR) (cm⁻¹): 2926, 3152, 2840, 1584, 1525, 1233, 631, 550, and 520.

$[Co(BHDH)_2]$

Color: brown. M.W: 640.39 g/mol. % yield: 76.44. M.P.: 203. % composition: C, 52.47; H, 3.12; N, 8.75; O, 15.00; S, 11.10; Co, 9.12. UV-Vis (λ_{nm}): 900 and 675. FT(IR) (cm⁻¹): 2919, 3448, 2849, 1596, 1542, 1217, 653, 590, and 511.

[Ni(BHDH)₂]

Colour: light green. M.W.: 640.09 g/mol. % yield: 79.13. M.P.: 204. % composition: C, 52.49; H, 3.12; N, 8.75; O, 15.00; S, 11.10; Ni, 9.17. UV-Vis (λ_{nm}): 995 and 667. FT(IR) (cm⁻¹): 2924, 3408, 2848, 1593, 1549, 1217, 639, 618, and 532.

[Pd(BHDH)₂]

Colour: Green. M.W.: 687.40 g/mol. % yield: 83.87. M.P.: 209. % composition: C, 48.48; H, 2.91; N, 8.15; O, 14.00; S, 10.33; Pd, 15.42. UV-Vis (λ_{nm}): 396, 331, and 285. FT(IR) (cm⁻¹): 2923, 3288, 2851, 1552, 1518, 1218, 535, and 525. ¹H-NMR (δ); 11.51 (-OH, C6), 9.74 (-NH), 8.65 (-CH=), 6.41-8.04 (Ar. protons).

$[Cu(BHDH)_2]$

Colour: Green. M.W.: 644.95 g/mol. % yield: 81.61. M.P.: 206. % composition: C, 52.10; H, 3.10; N, 8.68; O, 14.90; S, 11.00; Cu, 9.85. UV-Vis (λ_{nm}): 638. FT(IR) (cm⁻¹): 2923, 3152, 2852, 1581, 1513, 1219, 595, 521, and 515.

$[Zn(BHDH)_2]$

Colour: Yellow. M.W.: 646.76 g/mol. % yield: 77.40. M.P.: 206. % composition: C, 52.10; H, 3.10; N, 8.68; O, 14.90; S, 11.00; Zn, 9.85. UV-Vis (λ_{nm}): 332, 310, and 230. FT(IR) (cm⁻¹): 2922, 3341, 2851, 1592, 1547, 1215, 595, and 528. ¹H-NMR (δ); 11.28 (-OH, C6), 9.61 (-NH), 8.48 (-CH=), 7.11-7.55 (Ar. protons).

[Cd(BHDH)₂]

Colour: Yellow. M.W.: 693.81 g/mol. % yield: 81.55. M.P.: 210. % composition: C, 48.43; H, 2.88; N, 8.07; O, 13.80; S, 10.20; Cd, 16.20. UV-Vis (λ_{nm}); 340, 253. FT(IR) (cm⁻¹): 2979, 3197, 2835, 1579, 1548, 1226, 602, and 552. ¹H-NMR (δ); 11.27 (-OH, C6), 10.04 (-NH), 8.65 (-CH=), 6.41-8.67 (Ar. protons).

Journal of Applied Organometallic Chemistry

$[Hg(BHDH)_2]$

Colour: Yellow. M.W.: 781.40 g/mol. % yield; 69.77. M.P.: 206. % composition: C, 43.00; H, 2.56; N, 7.17; O, 12.29; S, 9.09; Hg, 25.67. UV-Vis (λ_{nm}); 340, 310, 230. FT(IR) (cm⁻¹); 2922, 3321, 2852, 1604, 1540, 1241, 529, and 510. ¹H-NMR (δ); 11.56 (-OH, C6), 9.89 (-NH), 8.55 (-CH=), 6.36-7.99 (Ar. protons).

$[Mn(BHDH)_2]$

Colour: Brown. M.W.: 634.34 g/mol. % yield: 69.88. M.P.: 202. % composition: C, 52.97; H, 3.15; N, 8.83; O, 15.10; S, 11.20; mn, 8.66. UV-Vis (λ_{nm}); 561 and 398. FT(IR) (cm⁻¹): 2925, 3257, 2836, 1552, 1515, 1218, 618, 571, and 534.

Biological evaluation

To investigate the chemicals' efficacy against various microorganisms, including *Staphylococcus aureus* (MCC 2408), *B. subtilis* (MCC 2010), *Escherichia coli* (MCC 2412), *Pseudomonas aeruginosa* (MCC 2080), *fungi* (*C. albicans* (MCC 1439), and *S. cerevisiae* (MCC 1033) [16], the conventional disc-agar diffusion technique was employed. *Fluconazole* was considered the best treatment for Gramnegative bacteria.

This is in contrast to the usage of cycloheximide against fungi and cephalothin against Grampositive bacteria. A 2 and 1 mg/mL solution of the compounds was prepared before testing using DMF, which did not have any inhibitory effect. The experimental medium used was PDA, which stands for potato dextrose agar and is composed of 200 g of potato infusion, 6 g of dextrose, and 15 g to measure.

Before being delicately put on incubated agar surfaces, discs of filter paper were soaked with 10 mL of a solution containing a controllable concentration of test compounds. Following incubation for 36 hours at 27 °C for bacteria and 48 hours at 24 °C for fungus, the distance between each disc and the surrounding clear zone was measured. The mean inhibition zones were then calculated.

Results and discussion

The HBHDH ligand and its complexes exhibit resistance to the negative impacts of oxygen and moisture when stored at room temperature. The complexes exhibit limited solubility in water, although they are partially soluble in ethanol and methanol and completely soluble in DMF, nitrobenzene, and DMSO solvents. Table 1 presents the analytical data for both the ligand and the complexes. The results corroborate the chemical formula of the ligand, $C_{14}H_{11}ClN_2O_3$, and the 1:2 metal-to-ligand stoichiometry in the complexes.

Compound	Colour	MW	% Yield	MP/DP	DP Element Content					Cond	MM	
					М	С	Н	Ν	0	S		
HBHDH	Yellow	290.70	69.07	183	-	57.84	3.81	9.64	16.51	12.20	-	-
Fe(BHDH) ₂	Blue	635.24	73.46	200	8.79	52.89	3.15	8.82	15.11	11.20	1.49	5.12
Co(BHDH) ₂	Brown	640.39	76.44	203	9.21	52.47	3.12	8.75	15.00	11.10	1.56	4.45
Ni(BHDH) ₂	L. Green	640.09	79.13	204	9.17	52.49	3.12	8.75	15.00	11.10	0.76	3.14
Pd(BHDH) ₂	Green	687.40	83.78	209	15.42	48.88	2.91	8.15	14.00	10.33	2.25	-
Cu(BHDH) ₂	Green	644.95	81.61	206	9.85	52.10	3.10	8.68	14.90	11.00	2.10	2.10
Zn(BHDH) ₂	Yellow	646.76	77.40	203	10.11	51.94	3.09	8.66	14.80	10.97	2.40	-
Cd(BHDH) ₂	Yellow	693.81	81.55	210	16.20	48.43	2.88	8.07	13.80	10.20	3.02	-
Hg(BHDH) ₂	Yellow	781.40	69.77	206	25.67	43.00	2.56	7.17	12.29	9.09	0.73	-
Mn(BHDH) ₂	Brown	634.34	69.88	202	8.66	52.97	3.15	8.83	15.10	11.20	2.25	5.27

Table 1. Analytical and physicochemical data of metal complexes with the HBHDH ligand

The molar conductance values of the complexes in DMF at a concentration of 10⁻³ M, ranging from 9.8 to 20 ohm⁻¹cm²mol⁻¹, suggest that the complexes lack electrolytic properties [18].

¹*H*-*NMR study of HBHDH ligand*

Table 2 lists the pertinent data for determining the structure of the HBHDH ligand from the ¹H NMR spectra that were acquired in CDCl₃ (suplementary file). At 12.02 ppm, the HBHDH ligand's ¹H-NMR spectra revealed a singlet (S, 1H. -OH C2). The C2 position's phenolic -OH group was designated as the site of this singlet. This peak is absent in all prepared metal complexes, indicating the deprotonation of the hydroxyl group during complexation. At 11.46 ppm, there was another singlet that was identified as the proton of the C6 position's phenolic -OH group (S, 1H, -OH C6) [19-20]. The singlet was observed at 10.02 ppm, assigned as the proton of the imino-NH group. The -CH= group of the prepared ligand and its metal complexes was observed at 8.55 ppm. The multiplets seen in the range from 6.38 to 7.99 ppm are assigned by aromatic protons.

Absorption and magnetic moment spectra

The HBHDH ligand and its complexes were subjected to electronic spectra measurements in DMF (suplementary file). The projected structure of synthesized transition complexes and data on magnetic moments are displayed in Table 3, which is based on recorded electronic absorption spectra. The aromatic ring $\pi \rightarrow \pi^*$ transition, the >C=O transition, and the $n \rightarrow \pi^*$ transitions of the C=N groups were identified in

the ligand's electronic absorption spectra, which showed high-energy bands at 235, 295, and 335 nm, respectively. It was considered that intra-ligand and LMCT transitions of coordinated ligands caused these bands to be moved to 230-340, 253-378, and 331-480 nm in complexes. The ${}^{5}T_{2g} \rightarrow {}^{5}E_{g}$ transition, identified as occurring at 613 nm and assigned to one atom, validated the complex's octahedral geometry, and a magnetic moment value of 5.12 B.M. was determined for the Fe (II) complex. The magnetic moment values of Fe(II) complexes were 3.14 B.M. and 4.45 B.M., respectively, in the literature [21,22]. The Co(II) complex's electronic spectra display three transitions at 900 and 621 nm, which can be attributed to ${}^{4}T_{1g}$ (F) \rightarrow ${}^{4}T_{1g}$ (F) and ${}^{4}T_{2g}$ (F) \rightarrow ${}^{4}T_{1g}$ (P) transitions, respectively.

In contrast, the Ni(II) complex exhibits three transitions at 995, 667, and 545 nm, which can be attributed to ${}^{3}T_{2g} \rightarrow {}^{3}T_{2g}$, ${}^{3}T_{2g} \rightarrow {}^{3}T_{1g}(F)$, and ${}^{3}T_{2g} \rightarrow {}^{3}T_{1g}(P)$ transitions, each of which suggests an octahedral geometry. The electronic spectra of the Cu(II) compound showed a wideband with low intensity at around 638 nm in the visible region [23], which is caused by the *d*-*d* (${}^{2}B_{1g} \rightarrow {}^{2}E_{g}$) transition. All signs led to a distorted octahedral arrangement.

In the Mn(II) complex, the octahedral structure was brought about via the d-d (${}^{6}A_{1g} \rightarrow {}^{5}E_{g}$, ${}^{4}D$, and ${}^{6}A_{1g} \rightarrow {}^{5}T_{1g}$) transition, which was seen as the lowest-energy band at 561 and 398 nm, respectively. The electronic spectra of the Pd(II) complex showed transitions at 396, 331, and 285 nm [25]. These alterations were given a square, flat form. Because their d¹⁰ transition is complete, complexes containing Zn(II), Cd(II), and Hg(II) do not display a d-d band.

Compound	-OH (C2)	-OH (C6)	-NH-	-CH=	Aromatic Protons
HBHDH	12.02	11.46	10.02	8.55	6.38-7.99
Pd(BHDH) ₂	-	11.51	9.74	8.65	6.41-8.04
Zn(BHDH) ₂	-	11.28	9.61	8.48	7.11-7.55
Cd(BHDH) ₂	-	11.27	10.04	8.65	6.41-8.67
Hg(BHDH) ₂	-	11.56	9.89	8.55	6.36-7.99

Table 2.¹H-NMR spectroscopic data of the HBHDH ligand and its metal complexes

Table 3. HBHDH ligand electronic spectral data andmetal complexes

Compound	λnm	Transition
	335	$n \rightarrow \pi^*$
HBHDH	295	$n \rightarrow \pi *$
	235	$n \rightarrow \pi^*$
Mn(BHDH)2	561	${}^{6}A_{1g} \rightarrow {}^{4}T_{1g}({}^{4}P)$
	398	${}^{6}\text{A1g} \rightarrow {}^{4}\text{E}_{g}(4\text{D})$
Fe(BHDH) ₂	613	${}^{5}T_{2g} \rightarrow {}^{5}E_{g}$
	503	$L \rightarrow M$ charge transfer
Co(BHDH)2	900	${}^{4}T_{1g(F)} \rightarrow {}^{4}T_{2g(F)}(v_{1})$
	675	${}^{4}T_{1g(F)} \rightarrow {}^{4}T_{2g(P)}(\nu_{2})$
Ni(BHDH)2	995	$^{3}A_{2g(F)} \rightarrow ^{3}T_{2g(F)} (\nu_{1})$
	667	$^{3}A_{2g(F)} \rightarrow ^{3}T_{1g(F)} (\nu_{2})$
Pd(BHDH)2	396, 331, 285	$L \rightarrow M$ charge transfer
Cu(BHDH)2	638	$^{2}B_{1g} \rightarrow ^{2}A_{1g}(\nu_{1})$
Zn(BHDH)2	332, 257, 288	$L \rightarrow M$ charge transfer
Cd(BHDH) ₂	340, 253	$L \rightarrow M$ charge transfer
Hg(BHDH) ₂	340, 310, 230	$L \rightarrow M$ charge transfer

FT(IR) spectral studies

As a result of comparing the IR spectra indicated in Table 4 with those of free HBHDH ligands, we can see the different kinds of bonds that form in complexes (suplementary file). At 3190, 1629, 1605, and 1295 cm⁻¹, respectively, the infrared bands for v(N-H), v(C=0), v(C=N), and phenolic (C-O) are visible in the free HBHDH ligand. The wavelengths that were measured are these. At 3295 cm⁻¹, however, a medium broad band was observed due to the ohydroxy group's intramolecular solid hydrogen bonding. The breadth of this band was approximately 3295 cm⁻¹. When this band vanished in complexes, it meant that phenolic oxygen had deprotonated and was coordinating with metal ions. The phenolic (C-O) frequency decreased from 1295 cm⁻¹ to 1215-1233 cm⁻¹, in contrast to the ligand's frequency of 1350 cm⁻¹, lending credence to this theory. It is worth noting that this band vanished in complexes, suggesting that metal ions coordinate with phenolic oxygen by deprotonation. The stretching frequency of (C=N), which is present in free ligands, changes to a value of 63-90 cm⁻¹ lower [24] when complexes are present. In this case, the coordination of the metal ion with the

nitrogen of the azomethine molecule is seen. The (C=O) vibrations moved to the lower side of the spectrum in all transition metal complexes, indicating that this group was involved in the complexation process [26-27].

That HBHDH ligands in metal complexes are monobasic tridentate was thus proven by this finding. A band in the vicinity of 3152-3448 cm⁻¹ is allocated to the 6-position hydroxy group of the aromatic ring by the HBHDH ligand and its complexes. It appears from this band that the complexation mechanism does not involve the 6-position hydroxyl group [28-30]. The bands observed in the 535-653, 521-618, and 510-534 cm⁻¹ far-infrared regions can be ascribed to the $\nu(M\leftarrow O)$, $\nu(M)$, and $\nu(M\leftarrow N)$ vibrations, respectively. The structures of complexes are allocated based on spectral investigations.

Antimicrobial study

Testing the HBHDH ligand and its compounds for antibacterial properties is shown in Table 5. The standard and its metal complexes had higher activity against all microorganisms, but the ligand HBHDH had lesser activity. The chelation that took place is the reason behind this, according to Tweedy's hypothesis of chelation.

Antibacterial activity

The antibacterial efficacy associated with each metal complex is nearly uniform against all bacterial strains.

The average *B. subtilis* has an inhibition zone of 13mm, but Cu(II), Zn(II), and Co(II) all have far larger inhibition zones of 14 mm, 13 mm, and 13 mm, respectively, against *E. coli*. The Hg(II) molecule had no impact on *S. aureus* (Figure 1).

Antifungal activity

The Ni(II) complex exhibits potent antifungal activity, as demonstrated by its efficacy against *C. albicans. S. cerevisiae* demonstrates a growth rate reduction of over 90% in the presence of the Mn(II) complex (Table 6). The Co(II) complex inhibits the development of all fungal strains (Figure 2).



IIDIIDII	3257	2926	3190	2937	1629	1605	1295	-	-	-
Fe(BHDH) ₂	-	2926	3152	2840	1584	1525	1233	631	550	520
Co(BHDH) ₂	-	2919	3448	2849	1596	1542	1217	653	590	511
Ni(BHDH) ₂	-	2924	3408	2848	1593	1549	1217	639	618	532
Pd(BHDH) ₂	-	2923	3258	2851	1552	1518	1218	535	-	535
Cu(BHDH) ₂	-	2923	3152	2852	1581	1513	1219	595	521	515
Zn(BHDH) ₂	-	2922	3341	2851	1592	1547	1215	595	-	528
Cd(BHDH) ₂	-	2979	3197	2835	1579	1548	1226	602	-	552
Hg(BHDH) ₂	-	2922	3321	2851	1604	1540	1241	529	-	510
Mn(BHDH) ₂	-	2925	3257	2836	1552	1515	1218	618	571	534

Compound	Antibacterial Activity (zone of inhibition)					
	S. aureus	B. subtilis	E. coli	Р.		
				aeruginosa		
HBHDH	0	0	10	0		
Fe(BHDH)2	11	7	12	9		
Co(BHDH) ₂	13	21	13	18		
Ni(BHDH) ₂	9	9	8	9		
Pd(BHDH) ₂	14	13	10	0		
Cu(BHDH) ₂	13	11	14	10		
Zn(BHDH)2	16	7	13	12		
Cd(BHDH) ₂	9	0	0	0		
Hg(BHDH) ₂	21	8	9	13		
Mn(BHDH)2	0	0	7	0		
Streptomycin	12	11	8	9		

Table 5. HBHDH ligand and its metal complexes have been studied for their antibacterial properties



Figure 1. Antibacterial activity of HBHDH ligand and its metal complexes

Table 6. Antifungal investigation on the HBHDH ligand and its metal complexes

Compound	Antibacterial Activity (zone of inhibition				
	C albican	<i>S. C</i> .			
HBHDH	8	8			
Fe(BHDH) ₂	14	13			
Co(BHDH) ₂	15	9			
Ni(BHDH) ₂	25	21			
Pd(BHDH) ₂	8	14			
Cu(BHDH) ₂	9	8			
Zn(BHDH) ₂	14	8			
Cd(BHDH) ₂	8	12			
Hg(BHDH) ₂	14	8			
Mn(BHDH)2	12	19			
Fluconazole	10	7			



Figure 2. HBHDH ligand and its metal complexes have antifungal behavior

Conclusion

Various spectroscopy techniques, including analytical, infrared (IR), electrical, magnetic, and electronic absorption spectra, were used to validate the binding of ligands to metal. The research revealed that the HBHDH ligand exhibited a monobasic tridentate structure with transition metal ions. The Mn(II), Fe(II), Ni(II), Cu(II), and Co(II) complexes were determined to have octahedral geometry, whereas the Zn(II), Mn(II), Hg(II), and Cd(II) complexes were found to have square planar and tetrahedral geometry for Pd(II) complex. These conclusions were drawn from the findings of analytical and physicochemical examinations. The metal complexes exhibit a considerably greater antibacterial activity compared to the ligand. Typically, the ligand and its complexes have significantly greater efficacy against E. coli bacteria compared to normal Penicillin.

Conflict of interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

Acknowledgements

Authors thank Dr. Chandrakant Jawale (Principal, Kai. Rasika Mahavidyalaya Deoni, Dist. Latur) for support, suggestions and encouragement during the execution of this project.

Orcid

Debraj Pallai https://orcid.org/0009-0002-4879-9416

Raj Badekar https://orcid.org/0000-0002-9425-4743

Ganpat R. Nagargoje https://orcid.org/0009-0003-3796-8496

Abhay S. Bondge https://orcid.org/0000-0002-6249-7185

Prasad D. Kadam https://orcid.org/0009-0004-1480-2271

Kalimoddin I. Momin https://orcid.org/0000-0001-6297-6509

Sharad P. Panchgalle https://orcid.org/0000-0001-9706-7567

Vijaykumar S. More *https://orcid.org/0000-0002-6110-8774*

References

[1]. S. Kelode. P. Mandlik. Synthesis. characterization, thermal and antibacterial studies of cobalt (II), nickel (II), copper (II) and zinc (II) complexes of hydrazone Schiff base, International Journal of Chemical and Pharmaceutical Sciences, 2012, 3, 8-13. [Crossref], [Google Scholar]

[2]. S. Gayathri, P. Viswanathamurthi, V. Thuslim, M. Sathya, M. Ranjani, R. Prabhakaran, J. Haribabu, C. Echeverria, Synthesis, structural, DNA/protein binding and cytotoxic studies of copper (I)∝-diimine hydrazone complexes, *Inorganica Chimica Acta*, **2022**, *533*, 120780. [Crossref], [Google Scholar], [Publisher]

[3]. N. Raghav, M. Singh, Biological activities of hydrazones, *International Journal of Pharmacy and Pharmaceutical Sciences*, **2011**, *3*, 26-32. [Google Scholar]

[4]. W.A. Wani, E. Jameel, U. Baig, S. Mumtazuddin, L.T. Hun, Ferroquine and its derivatives: new generation of antimalarial agents, *European journal of medicinal chemistry*, **2015**, *101*, 534-551. [Crossref], [Google Scholar], [Publisher]

[5]. F.K. Camellia, M. Ashrafuzzaman, M.N. Islam, L.A. Banu, Synthesis, characterization and thermal study of some transition metal complexes of *N*-(4-hydroxybenzylidene) isonicotinohydrazone and investigation of their antibacterial and antioxidant properties, *Asian Journal of Chemical Sciences*, **2022**, *11*, 8-22. [Crossref], [Google Scholar], [Publisher]

[6]. J. Devi, S. Kumar, D. Kumar, D.K. Jindal, Y. Poornachandra, Synthesis, characterization, in vitro antimicrobial and cytotoxic evaluation of Co (II), Ni (II), Cu (II) and Zn (II) complexes derived from bidentate hydrazones, *Research on Chemical Intermediates*, **2022**, 1-33. [Crossref], [Google Scholar], [Publisher]

Journal of Applied Organometallic Chemistry

[7]. A.N. Alhakimi, Synthesis, characterization and microbicides activities of *N*-(hydroxy-4-((4nitrophenyl) diazenyl) benzylidene)-2-(phenylamino) acetohydrazide metal complexes, *Egyptian Journal of Chemistry*, **2020**, *63*, 1509-1525. [Crossref], [Google Scholar], [Publisher]

[8]. R. Bhaskar, C. Ladole, N. Salunkhe, A. Aswar, Synthesis, Characterization and biological studies of some transition metal complexes with pyrazine Schiff base hydra zone ligand, *Jordan Journal of Chemistry (JJC)*, **2020**, *15*, 61-72. [Crossref], [Google Scholar], [Publisher]

[9]. R. Bhaskar, N. Salunkhe, A. Yaul, A. Aswar, Bivalent transition metal complexes of ONO donor hydrazone ligand: Synthesis, structural characterization and antimicrobial activity, *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, **2015**, *151*, 621-627. [Crossref], [Google Scholar], [Publisher]

[10]. S. Aly, S.K. Fathalla, Preparation, characterization of some transition metal complexes of hydrazone derivatives and their antibacterial and antioxidant activities, *Arabian Journal of Chemistry*, **2020**, *13*, 3735-3750. [Crossref], [Google Scholar], [Publisher]

[11]. R.B. Singh, N. Das, G.K. Singh, S.K. Singh, K. Zaman, Synthesis and pharmacological evaluation of 3-[5-(aryl-[1, 3, 4] oxadiazole-2-yl]-piperidine derivatives as anticonvulsant and antidepressant agents, *Arabian Journal of Chemistry*, **2020**, *13*, 5299-5311. [Crossref], [Google Scholar], [Publisher]

[12]. A. Vogel, A Text Book of Quantitative Inorganic Analysis, 3rdEdition, London: Longman ELBS; **1968**. [Crossref], [Google Scholar], [Publisher]

[13]. A.M. Hassan, A. Osman said, B.H. Heakal, A. Younis, M.A. Abdelmoaz, M.M. Abdrabou, Conventional and microwave-assisted synthesis, antimicrobial and antitumor studies of tridentate schiff base derived from *O*-vanillin and phenyl urea and its complexes, *Advanced Journal of Chemistry, Section A*, **2020**, *3*, 621-638. [Crossref], [Google Scholar], [Publisher]

[14]. M. Nencki, N. Sieber, Ueber die Verbindungen der ein-und zweibasischen Fettsäuren mit Phenolen, *Journal fuer praktische Chemie*, **1881**, *23*, 147-156. [Google Scholar], [PDF]

[15]. A. Reiss, M.C. Chifiriuc, E. Amzoiu, N. Cioateră, I. Dăbuleanu, P. Rotaru, New metal (II) complexes with ceftazidime Schiff base, *Journal of Thermal Analysis and Calorimetry*, **2018**, *131*, 2073-2085. [Crossref], [Google Scholar], [Publisher]

[16]. M. Vijayalakshmi, Synthesis and characterization of some transition metal complexes of Schiff base derived from 2, 4–dihydroxybenzaldehyde: Synthesis and characterization of some transition, *Iranian Journal of Pharmaceutical Sciences*, **2019**, *15*, 29-40. [Crossref], [Google Scholar], [Publisher]

[17]. P.G. Jamkhande, A.S. Wattamwar, S.S. Pekamwar, P.G. Chandak, Antioxidant, antimicrobial activity and in silico PASS prediction of Annona reticulata Linn. root extract, *Beni-Suef University Journal of Basic and Applied Sciences*, **2014**, *3*, 140-148. [Crossref], [Google Scholar], [Publisher]

[18]. W. Cao, Y. Liu, T. Zhang, J. Jia, Synthesis, characterization, theoretical and antimicrobial studies of tridentate hydrazone metal complexes of Zn (II), Cd (II), Cu (II) and Co (III), *Polyhedron*, **2018**, *147*, 62-68. [Crossref], [Google Scholar], [Publisher]

[19]. M.S. Hossain, F.K. Camellia, N. Uddin, L.A. Banu, M.M. Haque, Synthesis, characterization and antimicrobial activity of metal complexes of *N*-(4-methoxybenzylidene)

isonicotinohydrazone schiff base, *Asian Journal of Chemical Sciences*, **2019**, *6*, 1-8. [Google Scholar]

[20]. L.W. Xue, H.J. Zhang, P.P. Wang, Synthesis, crystal structures, and antimicrobial activity of copper (II) complexes derived from *N'*-(1-(pyridin-2-yl) ethylidene) isonicotinohydrazide, *Inorganic and Nano-Metal Chemistry*, **2020**, *50*, 637-643. [Crossref], [Google Scholar], [Publisher]

[21]. A. Mosa, A.A. Emara, J.M. Yousef, A. Saddiq, Novel transition metal complexes of 4-hydroxycoumarin-3-thiocarbohydrazone:

Pharmacodynamic of Co (III) on rats and antimicrobial activity, *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, **2011**, *81*, 35-43. [Crossref], [Google Scholar], [Publisher]

[22]. J. Singh, P. Singh, Synthesis, spectroscopic characterization, and in vitro antimicrobial studies of pyridine-2-carboxylic acid *N'*-(4-chloro-benzoyl)-hydrazide and its Co (II), Ni (II), and Cu (II) complexes, *Bioinorganic Chemistry and Applications*, **2012**, *2012*. [Crossref], [Google Scholar], [Publisher]

[23]. R. Kannappan, S. Tanase, I. Mutikainen, U. Turpeinen, J. Reedijk, Square-planar copper (II) halide complexes of tridentate ligands with π - π stacking interactions and alternating short and long Cu \cdots Cu distances, *Inorganica Chimica Acta*, **2005**, *358*, 383-388. [Crossref], [Google Scholar], [Publisher]

[24]. S. Chandra, L.K. Gupta, EPR, IR and electronic spectral studies on Mn (II), Co (II), Ni (II) and Cu (II) complexes with a new 22-membered azamacrocyclic [N4] ligand, *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, **2004**, *60*, 1751-1761. [Crossref], [Google Scholar], [Publisher]

[25]. M.A. Ali, A.H. Mirza, R.J. Butcher, M. Tarafder, T.B. Keat, A.M. Ali, Biological activity of palladium (II) and platinum (II) complexes of the acetone Schiff bases of *S*-methyl-and *S*-benzyldithiocarbazate and the X-ray crystal structure of the [Pd (asme)₂](asme= anionic form of the acetone Schiff base of *S*-methyldithiocarbazate) complex, *Journal of Inorganic Biochemistry*, **2002**, *92*, 141-148. [Crossref], [Google Scholar], [Publisher]

[26]. F. Rahaman, B. Mruthyunjayaswamy, Synthesis, spectral characterization and biological activity studies of transition metal complexes of Schiff base ligand containing indole moiety, *Complex Metals*, **2014**, *1*, 88-95. [Crossref], [Google Scholar], [Publisher]

Journal of Applied Organometallic Chemistry

[27]. F.Y. Chen, S.Y. He, Studies on Lanthanide (III) ternary mixed-ligand complexes with *N*-(2propionic acid)-salicyloyl hydrazone and isonicotinic acid: Synthesis, characterization, and antibacterial activities, *Synthesis and Reactivity in Inorganic, Metal-Organic, and Nano-Metal Chemistry*, **2008**, *38*, 642-646. [Crossref], [Google Scholar], [Publisher]

[28]. A.O. Olamide, K.A. Bello, A.A. Kogo, Dyes based on substituted pyridone using metal complexes (1: 2) and study of their application on nylon fabrics (6.6), *Journal of Apllied* *Organometallic Chemistry*, **2023**, *3*, 61-72, [Crossref], [Google Scholar], [Publisher]

[29]. C. Duru, I. Duru, Computational investigation of infrared vibrational frequency Shift modes in Schiff base-transition metal complexes, *Journal of Applied Organometallic Chemistry*,**2022**, *2*, 54-65. [Crossref], [Google Scholar], [Publisher]

[30]. D. Palke, Synthesis, Physicochemical and Biological studies of transition metal complexes of DHA Schiff bases of aromatic amine, *Journal of Applied Organometallic Chemistry*, **2022**, *2*, 81-88. [Crossref], [Google Scholar], [Publisher]

Copyright © 2024 by SPC (Sami Publishing Company) + is an open access article distributed under the Creative Commons Attribution License (CC BY) license (https://creativecommons.org/licenses/by/4.0/), which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.