
Pb(II) complexes of Schiff bases derived from benzoylhydrazine as the antibacterial agents

Mohammad Nasir Uddin, Md. Abdus Salam, Jannat Sultana

Department of Chemistry, University of Chittagong, Chittagong-4331, Bangladesh

Email address:

nasircu72@gmail.com (M. N. Uddin)

To cite this article:

Mohammad Nasir Uddin, Md. Abdus Salam, Jannat Sultana. Pb(II) Complexes of Schiff Bases Derived from Benzoylhydrazine as the Antibacterial Agents. *Modern Chemistry*. Special Issue: Synthesis and Microbial Screening of Coordination and Organic Compounds. Vol. 3, No. 1-1, 2015, pp. 7-14. doi: 10.11648/j.mc.s.2015030101.12

Abstract: Some Pb(II) complexes of dibasic tridentate Schiff base ligands derived from aldehydes/ketones and benzoylhydrazine have been synthesized and characterized by some physico-chemical studies; elemental, spectral (IR, UV, NMR, Mass), magnetic, and conductance analyses. The CHN elemental analysis data showed the formation of 1:1 [M:L] complexes. The obtained molar conductance values revealed non-electrolytic nature. The results of magnetic moment measurements supported diamagnetic phenomenon for the complexes. The infrared spectral data displayed the main coordination sites of ligands towards Pb^{2+} ions. The electronic spectral results of the complexes showed $\pi \rightarrow \pi^*$ (phenyl ring), $n \rightarrow \pi^*$ (HC=N), CT transition. Based on analytical data trigonal pyramidal geometry for the prepared complexes has been proposed. The antibacterial activity of the prepared complexes was also studied against some gram positive and gram negative bacteria. It is found that some of the complexes are quite effective against tested bacteria.

Keywords: Tridentate Dibasic Schiff Base, Benzoylhydrazine, Pb(II) Complexes, Antibacterial Activity, Trigonal Pyramidal Geometry

1. Introduction

Schiff base is a compound that contains azomethine group ($>C=N-$) connected to an aryl or alkyl group but not hydrogen. Schiff bases can be synthesized from an amine and a carbonyl compound by nucleophilic addition forming a hemi-aminal group followed by dehydration to generate an imine compound. A number of metal complexes with dibasic tridentate Schiff bases derived from various thiosemicarbazide hydrazides, *o*-hydroxybenzylamine or ethanolamine and *o*-hydroxy-aromatic aldehydes/ketones have been synthesized and characterized by Syamal *et al.* [1-3]. A number of authors [4-6] studied different aspects, physical characterization, spectroscopic and potentiometric investigations and antibacterial activity of several M (II) complexes including Mn, Ni, Fe, Zn, Cu and Co with the Schiff bases derived from 2-amino benzoic acid and salicylaldehyde. Metal Complexes with Ligands derived from aroylhydrazines and aromatic or aliphatic carbonyls gained a great interest of studies. Synthesis, characterisation and structural aspects of diorganotin(IV) complex with *N*-(5-bromo-2-hydroxybenzylidene) benzoylhydrazone [7] or *N*-

(2-Salicylidene)-*N*-benzoylhydrazone ligand [8]. Mn(II), Co(II), Ni(II), Cu(II), and Zn(II) metal complexes with NNO functionalized ligands, 2-acetyl pyridine benzoylhydrazone, etc. were synthesized and structures were elucidated by Jang *et al.* [9]. Mixed-ligand oxovanadium(IV) and oxovanadium(V) complexes incorporating the tridentate ONO donor Schiff base derived from acetylacetone and benzoylhydrazine were studied [10]. Chowdhury *et al.* [11] synthesized and characterized dioxo-uranium(VI) complexes of some aroylhydrazines and their Schiff bases with acetone.

Transition metal Schiff base complexes are used in various fields, such as medicine, agriculture, industries etc. Transition metal complexes derived from Schiff bases have been reported to have biological activity [4]. Pb(II) complex with the Schiff base derived from salicylaldehyde and *o*-phenylenediamine has been prepared and its geometry was investigated [12]. El-ajaily *et al.* [13] prepared Pb(II) complex with Schiff base derived from the reaction of salicylaldehyde and urea. Condensation and subsequent reaction with Pb(II) halides yielded complex of composition $Pb[DBQDI]$, where DBQDI is 3,5-di-tert-butyl-1,2-quinone-1-(2-hydroxy-3,5-di-tert-butylphenyl) imine anion [14]. Liu *et al.* [15] prepared $[Pb(Saloph)]$ complex by electrochemical

oxidation of anodic Pb from an acetonitrile solution of N,N-bis(salicylidene)-o-phenylenediamine. Shaker [16] presented a mixed-ligand Pb(II) complex with the general formula, $[Pb(BT)(BI)_2X_2]$, where BT= Benzotriazole, BI= benzimidazole and X=SCN. Subudhi *et al.* [17] prepared Pb(II) complexes of 4-(2-hydroxyphenylimino)phenylsulfonamide and studied its antimicrobial activities. Synthesis, characterization, antimicrobial activity and electrochemical behaviors of lead(II) complexes of thiosemicarbazone ligands have been carried out [18]. Bhattacharyya *et al.* [19] synthesized a unique Pb(II) Schiff base complex where the ligand was tripodal Schiff base $N[CH_2CH_2N=C(H)C_6H_3(OMe)-3-(OH)-2]_3$, having a distorted monocapped octahedron geometry. Lead(II) ion in the nicotinato lead(II) complex $[Pb(C_5H_4NCOO)_2]$ adopts 2-coordinate geometry, which is different from the 4-coordinate geometry of isonicotinato lead(II) complex [20].

The present work has been an attempt to study the Pb(II) complexes of some Schiff base ligands derived from aldehydes/ketones and benzoylhydrazine. The chemical structure of the prepared complexes has been elucidated by some physico-chemical studies. The antibacterial activity was studied against some gram positive and gram negative bacteria.

2. Experimental

2.1. Chemicals

2-hydroxy-1-naphthaldehyde, 2-hydroxyacetophenone and hydrazine hydrate were obtained from M/S. E. Merk (Germany). Salicylaldehyde, benzoylacetone, methanol, chloroform, N, N-dimethylformamide (DMF), dimethylsulphoxide (DMSO) and carbon-tetrachloride were obtained from Aldrich Chemical Company Ltd. Perchloric acid, nitric acid, sulphuric acid and disodium salt of ethylenediaminetetraacetic acid were obtained from BDH Chemicals Ltd. All chemicals except solvent were used as received. Solvents were dried by standard methods and distilled under an inert atmosphere.

2.2. Analytical Methods

The analysis of the metal content, Pb(II) of the prepared complexes were obtained by Atomic Absorption Spectrophotometer (Model Thermo Scientific ICE-3000) from B.C.S.I.R. Laboratories, Chittagong. Microanalyses on CHN CORDER MT-5, Yanaco, were performed in Prof. Shiumyozu's Lab. in the Dept. of Applied Chemistry at Kyushu University, Japan. The Infrared spectra of the prepared complexes were obtained by FTIR spectrophotometer (Model- 8900, Shimadzu, Japan) using KBr as the matrix in the range $400-4000\text{ cm}^{-1}$ from B.C.S.I.R. Laboratories, Dhaka. Polystyrene was used as the standard to calibrate the spectrophotometer. Electronic absorption spectra were run on Shimadzu UV-Visible Recording Spectrophotometer (Model-1800) using 1 cm cell in the research laboratory of the Department of Chemistry,

University of Chittagong. Mass spectra were obtained from Shiumyozu's Lab. in the Dept. of Applied Chemistry at Kyushu University, Japan, by Ultra-light Performance Mass Spectrometer (Model- JMS-HX 11 OA).

An electrothermal melting point apparatus was used for the determination of the melting or decomposition points of the complexes and the ligands. Using N, N-dimethylformamide (DMF) as the solvent the solutions of the complexes (of the order of 10^{-3} M) were used for conductivity measurements. Conductivity measurement was performed on a Philips Conductivity Meter (Model-HI 9255). Determination of conductivity of an electrolytic solution involves measuring of the electrical resistance of that solution at a particular temperature, usually 25°C . Magnetic susceptibility of some of the prepared complexes was determined using the Sherwood Scientific Magnetic Susceptibility Balance from Rajshahi University, Bangladesh.

2.3. Schiff Bases of Benzoylhydrazine

2.3.1. Synthesis of Benzoylhydrazine (BH)

Benzoylhydrazine (BH) is one of the starting materials for the preparation of some Schiff base ligands. This compound was prepared by the following published procedure¹¹. Ethylbenzoate (500 mmol) was added to hydrazine hydrate (500 mmol) in a round bottomed flask with a reflux condenser. It was heated under refluxed at 140°C for about 24 hours with continuous stirring using a magnetic stirrer. Then it was kept to stand overnight when white product separated out. The product so obtained was filtered off, washed with ethanol and was dried in air first and then in a desiccator over silica gel. The collected crystalline product was then re-crystallized from the same solvent. The white crystalline product of benzoylhydrazine was washed, dried in air and finally in a desiccator under vacuum over silica gel. (Yield: 80%, M.P. $112-114^\circ\text{C}$)

2.3.2. Schiff Bases of Benzoylhydrazine

Benzoylhydrazine (25 mmol) was dissolved in 30 ml of ethanol and aldehyde or ketones (25 mmol) was added drop wise to this solution with continuous stirring. The solution was refluxed for about one hour. Then it was cooled, allowed to stand for crystallization when white crystalline product separated out. The product was then filtered off, washed with ethanol and dried in a desiccator over calcium chloride. The following types of ligands have been prepared.

- Salicylaldehyde-benzoylhydrazine, Sal-BHH₂
- Bromosalicylaldehyde-benzoylhydrazine, BrSal-BHH₂
- 2-Hydroxyacetophenone-benzoylhydrazine, HAP-BHH₂
- 2-Hydroxy-1-naphthaldehyde-benzoylhydrazine, HNP-BHH₂
- Benzoylacetone-benzoylhydrazine, Bzac-BHH₂

2.5. Preparation of Complexes

For the preparation of various complexes of the Schiff base ligands, the metal ion, Pb^{2+} , was used in the form of lead acetate trihydrate, $Pb(CH_3COO)_2 \cdot 3H_2O$. Pure and dry solvent was used in all the preparations. White colored lead acetate

trihydrate [Pb(CH₃COO)₂·3H₂O] (10 mmol) was dissolved in ethanol (30 mL) with gentle warming. In another beaker, the prepared Schiff base ligand (10 mmol) was dissolved in the same solvent. The Pb(II) solution was then added to the ligand solution drop wise with continuous stirring. In maximum cases immediate precipitation occurred on mixing of the components. The mixture was refluxed for about two-three hours with continuous stirring by magnetic stirrer to complete the reaction. Then the mixture was allowed to stand and the precipitate formed was filtered off. It was then washed with ethanol and dried over calcium chloride.

2.6. Antibacterial Evaluation

For the detection of antibacterial activities and sensitivity spectrum analysis, the disc diffusion method by Gaydos *et al.* [21] was followed. Nutrient Agar (NA) was used as basal medium for culture of test bacteria and N, N-dimethylformamide (DMF) was used as the solvent to prepare the desired solution (1%) of the compounds initially. Nutrient Agar (NA) medium was prepared using the composition Beef extract (3 g), Peptone (5 g), NaCl (0.5 g), Agar (15 g) and Distilled water (1000 mL). All substances were added slowly in and they were mixed thoroughly with a glass rod and then heated to boiling for 10 minutes. After 10 minutes of boiling, the medium was transferred in 250 mL conical flasks at the rate of 200 mL per flask. The conical flasks were closed with the cotton plugs and autoclaved at 121°C and 15 psi pressure for 15 minutes and then culturing of different micro-organisms was performed.

Sensitivity spectrum analysis: Paper discs of 5 mm diameter were soaked with 10 µL of 2% solution of the test complexes. 0.2 mL of the suspension of test organism was taken in sterilized glass Petri plates of 100 cm diameter and then the molted and cooled (45°C) NA medium was poured at the rate of 10 mL per Petri plate and shaken gently. Then the discs with test complexes were placed on the seeded agar plate. A control plate was also maintained in each case with solvent. The plates were kept firstly for 24 hours at low temperature (4°C) and the test complex diffused from the disc to the surrounding medium by this time. The plates were then

incubated at 35±2°C for growth of test organisms and were observed at 24 hours interval for two days. The activity was determined by measuring the diameter of the zone of inhibition in mm.

2.7. Pathogenic Bacteria

Antibacterial activity of the prepared lead(II) complexes have been evaluated against the five selective human pathogenic bacteria such as: *Bacillus cereus* (BTCC 19, Gram-positive), *Salmonella typhi* (AE 14612, Gram-negative), *Escherichia coli* (ATCC 25922, Gram-negative), *Bacillus megaterium* (BYCC 18, Gram-positive), *Staphylococcus aureus* (ATCC 25923, Gram-positive).

3. Results and Discussion

3.1. Preparation of Schiff Bases

When an active carbonyl group reacts with -NH₂ group of this compound, a Schiff base product containing the azomethine (>C=N-) functional group is obtained. Series of Schiff base ligands were prepared using ethanol as the solvent. The ligands were washed properly but with minimum amount of solvent to protect the loss due to solubility. All the products were filtered under suction and dried in a desiccator over calcium chloride. Finally all ligands were preserved in a desiccator over silica gel.

Five dibasic tridentate Schiff bases were prepared by the condensation of benzoylhydrazine with salicylaldehyde (Sal), 5-bromosalicylaldehyde (⁵BrSal), 2-hydroxyacetophenone (HAP), 2-hydroxy-1-naphthaldehyde (HNP) and Benzoylacetone (Bzac) in their 1:1 molar ratio. HNP-BHH₂ was obtained as brownish precipitate and the Sal-BHH₂ was obtained as off-white precipitate almost immediately after mixing of the ethanolic solutions of the component reagents. On the other hand, Bzac-BHH₂ was obtained after reflux and then on cooling as off-white precipitate and HAP-BHH₂ as lemon. Physical properties and infrared Spectral bands of the prepared Schiff base ligands are in Table 1.

Table 1. Physical properties and infrared spectral bands of the prepared Schiff base ligands.

Ligands	Color	Yield %	M.P. (°C)	Infrared Spectral bands (cm ⁻¹)			
				ν _{C=N}	ν _{C=C}	ν _{C-N}	ν _{C-O}
Sal-BHH ₂	White	85	176-178	1617(s)	1542(s)	1355(ms)	1276(w)
⁵ BrSal-BHH ₂	Brown	84	180-182	1583(s)	1545(ms)	1357(w)	1271(ms)
HNP-BHH ₂	Brown	75	208-210	1620(w)	1571(w)	1363(s)	1286(w)
HAP-BHH ₂	Lemon	70	180-182	1543(s)	1555(ms)	1377(w)	1281(ms)
Bzac-BHH ₂	White	70	135-137	1580(w)	1541(w)	1343(s)	1266(w)

3.2. Preparation of Pb(II) Complexes

Pb(II) complexes of the dibasic tridentate Schiff bases were prepared by the reaction of 1:1 molar ratio of Pb(II) acetate trihydrate and the ethanolic solution of Schiff bases. The reaction mixture was concentrated to half of its volume

and the solution was kept in a refrigerator for a few days. After about a week very deep brown (almost black) precipitate was observed. The precipitate was separated out from the solution and preserved in a desiccator. Here all ligands acted as dinegative anion on removal of their two hydroxyl protons. Most of the complexes are insoluble in

methanol. Some of them are moderately soluble in chloroform. The complexes are moderately soluble in DMF. From elemental analysis it is observed that the values are in good agreement with those of calculated ones indicating the formation of the desired complexes with 1:1 metal-ligand

ratio. Some physical properties and analytical data of the prepared Schiff base complexes are in Table 2. A representative schematic diagram for the preparation of one of the Schiff base ligands and its Pb(II) complex is shown in Fig. 1.

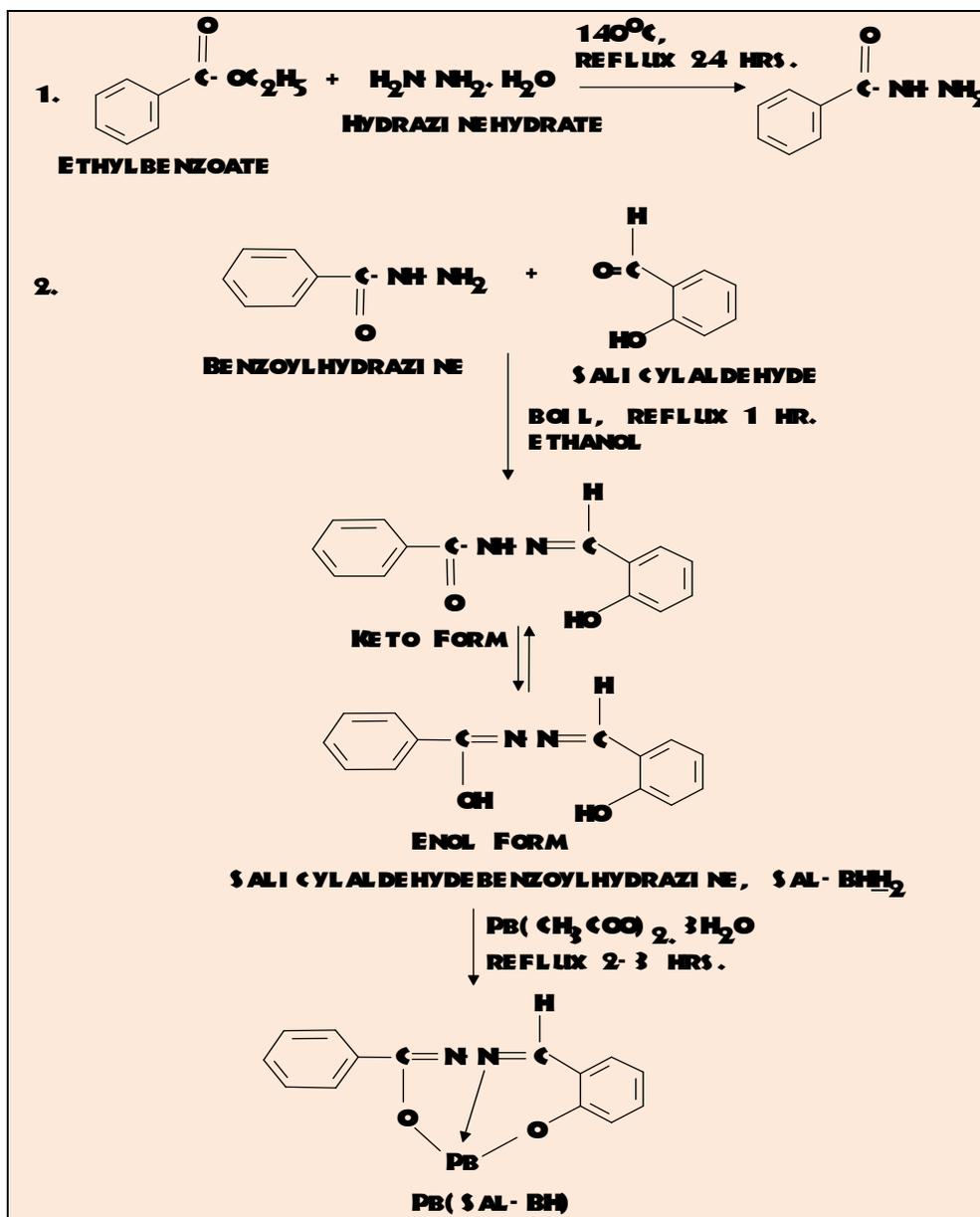


Fig. 1. A schematic diagram for the preparation of one of the Schiff bases (Sal-BHH₂) and its Pb(II) complex.

Table 2. Some physical properties and analytical data of the prepared Schiff base complexes.

Complexes	Color	M.P. (°C)	Elemental analysis (Calc.)				BM	$\lambda_{\text{m}} \text{ ohm}^{-1} \text{ cm}^2 \text{ mol}^{-1}$
			% M	% C	% H	% N		
[Pb(Sal-BH)]	Yellow	>250	45.24 (46.52)	37.48 (37.75)	2.26 (2.25)	6.24 (6.29)	Dia.	2.19
[Pb(⁵ BrSal-BH)]	Yellow	>250	38.60 (41.73)	31.50 (31.45)	1.68 (1.61)	2.78 (2.82)	Dia.	2.74
[Pb(HAP-BH)]	Lemon	>250	44.17 (45.09)				Dia.	1.82
[Pb(HNP-BH)]	Brown	>300	39.59 (41.81)				Dia.	2.28
[Pb(Bzac-BH)]	Yellow	>300	41.90 (42.68)	42.14 (42.06)	2.96 (2.89)	5.77 (5.77)	Dia.	2.98

* Calc. values are given in parentheses.

3.3. Qualitative Characterization

Primarily the complexes of were characterized qualitatively by the presence of Pb(II). A little amount of the prepared complex was taken in a test tube. Then concentrated nitric acid (HNO₃) was added to the sample and gently warmed on a water bath for the decomposition. Few drops of sulphuric acid (H₂SO₄) were added to the mixture when white precipitate of PbSO₄ was observed. The precipitate was separated out carefully from the solution. Ammonium acetate (conc.) solution was finally added. The precipitate was dissolved readily, which indicated the presence of Pb(II).

3.4. Infra-Red Spectra

For the present systems, the infrared spectra of the metal complexes provide information about their formation in respect to the presence and coordination of the ligands. The absence of any O-H and N-H vibrations indicate the complexes to contain the dibasic tridentate Schiff base as the ligand. Comparison of the metal complex spectra with those for the free ligands themselves was most useful in rationalizing the infrared results and assigning of the various stretching modes. The tentative assignments of the characteristic infrared peaks of various complexes have been made empirically by comparison of the spectra of the corresponding ligands. Tentatively made infrared band assignments of the present prepared complexes are given in Table 3. The $\nu_{\text{O-H}}$, $\nu_{\text{C=O}}$ (keto form), $\nu_{\text{C=N}}$, $\nu_{\text{C=C}}$, $\nu_{\text{M-O}}$, $\nu_{\text{N-H}}$ etc. stretching modes which are found in the ligands and their complexes have been identified by many authors [7,10]. It is a general observation that the spectral behavior of the free ligands changes by chelation but the interpretation of the band shifts depends on the attribution of the observed frequencies to the various bands, C-N, C-O etc. Fig. 2 shows the representative IR spectrum of [Pb(HNP-BH)] complex.

The present Schiff base ligands contain the $\nu_{\text{C=N}}$ modes. The peak between 1620 and 1580 cm⁻¹ have been assigned to $\nu_{\text{C=N}}$ by Aliyu and Ado [4] and Johari *et al.* [5]. The bands

appearing between 1520 and 1600 cm⁻¹ have been assigned to aromatic $\nu_{\text{C=C}}$. The $\nu_{\text{C=N}}$ modes are often mixed with higher frequency $\nu_{\text{C=C}}$ (near 1600 cm⁻¹) and are seen as a strong band. The bands appearing at 1315-1400 cm⁻¹ (specially the highest frequency ones near 1400 cm⁻¹) have been assigned to $\nu_{\text{C-N}}$ mode. Aliyu and Ado⁴ have assigned a band near 1400 cm⁻¹ to $\nu_{\text{C-N}}$ for M(II) (M=Mn, Ni, Cu) Schiff base complexes. The $\nu_{\text{C-O}}$ (Phenolic) stretching frequency of ligand seen at around 1380 cm⁻¹ gets shifted to a lower frequency region in the complexes in the range of 1372- 1326 cm⁻¹, and this is indicative of bonding through the phenolic oxygen [6]. On the basis of these studies, the band observed for the present complexes in the regions of 1580-1670, 1520-1600, 1360-1401 and 1260-1300 cm⁻¹ have been assigned as due to $\nu_{\text{C=N}}$, aromatic $\nu_{\text{C=C}}$, $\nu_{\text{C-N}}$ and $\nu_{\text{C-O}}$, respectively. Strong bands at 1606 cm⁻¹ can be attributed to $\nu(\text{C=N-N=C})$ stretching [7]. The N-N stretching frequency is observed at 910-970 cm⁻¹ for the free ligands which generally suffers a positive shift of 55-100 cm⁻¹ in the spectra of the complexes [7]. Comparing with the ligand bands the $\nu_{\text{N-N}}$ bands have been assigned in the region 900-950 cm⁻¹ for the prepared complexes. Chowdhury *et al.* [11] have prepared Schiff base complexes where $\nu_{\text{N-N}}$ frequency was observed at 910-950 cm⁻¹. The positive shift of the $\nu_{\text{N-N}}$ band upon coordination with the lead ion may be due to the reduction of repulsion between lone pairs of electrons on the nitrogen atoms.

The presence of $\nu_{\text{C=N}}$ and $\nu_{\text{C-O}}$ modes in the ligand and their negative shift on complexation confirm that the ligands act as N- donor and O- donor bases. El-ajaily *et al.* [12,13] have prepared and investigated new Schiff base complexes of Cr(II), Pb(II) and TiO(IV) and observed new bands at 575-671 cm⁻¹ and 444-540 cm⁻¹ in the spectrum which were attributed to $\nu_{\text{M-O}}$ and $\nu_{\text{M-N}}$ vibrations, respectively For the complexes, the presence of $\nu_{\text{Pb-N}}$ vibration in the region 575-671 cm⁻¹ indicates coordination of N- donors and $\nu_{\text{Pb-O}}$ vibrations in the region 444-540 cm⁻¹ indicate the O- donor character of the ligands to the lead(II) ion.

Table 3. Spectral bands (infrared and electronic) of some Schiff base complexes of lead (II).

Complexes	Infrared Spectral Bands (cm ⁻¹)						Electronic Spectral Bands (nm) (DMF/CHCl ₃)
	$\nu_{\text{C=N}}$	$\nu_{\text{C=C}}$	$\nu_{\text{C-N}}$	$\nu_{\text{C-O}}$	$\nu_{\text{Pb-O}}$	$\nu_{\text{Pb-N}}$	
[Pb(Sal-BH)]	1596(s) 1544(vs)	1544(s) 1498(s)	1352(s) 1367(s)	1267(s) 1298(ms)	617(w) 650(w)	466(w) 516(ms)	473,408,322, 296,287,276
[Pb(⁵ BrSal-BH)]	1577(w) 1604(ms)	1521ms 1541(sh)	1371ms 1299 ms	1269(sh) 1284(sh)	590(w) 634(ms)	491ms 540(w)	443, 388, 342
[Pb(HAP-BH)]	1577(w)	1544(s)	1344(ms) 1363(sh)	1286(ms) 1313(ms)	617(w) 646(ms)	455(s) 493(s)	382,325,323
[Pb(HNP-BH)]	1595(w) 1566(w)	1541(s)	1355(ms) 1388(w)	1284(s) 1242(s)	615(w) 648(s)	447(s) 464(s)	436,342,334, 317,386,286
[Pb(Bzac-BH)]	1587(s) 1618(w)	1564(ms) 1514(vw)	1363((sh) 1375(ms)	1276(s) 1299(s)	617(w) 632(ms)	484(s) 518(ms)	429,380,350

* vs = very strong; s = strong; ms = medium strong; m = medium; w = weak; vw = very weak; sh = shoulder

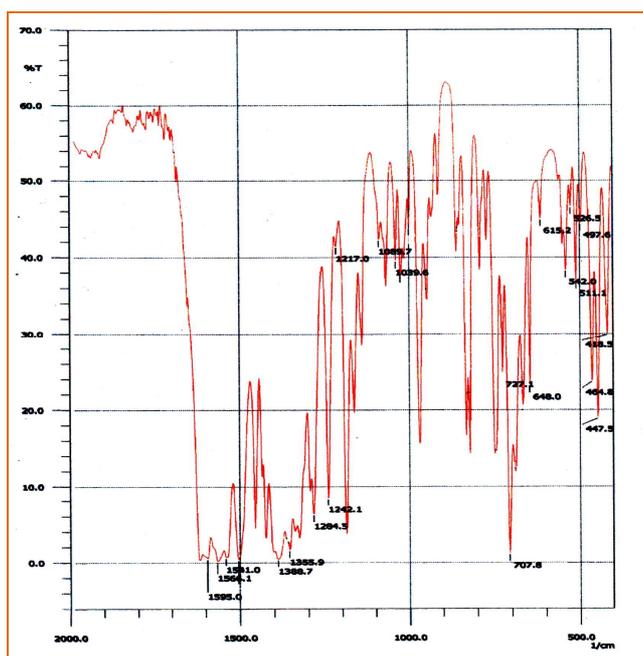


Fig. 2. Representative IR spectrum of [Pb(HNP-BH)] complex

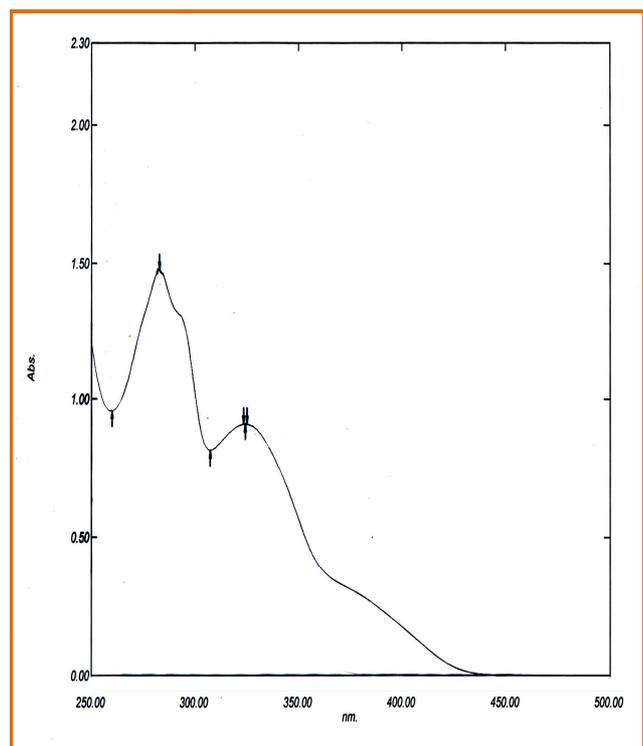


Fig. 3. Electronic spectra of [Pb(HAP-BH)] complex.

3.5. Mass Spectra

Mass spectra of the compounds, [Pb(Sal-BH)] and [Pb(Bzac-BH)] were obtained. The spectra, however, didn't look that clean and explicit as lead has isotopes ranging from $^{82}\text{Pb}^{194}$ to $^{82}\text{Pb}^{214}$. The spectrum of [Pb(Sal-BH)] complex shows the molecular ion peak at m/e 446 and 447. The peak

observed at m/e 419 is thought to be due to the loss of N_2 . The peak at m/e 307 is assigned to due to loss of $\text{C}_7\text{H}_5\text{O}$. The spectrum of [Pb(Bzac-BH)] complex shows the molecular ion peak at m/e 486 and 485. The peak observed at m/e 460 is thought to be due to the loss of N_2 . The peak at m/e 307 is assigned to due to the loss of $\text{C}_{10}\text{H}_9\text{O}$. The mass spectra of the complexes indicated formation of the titled complexes.

3.6. ^1H NMR Spectra

The ^1H NMR spectrum of the prepared complex [Pb(Sal-BH)] was measured and interpreted. The phenyl multiplet is seen at 6.5 - 7.3 δ and the azomethine ($\text{N}=\text{CH}$) proton is seen at 7.6-8.76 δ (singlet). The peak at 7.85-10.8 δ is attributed to phenolic OH group present in the ligand which is being absent in respective complex. These are comparable with the reported values [6].

3.7. Electronic Spectra

Being a d^{10} system, the present Pb(II) complexes show no d-d transition in the visible region, and are colored only through their intense charge transfer absorptions tailing in from the ultraviolet. Besides, the other bands are due to intra-ligand transitions. The peaks observed below 350 nm are tentatively assumed as due to $\pi \rightarrow \pi^*$, $n \rightarrow \pi^*$ and $n \rightarrow \sigma^*$ transitions. The peaks observed above 350 nm are assumed to be due to charge transfer [13]. Fig. 3 shows the electronic spectra of [Pb(HAP-BH)] complex.

3.8. Magnetic Measurements

From the experimental values of magnetic measurements of some of the prepared complexes, it is revealed that lead(II) complexes of the tridentate Schiff bases are diamagnetic thus indicating the d^{10} electronic configuration of Pb(II) in such complexes.

3.9. Molar Conductance

The molar conductance values of the presently prepared complexes are shown in Table 2. The low conductance values of the prepared complexes in DMF solutions indicate their non-electrolytic nature. This suggests +2 oxidation state of lead which is satisfied by the dinegatively charged tridentate ligands as expected.

3.10. Microbial Activities

It is known from literature that a large number of biologically active compounds possess aromatic and heteroaromatic nucleus. It is also known that, if an active nucleus is linked to another nucleus, the resulting molecule may possess greater potential for biological activity. It is recognized that nitrogen and sulphur containing heterocyclic compounds show marked microbial activities. Antibacterial and antifungal activities of a series of dithiocarbamate and Schiff base complexes have been carried out in this laboratory [22,23]. In the present investigation, antibacterial

activity of the prepared lead(II) complexes have been evaluated against the five selective human pathogenic bacteria as mentioned in experimental section. The inhibition zone of test organisms for different complexes is presented in Fig. 4. All the tested complexes are quite effective against *Bacillus cereus*, *Salmonella typhi* and *Staphylococcus aureus*. On the other hand, they were not effective against *Escherichia coli* and *Bacillus megaterium*. All the tested complexes are quite effective against *Bacillus cereus*, *Salmonella typhi* and *Staphylococcus aureus*. On the other hand, they were not effective against *Escherichia coli* and *Bacillus megaterium*.

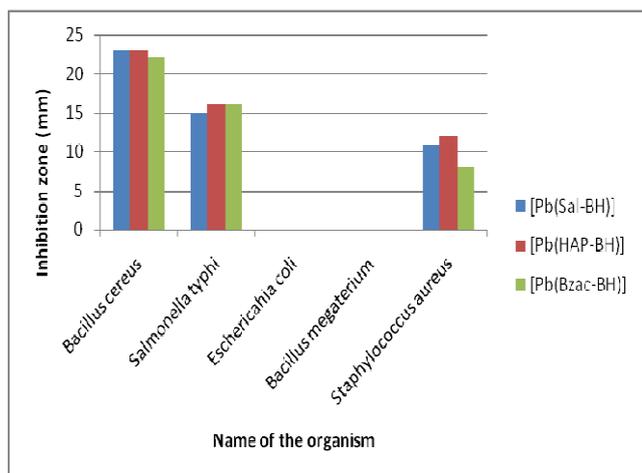


Fig. 4. Zone of inhibition produced by the prepared complexes against the test organism.

Different effect of the compounds against microorganism may be because of the structure of the compound, types of ligands and also the nature of solvent. The diffusion capacity of the compounds varies with the employed solvent, which may be because of the polarity of the solvent. This high antibacterial activity of complexes can be attributed due to the chelation trends to make a liquid more potential bacterial agent. The increased activity depends upon chelation is attributed to the positive charge of the metal partially shared with donor atom present on the Ligand and possible π -electron delocalization over the whole chelate ring.

4. Conclusion

The Schiff bases were derived by the condensation of o-hydroxyaldehydes or ketones with amines. The prepared Pb(II) complexes with such ligands have been found to be of the type PbL (where, LH₂ = dibasic tridentate ligands). The analytical data indicate that the complexes have 1:1 (metal: ligand) stoichiometry. Conductivity measurement indicates their non-electrolytic nature. The magnetic measurement along with electronic spectral data of the prepared complexes support 5d¹⁰ electronic configuration of the central metal ion consistent with the +2 oxidation state of lead ion. The IR data and mass spectra are indicative of the monomeric structures of the complexes. The Schiff base ligands possess a planar configuration with respect to the positions of the donor atoms

or ions. The structures of the prepared PbL complexes seem to be trigonal pyramidal with the 6s² electron pair in the 4th position of the sp³ hybrids. However, it is difficult to suggest the exact structure of any of the prepared compounds without crystal structural evidences.

Acknowledgement

Authors are pleased to acknowledge the Department of Microbiology, University of Chittagong, Bangladesh for cordial cooperation in collecting bacterial strains and performing antibacterial tests.

References

- [1] Syamal, A. & Maurya, M. R. (1986). Synthesis and characterisation of nickel(II), cobalt(II), manganese(II), copper(II), zinc(II), dioxouranium(VI) and dioxomolybdenum(VI) complexes of tridentate dibasic ONO donor Schiff bases derived from 2-benzothiazolecarbohydrazide and salicylaldehyde/2-hydroxy-1-naphthaldehyde. *Transition Metal Chem.*, 11(6), 201-205.
- [2] Syamal, A., & Maurya, M. R. (1986). Dioxomolybdenum(VI) complexes with tridentate dibasic Schiff bases derived from various hydrazides. *Transition Metal Chem.*, 11(6), 235-238.
- [3] Syamal, A. & Maurya, M. R. (1986). Synthesis and characterization of new dioxomolybdenum(VI) complexes of ONS donor Schiff bases derived from thiosemicarbazide, S-methyldithiocarbamate, S-benzylidithiocarbamate and o-hydroxy-aromatic aldehydes/ketones. *Transition Metal Chem.*, 11(7), 255-258.
- [4] Aliyu, H. N. & Ado, I. (2010) Studies of Mn(II), and Ni(II) complexes with Schiff base derived from 2 amino benzoic acid and salicylaldehyde. *Bayero J. Pure Appl. Sci.*, 3(1), 245 – 249.
- [5] Johari, R., Kumar, G., Kumar, D. & Singh, S. (2009). Synthesis and antibacterial activity of M (II) Schiff base complex. *J. Ind. Council Chem.*, 26(1), 23-27.
- [6] Mounika, K., Anupama, B., Pragathi, J. & Gyanakumari, C. (2010). Synthesis, characterization and biological activity of a Schiff base derived from 3-ethoxy salicylaldehyde and 2-amino benzoic acid and its transition metal complexes. *J. Sci. Res.* 2(3), 513-524.
- [7] Samanta, B., Chakraborty, J., Dey, D. K., Gramlich, V. & Mitra, S. (2007). Synthesis, characterization and structural aspects of a new diorganotin(IV) complex with N-(5-bromo-2-hydroxybenzylidene) benzoylhydrazone ligand. *Struct Chem.*, 18, 287–293.
- [8] Lalia-Kantouri, M., Tzavellas, L. & Paschalidis, D. (2008). Novel lanthanide complexes with di-2-pyridyl ketone-p-chloro-benzoylhydrazone, Thermal investigation by simultaneous TG/DTG-DTA and IR spectroscopy. *J. Thermal Anal. Calori.*, 91(3), 937–942.
- [9] Jang, Y. J., Lee, U. K. & Koo, B. K. (2005). Synthesis and crystal structures of Mn(II), Co(II), Ni(II), Cu(II), and Zn(II) metal complexes with NNO functionalized ligands. *Bull. Korean Chem. Soc.*, 26(6), 925-929.

- [10] Ghosh, T., Bandyopadhyay, C., Bhattacharya, S. & Mukherjee, G. (2004). Synthesis, spectral and electrochemical studies of mixed-ligand oxovanadium(IV) and oxovanadium(V) complexes incorporating the tridentate ONO donor schiff base derived from acetylacetone and benzoylhydrazine. *Transition metal chem.*, 29(4), 444-450.
- [11] Chowdhury, D. A., Uddin, M. N. & Sarker, M. A. H. (2008). Synthesis and characterization of dioxo-uranium(VI) complexes of some aroylhydrazines and their Schiff bases with acetone. *Chiang Mai J. Sci.* 35(3), 483-494.
- [12] El-Ajaily, M. M., Abdseed, F. A. & Gweirif, B. (2007). Preparation, characterization and antibacterial activity of some metal ion complexes. *E-Journal of Chem.*, 4, 461-466.
- [13] Abdseed, F. A. & El-ajaily, M. M. (2009). Preparation and spectroscopic investigation of a Schiff base metal complexes. *Inter. J. Pharm Tech Res.*, 1(4), 1097-1103.
- [14] McGaryey, B. R., Ozarowski, A., Tian, Z. & Tuck, D. G. (1995). Tin and lead derivatives of a Schiff Base-bis(orthoquinone) ligand. *Can. J. Chem.* 73, 1213-1222.
- [15] Liu, J., Zhang, B., Bowan, W. U., Zhang, K. & Shenli, H. U. (2007). The direct electrochemical synthesis of Ti(II), Fe(II), Cd(II), Sn(II), and Pb(II) complexes with N, N-bis(Salicylidene)-o-phenylenediamine. *Turk J Chem*, 31, 623-629.
- [16] Shaker, S. A. (2010). Preparation and study of some Mn(II), Co(II), Ni(II), Cu(II), Cd(II) and Pb(II) complexes containing heterocyclic nitrogen donor ligands. *E-Journal of Chem.*, 7(4), 1598-1604.
- [17] Subudhi, B. B., Panda, P. K. & Sahoo, S. (2007). Synthesis and Evaluation of Antimicrobial Activity of Metal Complexes of 4-(2'-Hydroxy Phenyl Imino)Phenyl Sulphonamide. *Iranian J. Pharm. Sci.*, 3(4), 245.
- [18] Malik, S., Kumari, M., Chauhan, S. & Sharma, D. K. (2010). Sulfur-containing Schiff base complexes of iron(III) and lead(II): Synthesis, characterization, antimicrobial activity, and their electrochemical behaviors. *Phosphorus, Sulfur, and Silicon and Related Elements*, 185(8), 1759 – 1771.
- [19] Bhattacharyya, P., Parr, J., Ross, A.T. & Slawin, A. M. Z. (1998). First synthesis of a unique dilead Schiff base complex. *J. Chem. Soc., Dalton Trans.*, 19, 3149-3150.
- [20] Zhao, P. S., Li, R. Q., Song, J. & Guo, M. P. (2008) Quantum chemical studies on nicotinato lead(II) complex [Pb(II)(C₅H₄NCOO)₂], *Bull. Korean Chem. Soc.*, 29(3), 546-550.
- [21] Gaydos, J. M. & Harrington, B. J. (1982). Agar disk diffusion for the quality control testing of Autobac elution disks. *Antimicrob Agents Chemother.* 21(3), 516–518.
- [22] Chowdhury, D. A., Uddin, M. N. & Lucky, N. (2003). Antimicrobial activities of some dithiocarbamates and their Titanium(IV) complexes. *Bull. Pure Appl. Sci.*, 22C(1), 39-45.
- [23] Chowdhury, D. A., Uddin, M. N., Islam, M. T., & Haque, F. (2012). Evaluation of biological activity of dioxouranium complexes of some Schiff base and dithiocarbamate ligands. *Orbital-The electronic J. Chem.*, 4(4), 273-287.