



Synthesis, Characterization and Biological Activity of Mn(II), Co(II), Ni(II), Cu(II), Zn(II), Pd(II), Cd(II), Pt(II), Hg(II) and Pb(II) Complexes of 2-Methyl-6-(3-phenylthioureido)benzoic Acid

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In this work, the synthesis, characterization and biological activities of Mn(II), Co(II), Ni(II), Cu(II), Pd(II), Pt(II), Hg(II) and Pb(II) complexes with 2-methyl-6-(3-phenylthioureido)benzoic acid (BH) are reported. The synthesized ligand (BH) and its metal complexes were characterized by elemental analysis, infrared and electronic spectral data, magnetic susceptibility and molar conductance. The biological activities of the synthesized complexes were tested against *Staphylococcus aureus*, *Escherichia coli* and *Pseudomonas aeruginosa* using a well diffusion method. Dunnett method was used to compare the biological activities of the complexes with the control.

Key Words: Thiourea derivative, Transition metal complexes, Biological activity.

INTRODUCTION

Thiourea and its derivatives have been shown to possess antibacterial, antifungal, antitubercular, antithyroid and insecticidal properties^{1,2} and have wide applications in wood protection³, rubber industry, photography and agriculture⁴.

Study of the thiourea derivatives has attracted the attention of many research groups due to their interest as selective ligands for the concentration and separation of metal cations⁵. Thiourea derivatives are selective analytical reagents, especially for the determination of transition metals, in complex interfering matrices⁶. Thiourea moieties are important chemical building blocks which have numerous chemical and pharmaceutical applications⁷. The complexing capacity of thiourea derivatives have been reported in several papers^{6,8-10}. Transition metal complexes of thiourea derivatives have been an active area of some recent researches¹¹.

Thiourea and its derivatives form a variety of complexes of different symmetries with various ions¹². The structures of several types of thioureas derivatives and its several metal complexes have been determined during the past decades^{2,12-27}. It is well known that nitrogen and sulfur atoms, which are present in thiourea and its derivatives, play a key role in the coordination of metals at the active sites of numerous metallo-biomolecules^{28,29}.

The biological activity of complexes with thiourea derivatives have been successfully screened for various biological actions^{6,8,10,11,22,23}. From the literature review, it is obvious that little work has been conducted on the chemistry of thiourea

derivatives of anthranilic acid, their metal complexes and their biological activities. In this work, 2-methyl-6-(3-phenylthioureido)benzoic acid complexes of Pd(II), Pt(II), Co(II), Cu(II), Cd(II), Mn(II), Zn(II), Ni(II), Hg(II) and Pb(II) have been synthesized and characterized.

EXPERIMENTAL

Phenylisothiocyanate, potassium chloride, potassium bromide, $K_2[PtCl_4]$ and $NiCl_2 \cdot 6H_2O$ were purchased from Fluka AG. $Zn(II)$ chloride, benzene, ether and acetone are Merck products. $PdCl_2$, $CoCl_2 \cdot 6H_2O$, $CdCl_2 \cdot 6H_2O$, $HgCl_2$, $Pb(CH_3COO)_2 \cdot 4H_2O$, $CuCl_2 \cdot 2H_2O$, 2-amino-6-methylbenzoic acid from ACROSS products and $MnCl_2 \cdot 2H_2O$ were supplied by BDH. Melting points were measured using Toshinwal electro-thermal melting point apparatus. Electronic spectra of the ligand and the complexes were measured in DMSO using a Jenway 6485 spectrophotometer. The Infrared spectra of the ligands and complexes were recorded on a Shimadzu infrared spectrophotometer in the $4000-200\text{ cm}^{-1}$ range using potassium iodide discs. Elemental analysis was carried out on Perkin-Elmer 2400 CHN/O analyzer in Al AL-bayt University IIES/central Lab. The conductivities of the complexes were measured in DMSO using conductometer type; HANNA instruments, model EC215 Conductivity Meter. Magnetic measurements were recorded on a Bruker BM6 instrument at room temperature following Faraday method.

Preparation of the ligand: 2-methyl-6-(3-phenylthioureido)benzoic acid ($C_{15}H_{14}N_2O_2S$): (BH): The thiourea

ligand, 2-methyl-6-(3-phenyl-thioureido)benzoic acid, was prepared using a modified literature method¹⁷.

Preparation of the ligand (BH) metal complexes

[Mn(B)₂]: A solution of the ligand (BH) (0.1170 g, 0.4090 mmol) in a (1:1) mixture of ethanol and dichloromethane (10 mL) containing 2 mL of triethylamine was added dropwise to a solution of Mn(CH₃COO)₂·4H₂O (0.0501 g, 0.2045 mmol) in ethanol (10 mL). The solution mixture was stirred at room temperature for 24 h then cooled in an ice bath and a brown precipitate was obtained which was filtered off, washed with ethanol then with diethyl ether and dried under vacuum.

[Co(B)₂], [Ni(BH)₂], [Cu(B)₂] and [Zn(B)₂]: A solution of the corresponding metal acetate (or a metal chloride in case of Ni(II) (0.2045 mmol) in methanol (5-10 mL) was added dropwise to a solution of the ligand (BH) (0.1170 g, 0.4090 mmol) in a (1:1) mixture of methanol and dichloromethane (10 mL) containing three drops of triethylamine. The solution mixture was stirred at room temperature for 1 h (Zn(II) for 2.5 h) then cooled in an ice bath and the solid formed was filtered off washed with methanol, diethyl ether and dried under vacuum.

[Cd(B)₂], [Pb(B)₂] and [Hg(BH)Cl₂]: A solution of a metal acetate (or a metal chloride in case of Hg(II)) (0.2045 mmol) in methanol (5-10 mL) was added dropwise to a solution of the ligand (BH) (0.117 g, 0.4090 mmol) in a (1:1) mixture of methanol:dichloromethane (10 mL). The solution mixture was stirred at room temperature for 1 h. Then cooled in ice bath and the solid formed was filtered off washed with methanol, diethyl ether and dried under vacuum.

[Pt(B)₂] and [Pd(BH)₂]Cl₂: A solution of K₂[PdCl₄] (0.0652 g, 0.20 mmol) in acetone (5 mL) or Na₂[PtCl₄] (0.0766 g, 0.20 mmol) in distilled water (5 mL) was added to a warm solution of the ligand (BH) (0.114 g, 0.40 mmol) in acetone (5 mL). The mixture was stirred at room temperature for 1.5 h then cooled in ice bath. The solid formed was filtered off washed with acetone (or cold distilled water for Pd(II)) and dried under vacuum.

Biological activity of BH ligand and its metal complexes:

Biological activity of the ligand, BH and its metal complexes have been screened against gram positive and gram negative bacteria as *E. coli* (gram negative), *P. aeruginosa* (gram negative) and *S. aureus* (gram positive) using Petri dish containing Muller-Hinton agar discs (9 cm in diameter) by well-diffusion method³⁰ Muller-Hinton agar used as bacterial nutrient medium and the well diameter is *ca.* 7 mm, using DMSO as solvent, the discs-well were dripped with DMSO as solvent control and test compound solutions of certain concentration of (0.25 mL of 1 × 10⁻³ M). The dripped discs were incubated at 35 °C for 24 h. The antibacterial activity was estimated on the bases of the size of inhibition zone formed around the well of the agar discs in mm.

RESULTS AND DISCUSSION

The ligand 2-methyl-6-(3-phenylthioureido)benzoic acid (BH) is a white solid, soluble in acetone, (1:1) mixture of methanol or ethanol and dichloromethane, but it is insoluble in water. The prepared ligand (BH) was characterized by means of melting point, IR-spectroscopy and elemental analysis. The

elemental analysis data of this ligand (Table-1) is in a well agreement with the suggested formula. The infrared spectrum of the prepared ligand (Table-2) shows four characteristic bands from medium to very strong intensity; 1543, 1415, 984 and 786 cm⁻¹ which are attributed to the thioamide (HNC=S) bands I, II, III and IV, respectively. These bands gave contributions from the groups as follows: band (I) has contributions from δ(N-H) + δ(C-H) + ν(C=N), band (II) has contributions from ν(C=S) + ν(C=N) + δ(C-H), band (III) has contribution from ν(C=S) + ν(C-N) and band(IV) has contribution from ν(C=S)^{11,31-33}. Also the IR-spectrum shows bands at 3414, 3182 and 3125 cm⁻¹, which are assigned to ν(O-H) vibration frequency of the carboxylic group and two ν(NH) secondary amine of thiourea ligand, respectively³⁴⁻³⁷.

Reaction of the deprotonated anion ligand (B⁻) with metal salts of Mn(II), Co(II), Ni(II), Cu(II), Zn(II), Cd(II), Pt(II) and Pb(II) in an alcoholic solvent containing triethylamine gave complexes of the type [M(B)₂], while reaction of the ligand (BH) with metal salts of Pd(II) and Hg(II) in acetone or alcoholic solvent gave complexes of the type [Pd(BH)₂]·Cl₂ and [Hg(BH)Cl₂].

The elemental analysis data for the prepared complexes are given in Table-1, which are consistent with the suggested stoichiometries. The colour, melting points and molar conductivities of the metal complexes are shown in Table-1.

The molar conductivities of the complexes **1-5** and **7-10** in DMSO (Table-1) are too low suggesting that they are non-electrolytes. The complex **6** has a conductivity of (73.27 Ω⁻¹ cm² mol⁻¹) which indicates that it is an electrolyte of the type (1:2)^{38,39}.

The main characteristic band frequencies are listed in Table-2 together with their assignments.

Thiourea derivatives and their metal complexes containing thioamide moiety (HNC=S) usually show four characteristic bands in their infrared spectra^{11,32,40-42}. These bands are; thioamide band I of medium to very strong intensity in the range 1497-1556 cm⁻¹, thioamide band II of weak to very strong intensity in the range 1415-1468 cm⁻¹, thioamide band III of very weak to strong intensity in the range 985-1022 cm⁻¹ and thioamide band IV which has medium to very strong intensity in the range 786-804 cm⁻¹. The ligand (BH) has four thioamide bands; 1542 (vs), 1415 (vs), 983 (m) and 786 (vs) as shown in Table-2. Upon coordination of the ligand to the metal ions in the prepared complexes **1-10**, its four thioamide bands are shifted either to higher or lower frequencies and their intensities are either decreased or increased as shown in Table-2.

The infrared spectrum of the ligand and its metal complexes **1-10** showed a broad band at the range of 3416-3414 cm⁻¹ which is attributed to ν(OH), Table-3³⁵⁻³⁷, this means that the ligand is not coordinated through the oxygen of its hydroxyl group to the metal centers in these complexes.

The infrared spectrum of the free ligand shows two bands at 3182 and 3125 cm⁻¹ which are assigned to two secondary amines of thioureido group^{11,43}.

The infrared spectra of the metal complexes **1, 2, 3, 4, 5, 7, 8** and **10** (Table-2), show a very weak band in the range 3255-3225 cm⁻¹ which is attributed to one secondary amine, but the other secondary amine band was disappeared upon coordination due to its deprotonation. The infrared spectrum

TABLE-1
COLOUR, MELTING POINT, ELEMENTAL ANALYSIS AND CONDUCTIVITY DATA FOR THE PREPARED COMPLEXES (1-10)

No.	Compounds	m.f.	Colour	m.p. (°C)	Elemental analysis (%): Found (Calcd.)				Λ_m (Ω^{-1} cm ² mol ⁻¹)
					C	H	N	S	
	BH	(C ₁₅ H ₁₄ N ₂ SO ₂)	White	185-187	63.11 (62.92)	4.85 (4.93)	10.03 (9.78)	10.96 (11.17)	1.5
1	[Mn(B) ₂]	[Mn(C ₁₅ H ₁₃ N ₂ SO ₂) ₂]	Brown	237-241	58.23 (57.58)	3.52 (4.19)	8.50 (8.96)	9.96 (10.24)	6.8
2	[Co(B) ₂]	[Co(C ₁₅ H ₁₃ N ₂ SO ₂) ₂]	Pale brown	>300	56.99 (57.23)	3.74 (4.16)	9.40 (8.90)	9.90 (10.18)	12.41
3	[Ni(B) ₂]	[Ni(C ₁₅ H ₁₃ N ₂ SO ₂) ₂]	Pale green	330d	57.75 (57.25)	3.39 (4.16)	9.34 (8.90)	9.85 (10.19)	10.2
4	[Cu(B) ₂]	[Cu(C ₁₅ H ₁₃ N ₂ SO ₂) ₂]	Grassy	>325d	56.64 (56.81)	3.59 (4.13)	8.41 (8.83)	9.68 (10.11)	7.82
5	[Zn(B) ₂]	[Zn(C ₁₅ H ₁₃ N ₂ SO ₂) ₂]	White	>348d	58.82 (56.78)	3.89 (4.12)	9.18 (8.83)	10.00 (10.08)	6.97
6	[Pd(BH) ₂]Cl ₂	[Pd(C ₁₅ H ₁₄ N ₂ SO ₂) ₂]Cl ₂	Pale brown	>275d	47.93 (48.04)	3.36 (3.76)	7.14 (7.47)	7.91 (8.55)	73.27
7	[Cd(B) ₂]	[Cd(C ₁₅ H ₁₃ N ₂ SO ₂) ₂]	White	>360d	53.21 (52.75)	3.46 (3.84)	8.51 (8.20)	9.36 (9.39)	7.9
8	[Pt(B) ₂]	[Pt(C ₁₅ H ₁₃ N ₂ SO ₂) ₂]	Yellow	>322d	47.34 (47.06)	3.38 (3.42)	6.98 (7.32)	7.97 (8.37)	9.26
9	[Hg(BH)Cl ₂]	[Hg(C ₁₅ H ₁₄ N ₂ SO ₂)Cl ₂]	White	>360d	32.64 (32.30)	2.51 (2.53)	5.38 (5.02)	5.41 (5.74)	17.25
10	[Pb(B) ₂]	[Pb(C ₁₅ H ₁₃ N ₂ SO ₂) ₂]	White	312	46.01 (46.32)	3.08 (3.37)	7.48 (7.20)	8.41 (8.24)	7.57

TABLE-2
INFRARED ASSIGNMENTS OF MAJOR BANDS FOR THE PREPARED LIGAND (BH) AND ITS COMPLEXES (1-10)

No.	Thioamide bands				v(O-H)	v(N-H)	v(C-H)	v(C=O)	v(M-S)	v(M-O)	v(M-Cl)
	I	II	II	IV							
BH	1542(vs)	1415(vs)	983(m)	786(vs)	3414(b)	3182(w) 3125(w)	3034(w) 2979(w)	1702(vs)	-	-	-
1	1541(vs)	1416(m)	985(w)	791(s)	3414(b)	3233(w)	3057(w) 2979(w)	1700(vs)	392(w)	553(m)	-
2	1508(vs)	1423(m)	993(m)	804(m)	3414(b)	3235(vw)	3050(vw) 2924(w)	1661(s)	397(vw)	640(m)	-
3	1528(vs)	1418(m)	994(w)	797(s)	3414(b)	3235(vw)	3063(vw) 2925(w)	1689(vs)	392(vw)	644(m)	-
4	1517(vs)	1421(s)	992(w)	798(s)	3414(b)	3250(vw)	3061(vw) 2924(w)	1691(vs)	405(vw)	643(s)	-
5	1497(s)	1423(w)	991(w)	803(s)	3414(b)	3255(vw)	3055(vw) 2925(w)	1698(vs)	405(vw)	642(w)	-
6	1531(m)	1468(vs)	1005(vw)	795(m)	3414(b)	3290(w) 3233(w)	3092(w) 2926(w)	1673(vs)	393(m)	642(m)	-
7	1511(vs)	1426(w)	989(w)	799(m)	3416(b)	3250(vw)	3050(vw) 2926(vw)	1689(vs)	359(vs)	639(m)	-
8	1529(s)	1421(w)	995(vw)	793(s)	3414(b)	3225(vw)	3055(vw) 2924(m)	1687(vs)	395(s)	616(m)	-
9	1556(vs)	1453(m)	10022(s)	790(s)	3416(b)	3216(w) 3160(w)	3056(w) 2985(vw)	1720(vs)	413(vw)	582(m)	318(s) 272(m)
10	1509(vs)	1447(m)	987(m)	800(s)	3415(b)	3245(vw)	3057(vw) 2922(w)	1683(vs)	405(m)	634(m)	-

of each metal complexes **6** and **9** shows two bands at 3290 and 3233 cm⁻¹, and 3216 and 3160 cm⁻¹, respectively which are assigned to $\nu(\text{NH})$ of the two secondary amines of the thiourea ligand^{6,26,34}.

The infrared spectra of the ligand and all the prepared complexes showed two bands of very weak to medium in the range 2980-2922 and 3092-3034 cm⁻¹ which are attributed to $\nu(\text{C-H})$ of the methyl group and aromatic ring, respectively^{35-37,44}.

The infrared spectrum of the ligand shows a very strong bands at 1702 cm⁻¹, Table-2, which is attributed to $\nu(\text{C=O})$ ^{35-37,44}.

The metal complexes **1**, **2**, **3**, **4**, **5**, **6**, **7**, **8** and **10**, show strong to very strong intensities bands in the range 1661-1698 cm⁻¹ which are attributed to $\nu(\text{C=O})$ ^{35-37,44}. Upon coordination of the thiourea to the metal centers the $\nu(\text{C=O})$ bands are shifted to lower frequencies^{6,20,34}. The metal complex **9**, has a strong band at 1720 cm⁻¹ which is assigned to $\nu(\text{C=O})$ ^{35-37,44} and it is shifted to a higher frequency if compared with that of the free ligand^{6,20,34}.

The IR spectra of all the metal complexes **1-10**, show weak to medium intensity bands in the range 644-536 cm⁻¹ (Table-2), which are attributed to $\nu(\text{M-O})$ vibrations indicates that the

TABLE-3
ELECTRONIC SPECTRAL BANDS AND MAGNETIC SUSCEPTIBILITY OF THE PREPARED COMPLEXES (1-10)

No.	Complex	ϵ (L mol ⁻¹ cm ⁻¹)	Band absorption		Assignment	μ_{eff} (BM)
			cm ⁻¹	nm		
1	[Mn(B) ₂]	5760	30769	325	C.T.	5.78
2	[Co(B) ₂]	19700	40650	246	C.T.	4.65
		15000	34843	287	C.T.	
		66	15383	650	⁴ A ₂ (F) → ⁴ T ₁ (P)	
3	[Ni(B) ₂]	14304	40650	246	C.T.	3.55
		10000	33557	298	C.T.	
		4224	27397	365	A _{2g} → ³ T ₁ (P)	
		21.5	16474	607	³ T ₁ (F) → ³ T ₁ (P)	
		22.2	14577	686	³ T ₁ (F) → ³ T ₁ (P)	
4	[Cu(B) ₂]	24750	40000	250	C.T.	1.77
		18050	33113	302	C.T.	
		6250	24096	415	Internal ligand transition C.T., ² B _{1g} → ² E _g	
		361	23256	430	² B _{1g} → ² A _{1g}	
5	[Zn(B) ₂]	3500	30864	324	C.T.	0
6	[Pd(BH) ₂ Cl] ₂	332			C.T.	0
		15264	40000	250	¹ A _{1g} → ¹ A _{2g}	
			33898	295	¹ A _{1g} → ¹ B _{1g}	
7	[Cd(B) ₂]	164	41152	243	C.T.	0
		7112	30769	325	C.T.	
8	[Pt(B) ₂]	405	39024	256	C.T.	0
		11981	33784	296	¹ A _{1g} → ¹ A _{2g}	
					¹ A _{1g} → ¹ B _{1g}	
9	[Hg(BH)Cl] ₂	246	39024	256	C.T.	0
		6860	30675	326	C.T.	
10	[Pb(B) ₂]	353	42194	237	C.T.	0
		6564	30864	324	C.T.	

thiourea ligand is coordinated to the metal centers through its oxygen atom of carbonyl group⁴⁵⁻⁴⁷. The infrared spectra of the complexes **1-10**, show no bands expected to $\nu(\text{M-N})$ which means that the thiourea ligand is not coordinated to the metal centers through the nitrogen atom of its secondary amine.

The infrared bands in metal complexes appeared very weak to very strong intensity bands in the range 359-413 cm⁻¹ (Table-2), which are assigned to $\nu(\text{M-S})$ ^{33,40,42, 48,49}. This indicates the coordination of the thiourea ligand to the metal centers is through the sulfur atom of the thiocarbonyl group. The infrared spectrum of the complex **9**, shows a strong band at 310 cm⁻¹ which is attributed to the $\nu(\text{Hg-Cl})$ band^{12,36}.

Electronic spectra: The electronic spectra of the prepared thiourea metal complexes are recorded in DMSO at 1 × 10⁻³ M solution at room temperature in ultraviolet and visible region. The spectral bands are tabulated in Table-3.

The electronic spectrum of the manganese(II) complex **1** (Table-3), shows a band in ultraviolet region at 30769 cm⁻¹ which attributed to Mn-ligand charge transfer transition band⁴⁰. The Mn(II) complex **1**, has no electronic absorption bands in the visible region which may denote to that this complex is tetrahedral because tetrahedral manganese(II) complexes are not expected to exhibit *d-d* transition in the visible region since the *d-d* transitions are spin and Laporte forbidden^{50,51}.

The electronic spectrum of Co(II) complex **2**, exhibit three bands; 40650, 34834 and 15383 cm⁻¹ (Table-3). The bands at 40650 and 34834 cm⁻¹ are attributed to electronic charge transfer transitions⁴⁰. The electronic transition at 15383 cm⁻¹ is assigned to ⁴A₂(F) → ⁴T₁(P) of Co(II) in a tetrahedral arrangement^{1,40-42,50-52}.

The electronic spectrum of Ni(II) complex **3** (Table-3) shows several bands at 40650, 33557, 27397, 16474 and 14577

cm⁻¹. The bands at 40650 and 33557 cm⁻¹ are assigned to charge transfer transitions. The band at 27397 cm⁻¹ and a multiple absorptions at 16474 and 14577 cm⁻¹ are attributed to A_{2g} → ³T_{1g} and ³T₁(F) → ³T₁(P) electronic transitions of Ni(II) in a tetrahedral geometry, respectively^{41,50}.

The electronic spectrum of the Cu(II) complex **4** (Table-3) shows four bands at 40000, 33113, 24096 and 23256 cm⁻¹. The bands at 40000 and 33113 cm⁻¹ are assigned to charge transfer bands, while the band at 24096 cm⁻¹ is attributed to internal ligand transitions⁴⁰. The band at 23256 cm⁻¹ is attributed to ²B_{1g} → ²E_g and ²B_{1g} → ²A_{1g} transitions of Cu(II) square planar geometry⁵³.

The electronic spectra of Zn(II), Cd(II), Hg(II) and Pb(II) complexes (Table-3) show electronic bands in the range 30675-42194 cm⁻¹ which are either assigned to charge transfer transitions or internal ligand transitions^{54,55}.

The electronic spectrum of Pd(II) complex **6**, (Table-3) shows two bands at 40000 and 33898 cm⁻¹. The band at 40000 cm⁻¹ is assigned to charge transfer transition, while the band at 33898 cm⁻¹ is attributed to ¹A_{1g} → ¹B_{2g} and ¹A_{1g} → ¹B_{1g} transitions of Pd(II) in square planar geometry^{40,55}.

The electronic spectrum of Pt(II) complex **8**, (Table-3) shows two bands at 39024 and 33784 cm⁻¹. The band at 39024 cm⁻¹ is assigned to charge transfer transition, while the band at 33784 cm⁻¹ is attributed to a ¹A_{1g} → ¹B_{2g} and ¹A_{1g} → ¹B_{1g} transition of Pt(II) of square planar geometry^{40,55}.

Magnetic susceptibility: The magnetic susceptibility measurements of the prepared complexes are made at room temperature and the data are arranged in Table-3.

The magnetic susceptibility value of the Mn(II) complex **1**, is 5.78 BM (Table-3), which suggests a tetrahedral arrangement

around Mn(II) metal as compared with the reported values for tetrahedral Mn(II) complexes⁵⁶. The value of magnetic susceptibility of Co(II) complex **2**, is 4.65 BM (Table-3), which confirms a tetrahedral arrangement around Co(II) metal ion^{1,48}. The magnetic susceptibility of the Ni(II) complex **3**, is 3.55 BM (Table-3), which denotes a tetrahedral arrangement around Ni(II) metal center as compared with the reported values for tetrahedral Ni(II) complexes 3.5- 4.1 B.M.^{32,42,51}. The magnetic susceptibility of the prepared Cu(II) complex **4**, is 1.77 BM, (Table-3), which assigned to one unpaired electron of 9d-system of square planar Cu(II)^{2,84}. The magnetic susceptibility values of Pd(II) and Pt(II) complexes **6** and **8** (Table-3), are zero which indicate that these complexes are diamagnetic and have square planar arrangements^{20,56}.

The Zn(II), Cd(II), Hg(II) and Pb(II) complexes **5**, **7**, **9** and **10**, have zero magnetic susceptibility Table-3, which means that they are diamagnetic and expected that they do not show *d-d* transitions and consistent with the tetrahedral geometry around these metals^{48,49,57}.

No.	Compound	<i>E. coli</i> Gram(-ve)	<i>P. aeruginosa</i> Gram(-ve)	<i>S. aureus</i> Gram(+ve)
1	[Mn(B) ₂]	R	R	R
2	[Co(B) ₂]	R	S	R
3	[Ni(B) ₂]	R	R	R
4	[Cu(B) ₂]	R	R	S
5	[Zn(B) ₂]	R	R	R
6	[Pd(BH) ₂]Cl ₂	R	S	R
7	[Cd(B) ₂]	R	R	S*
8	[Pt(B) ₂]	R	R	S
9	[Hg(BH)Cl ₂]	S	S	S*
10	[Pb(B) ₂]	R	R	R
	BH	0	0	0
	DMSO (Control)	0	0	0

where: R = resistant, S = sensitivity,
S* = maximum sensitivity (maximum inhibition zone)

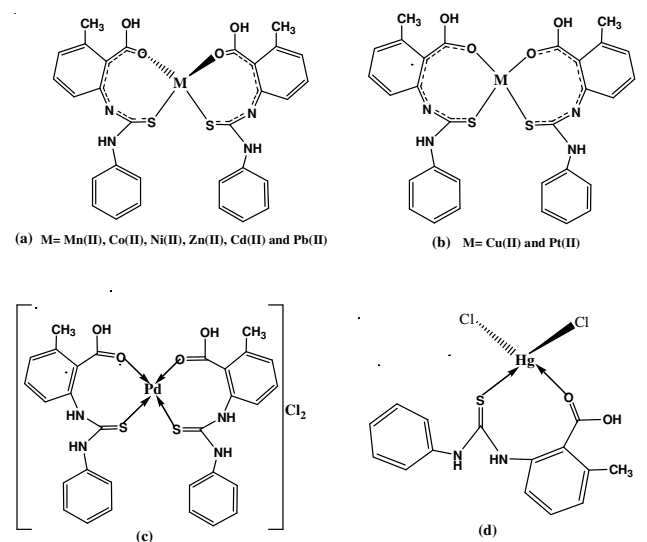


Fig. 1. Suggested structures of the prepared complexes; (a) tetrahedral [M(B)₂], (b) Square planar [M(B)₂], (c) Square planar [Pd(BH)₂]Cl₂ and (d) Tetrahedral [Hg(BH)Cl₂]

Biological activity: The biological activity of the ligand and its complexes were tested against gram positive and gram negative bacteria as *E. coli* (gram negative), *P. aeruginosa* (gram negative) and *S. aureus* (gram positive) using well-diffusion method³⁰. The biological activities were estimated on the basis of the size of inhibition zones formed around the wells of the agar discs in mm (Table-4).

On the basis of micro-elemental analysis, infrared and UV-Visible spectroscopic, conductivity and magnetic susceptibility data, the following structures are proposed for the prepared complexes as shown in Fig. 1.

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