Study on Synthesis and Antibacterial Activity of Co(II) and Ni (II) Complexes Including Isopropylacetone Thiosemicarbozone and Cresol

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Abstract

Complexes containing mixed ligands of cobalt (II) and nickel (II) have been synthesized by the reaction of cobalt chloride or nickel chloride with isopropylacetonethiosemicarbazone (LH) and substituted cresols (3–Methyl–2 nitrophenol (L_1H) or 3–Methyl–2–aminophenol (L_2H) or 2–Methyl–3–nitrophenol (L_3H) or 3–Methyl–4–nitrophenol (L_4H) in mole ratio 1:2:1 metal: isopropylacetonethiosemicarbazone: cresol, respectively and characterized phyico–chemically.

They have been found to have the general formulas $[M(LH)_2(L^-H)]CL_2$ or$[M(LH]_2(L^-nH)Cl]Cl$ and $K[M(L)_2L^-]$ or $M(L)_2(L^-n)(H_2O)]$ in neutral and basic medium, respectively where M = Co(II) or Ni(II); LH = isopropylacetonethiosemicarbazone; L^-H = L_1H or L_2H; L_n^-H= L_3H or L_4H; L^-= deprotonated L_1H or L_2H; L_n^-= deprotonated L_3H or L_4H, respectively.

Furthermore, the complexes have been found to be biologically active as demonstrated by antibacterial screening against seven human pathogenic bacterial strains. It has been found that the complexes were more effective agent than the ligand.

Keywords: isopropylacetonethiosemicarbazone, antibacterial agent, cresol complexes, cobalt complexes, nickel complexes.

الخلاصة

حضرت معقدات للكوبلـت (II) والنيكـل (II) مـع مـزيج مـن ليكانـد آيزوبروبيـل اسـيتون ثايوسيميكاربازون (LH) وبعض الكريسولات المعوضة (3-مثيل-2-نتروفينـول (L_1H) أو 3-مثيـل-2-امينوفينول (L_2H) او 2-مثيل-3-نتروفينول (L_3H) أو 3-مثيل-4-نتروفينول (L_4H) بنـسب مولاليـة : 1:2:1 فلتر : ليكاند : كريسول معوض . شخصت المعقدات المحضرة بطرق فيزيائية – كيميائية . ووجد ان لها الصيغ البنائيـة العامـة $[M(LH)_2(L^-H)]CL_2$ أو $[M(LH]_2(L_n^-H)Cl]Cl$ أو $K[M(L)_2L^-]$ أو $K[M(L)_2(L_n^-)(H_2O)]$ على التوالي في كل من الوسطين المتعادل والقاعدي حيث أن LH = آيزوبروبيل اسيتون ثايوسيميكاربازون ، L^-H = L_1H أو L_2H ؛ L_n^-H = L_3H أو L_4H ؛ $L_1=L^-$ أو L_2 (بـدون بروتون) ؛ $L_3=L_n^-$ أو L_4 (بدون بروتون) ؛ M= Co(II) أو Ni(II) على التوالي . كما شخصت الفعالية البكتيرية للمعقدات على سبعة جراثيم مرضية كمضادات للبكتيريا الموجبة والسالبة لصبغة كرام والمعزولـة من حالات مرضية وقد أثبتت المعقدات أن لها فعالية كمضادات للبكتيريا أكثر من الليكاند .

Introduction

Thiosemicarbazone complexes of transition metals have attracted special attention due to their activity against virus, protozoa and certain kinds of tumor and also antitubercular activity[1-3]. Recent years witness a growing interest in the chemistry of thiosemicarbazone owing to their ligational properties achieved through several available coordination sites[4-7].

In view of this, since mixed ligand complexes with isopropylacetone thiosemicarbazones , substituted cresols with Co(II) and Ni (II) have not yet been reported. It is matter of interest to determine the extent to which the biological properties of these ligands would be affected by incorporating metal (II) ion[8-10].

In the present work, cobalt (II) and nickel (II) complexes with mixed ligand (Figure 1) have been prepared and characterized physico -chemically besides, the study of antibacterial activity of these complexes against seven human pathogenic bacterial strains.

Experimental

a. Chemicals:

All chemicals used throughout these investigation avialble from Merck, B.D.H., Aldrich or Fluka.

B. Synthetic Methods:

Isopropylacetonethiosemicarba zone has been prepared according to literature method[11].

A general procedure has been adopted for the preparation of complexes in neutral and basic medium. In neutral medium, a solution of 0.01 mole (2.38g or 2.36g) of cobalt chloride or nickel chloride in 10ml of distilled water has been added to the solution of isopropylacetonethiosemicarbazone (0.02 mole (3.46g) and one of the substituted cresols (L_1H or L_2H or L_3H or L_4H 0.01 mole (1.53g, 1.53g, 1.53g and 1.53g) in 15 ml of hot ethanol, respectively. The mixtures have been refluxed for 3hrs. followed by evaporation of half then cooled. The products are isolated by filtration, washed with petroleum ether and dried. In basic medium, complexes have been prepared by applying the same amount used in neutral medium, and after mixing the metal chlorides with the ligands and heating on a water bath, potassium hydroxide solution (1M) has been added until pH of the solution have been (8–9). The mixtures have been heated on a water bath for half hr. Then allowed to cooled. The products have been filtered off, washed with petroleum ether and dried over $CaCl_2$.

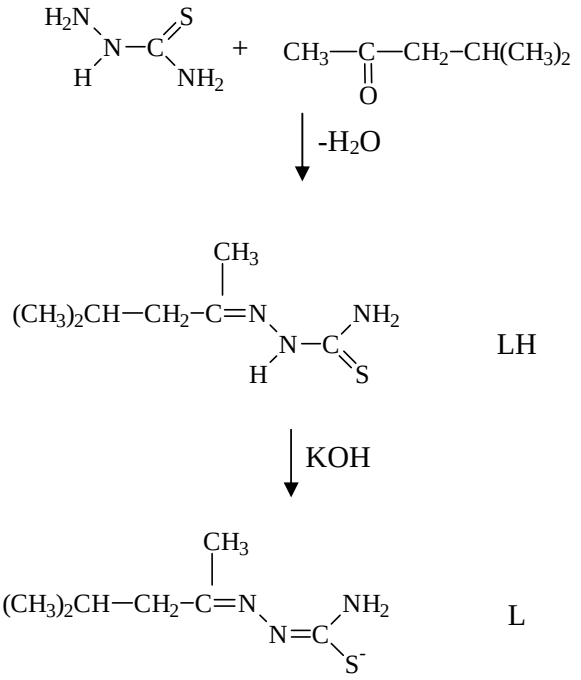
C. Analytical and Physical Measurements:

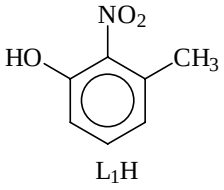
Metal and chloride contents have been determined by applying precipitation methods[12] decomposition of the complexes with concentrated nitric acid. Nitrogen content has been determined by Kjlediahl method. Conductivity measurements of the complexes have been measured using 10^{-3}M dimethylformamide solution at 25°C by using LF–42 conductivity meter. The infrared spectra has been recorded on Pye–Unicam Sp–1100 spectrometer as KBr discs at 4000 – 400 cm^{-1}. Electronic spectra have been recorded on a Shimadzu UV–160 spectrophotometer for 10 M^{-3} solution of the compounds on DMF at room temperature using at 1cm quartz cell. Magnetic susceptibility measurements have been determined by Bruker B.M6 of the complexes. Molecular weights of the complexes have been determined cryoscopically[13].

D. Antimicrobial Assay of the Complexes:

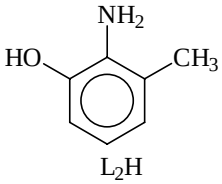
Seven pathogenic microorganisms have been selected to study the antibacterial activity of the complexes in this work. The gram

positive bacteria were *staphylococcus aureus*, *streptococcus pyogenes*, *Bacillus subtillis* and gram negative *Eschcterichia coli*, *Pseudomonas aeruginosa*, *Proteus Vulgaris* and *Salmonilla typhi*. In this method nutrient agar Petri dishes 6cm diameter have been seeded with 0.1ml of the broth culture of the tested microorganism containing $(10)^8$ cell/(ml). Filter paper discs of 6mm diameter have been impregnated with the tested materials have been placed on the surface of seeded nutrient agar dishes, then these dishes have been incubated at 37°C for 24hrs. The zone of inhibition have Been[14,15] measured using a special calibrated lences.

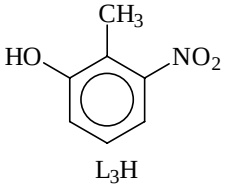




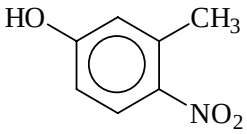
3–Methyl–2–nitrophenol
2–Nitro–m–cresol



2–Amino–3–methylphenol
2–Amino–m–cresol



2–Methyl–3–nitrophenol
3–Nitro–o–cresol



L4H

3–Methyl–4–nitrophenol
4–Nitro–m–cresol

Figure 1: Model structures of the ligands

E. Determination of Minimum Inhibitory Concentration (MIC):

Different concentrations of the tested compounds in dimethylsulphoxide solutions (7.600, 15.375, 31.250, 62.500, 125 and 500μg/ml) have been used for the determination of minimum inhibitory concentration (MIC)[16]. The lowest dilution which inhibits the growth have been recorded, each experiment has been carried out in triplicates for each concentration of the complexes as well as for the microorganisms alone as positive controls for the growth The antibiotic chloramphenicol and ciprofloxacin have been used as acontrol .

Results and Discussion

The reaction of $CoCl_2.6H_2O$ or $NiCl_2.6H_2O$ with isopropylacetone thiosemicarbazone and substituted cresols in 1:2:1 molar ratio in both neutral and basic medium can be represented by the following reactions:

$$MCl_2.6H_2O + 2LH + L^{-}H \rightarrow [M(LH)_2(L^{-}H)]Cl_2 + 6H_2O \quad \dots (1)$$

$$MCl_2.6H_2O + 2LH + L^{-}{}_nH \rightarrow [M(LH)_2(L^{-}{}_nH)Cl]Cl + 6H_2O \quad \dots (2)$$

$$MCl_2.6H_2O + 2LH + L^{-}H + 3KOH \rightarrow K[M(L)_2(L)] + 2KCL + 9H_2O \quad \dots (3)$$

$$MCl_2.6H_2O + 2LH + L_nH + 3KOH \rightarrow K[M(L)_2(L_n)(H_2O)] + 2KCL + 8H_2O \quad \dots (4)$$

where M = Co(II) or Ni(II), LH = isopropylacetone thiosemicarbazone

$L^{-}H = L_1H$ or L_2H; $L^{-}{}_nH = L_3H$ or L_4H; L^{-} and $L^{-}{}_n$ deprotonated L_1H or L_2H and L_3H or L_4H respectively.

The solid complexes are coloured, slightly soluble in water, moderately soluble in ethanol and soluble in dimethylformamide. The values of molar conductivities (Table1) approach those expected for 1:2 and 1:1 for complexes prepared in neutral and 1:1 for complexes prepared n basic medium[17]. Molecular weight measurements suggest that all the complexes are monomers. The room temperature magnetic moments of Co(II) complexes are in the range 3.75–4.25B.M (Table 1) suggesting the presence of unpaired electrons, and hence the complexes are spin free with octahedral stereochemistry[18]. The magnetic moments of Ni(II) complexes calculated also from the corrected magnetic susceptibilities are in the range 2.75–2.79B.M as shown in (Table 1), indicating octahedral geometry around the nickel ion[19].

The coordination sites of the ligands have been indicated by infrared spectroscopy, which is considered a useful technique for probing the structure of complexes. The infrared data of the main absorption bands of the ligands and the complexes are given in (Table 2).

A strong band at about 1560cm^{-1} which has been attributed to stretching frequency of azomethine groups (C=N) shifted towards a lower frequency of the order 10–30 cm^{-1} on coordination due to the decrease of the bond order as a result of metal nitrogen bond formation[20]. The next band at 1420cm^{-1} and 1340 cm^{-1} have been attributed to the stretching and bending frequencies of thiosulfur group (C=S). The first value shifted towards a lower frequency on coordination in neutral medium, indicating the formation of chelation between the sulfur at the C=S group and the metal ion[21]. Meanwhile, in basic medium, this band has been absent and a new band has been observed at 700–750cm^{-1} due to the stretching vibration of C–S group (because of the conversion of the ligand to the thiolic form and the deprotoration of the ligand has been took place in basic medium. The region of 3200–3300cm^{-1} of the ligand

spectra has been due to a broad band of stretching frequency of NH group as a result of consequence of hydrogen phenomenon has been more complicated due to different factor[22]. Such effect of hydrogen bonding and the effect of coordination and also the presence of other groups (NH_2, OH and H_2O) appeared in the same position. This band, however, has been remained unaltered in the complexes prepared in neutral medium, indicating that there is no coordination through the NH group. Meanwhile in basic medium, because of the presence of hydrogen bonding it has been more difficult to notice the absence of NH group, but it has been well known that this band has been disappeared in basic medium[22] due to the thiolic form. The other strong bands at 3130–3420 and 1480cm^{-1} have been due to υ_{NH2}[23]. These bands remained unaltered on complexation indicated that there is no coordination through this group and metal ion.

The infrared spectra of substituted cresols ligands (L_1H, L_2H, L_3H and L_4H) showed a wide bands in the region 3300–3600cm^{-1} and 3420cm^{-1} due to the stretching vibration of phenolic OH and NH_2 in L_2H, respectively. The wide range has been due to the hydrogen bonding. In the spectra of the complexes it has been more difficult to observe the coordination due to the presence of different groups and hydrogen bonding. Whatever, in the complexes have been prepared in neutral medium this wide band has been shifted to higher frequency, whereas for complexes have been prepared in basic medium it has been very difficult to observe. The disappearance of this band, but it has been well known that this band has been disappeared due to the deprotoration of cresol and the formation of ionic form[24]. As a matter of the fact the band at about 1230cm^{-1} which is attributed to C–O group shifts towards higher frequency on complexation in all complexes demonstrates an evidence of coordinating through phenolic oxygen to the metal ion[25]. The spectra of L_2H shows a band at 3300–3460cm^{-1}, this band has been shifted towards a lower frequency indicating the formation of chelation between the nitrogen of NH_2 group and the metal ion[25]. The ligand L_1H, L_3H and L_4H show a band at 1525 and 1355cm^{-1} due to the nitro group[25]**.** This band is shifted towards lower frequencies on coordination in the spectra of L_1H complexes indicating the formation of chelation between the oxygen of nitro group and the metal ion. While this band has been remained unaltered in the spectra of L_3H and L_4H complexes indicating the absence of coordination through this group. In addition for chloride complexes show a band at 590-610cm^{-1} which is observed in their infrared spectra has been attributed to ionic chloride[25]. This is not observed in the spectra of the complexes prepared in basic medium indicating the absence of this ion. For all complexes spectra new bands have been observed around 530–610cm^{-1}, 420–520cm^{-1}, and 620–680cm^{-1} which attributed to the stretching modes of M–S, M–N and M–O, respectively[26]. The presence of these bands support the formation of the complexes under investigation figures.

The aqua complexes gave one broad band in the region 3260–3320cm^{-1} due to stretching vibration OH of water[27]. The loss of water molecules at a relatively high temperature suggest that these are coordinated and not lattice held[28].

The coordination of Chloride could not be inferred form the infrared spectra of the complexes because the band due to these group occurred

beyond the range of our infrared spectrophotometer.

The electronic spectral bands of complexes in DMF solution have been recorded giving d–d spectra and charge transfer spectra. The spectra of cobalt(II) complexes exhibit three bands due to the spin–allowed transitions at 9495–9555cm^{-1}, 15800–16404cm^{-1} and 18350–19000^{-1}cm due to the transitions $^4T_{1g(F)} \rightarrow {}^4T_{2g(F)}(\upsilon_1)$, $^4T_{1g(F)} \rightarrow {}^4A_{2g(F)}(\upsilon_2)$ and $^4T_{1g(F)} \rightarrow {}^4T_{1g(p)}(\upsilon_3)$ respectively expected for d^7 system in octahedral field. The three bands in the spectra of nickel(II) complexes at 10084–10541cm^{-1}, 14365 – 15750 cm^{-1} and 19127–20500cm^{-1} due to the transitions $^3A_{2g(F)} \rightarrow {}^3T_{2g(F)}(\upsilon_1)$, $^3A_{2g(F)} \rightarrow {}^3T_{1g(F)}(\upsilon_2)$ and $^3A_{2g(F)} \rightarrow {}^3T_{1g(p)}(\upsilon_3)$, respectively. The number and positions of these bands corresponding to octahedral structure for both cobalt and nickel complexes.

The ligand field parameter B and the ligand field splitting energy ($10D_q$) have been calculated[29]. The free ion value of B have been 372–440 and 221–314cm^{-1} compared to the reported value of B (1120 and 1080cm^{-1}) for Co(II) and Ni(II) complexes, respectively. The $10D_q$ values of the complexes indicated that the ligands have been produced a weak field energy calculations pointed out to the high spin nature of the complexes. The values of β of the complexes are between 0.21–0.45cm^{-1} clearly indicated the covalent character of the bond concerned. However, the electronic spectral data of complexes (Table 3) showed that the values of D_q /(B, υ_3/υ_1 and υ_3/υ_1) confirmed the octahedral geometry for all the complexes[29].

The antibacterial agents were known to attack the bacterial cell in a variety of ways such as killing or inhibiting the growth of bacteria by affecting special target sites like the synthesis of cell wall, protein and nucleic acid through their effects on the synthesis of ribonucleic acid which could be resulted from the inhibition action of bacterial DNA enzyme which caused the separation of supercoiling or decantenation or unknotting of the DNA[30]. Numerous experiments have been done to determine the antibacterial influence of the complexes (Table 4).

The chemical complexes had a good ability to attack the cell wall of the bacteria through the heavy metal ions which preferentially bind to SH (sulfhydryl group) of the cell enzyme more strongly, it is logical to assume that the complexes screened were involved in a competitive equilibria involving the SH group of the cell enzyme. Therefore, we concluded the most of the complexes acquire a good antibacterial activity, this may be due to interaction of the metal ion more strongly than the donor atoms in the ligands and should have lower MIC these observations have been consistent with that observed by many authors[30, 31] as shown in (Table 4 and5) and (Figure 4).

Table 1: Analytical data and physical properties of the complexes

No.	Complexes	m.p or d	Color	Λ_M	Relative M.Wt. calc. (obser.)	$\mu_{eff.}$	% Analysis Calc. (Observ.)		
							M	N	Cl
1.	$[Co(LH)_2(L_1H)]Cl_2$	119	Brown	158	629 (637)	3.75	9.37 (91.7)	15.58 (15.63)	11.28 (11.52)
2.	$[Ni(LH)_2(L_1H]Cl_2$	135(d)	Green	137	628.7 (640)	2.75	9.33 (9.24)	15.58 (15.71)	11.29 (11.98)
3.	$[Co(LH)_2(L_2H]Cl_2$	111	Brown	162	599 (611)	3.85	9.84 (9.68)	16.63 (16.20)	11.85 (12.00)
4.	$[Ni(LH)_2(L_2H)]Cl_2$	104	Green	130	598.7 (600)	2.75	9.80 (10.00)	16.36 (16.47)	11.85 (11.98)
5.	$[Co(LH)_2(L_3H)Cl]Cl$	115	Brown	80	629 (624)	4.25	9.37 (9.31)	15.58 (15.36)	5.64 (5.37)
6.	$[Ni(LH)_2(L_3H)Cl]Cl$	137	Green	65	6287 (653)	2.76	9.33 (8.91)	15.58 (15.21)	5.64 (5.70)
7.	$[Co(LH)_2(L_4H)Cl]Cl$	124	Brown	73	629 (647)	3.95	9.33 (8.91)	15.58 (15.36)	5.64 (5.70)
8.	$[Ni(LH)_2(L_4H)Cl]Cl$	113	Green	67	628.7 (650)	2.76	9.33 (8.59)	15.58 (15.30)	5.64 (5.82)
9.	$K[Co(L)_2 (L_1)]$	125(d)	Brown	65	549 (562)	4.10	9.93 (10.01)	16.49 (16.32)	-
10.	$K[Ni(L)_2(L_1)]$	121	Green	82	593.7 (610)	2.79	9.88 (10.00)	16.50 (16.79)	-
11.	$K[Co(L)_2(L_2)]$	107	Brown	79	564 (578)	4.00	10.46 (10.21)	17.37 (17.48)	-
12.	$K[Ni(L)_2(L_2)]$	131	Green	65	5637 (574)	2.75	10.41 (10.17)	17.38 (17.17)	-
13.	$K[Co(L)_2(L_3)(H_2O)]$	117	Brown	87	612 (634)	3.99	9.46 (9.87)	16.01 (16.23)	-
14.	$K[Ni(L)_2(L_3)(H_2O)]$	161	Green	69	611.7 (588.5)	2.79	9.59 (9.82)	16.02 (15.82)	-
15.	$K[Co(L)_2(L_4)(H_2O)]$	110	Brown	77	612 (591)	3.75	9.46 (9.47)	16.01 (16.20)	-
16.	$K[Ni(L)_2(L_4)(H_2O)]$	139	Green	85	611.7 (637)	2.78	9.59 (9.43)	16.02 (15.98)	-

d= decomposition point

Λ_M= molar conductivities in Ω^{-1} cm^2 mol^{-1}

Table 2: Selected I.R. bands of the ligands and their complexes (in cm^{-1})

Comp.	υ_{NH} , $\upsilon_{OH\ (ph.)}$	$\upsilon_{C=N\ thiosemi}$	$\upsilon_{C=S\ thiosemi}$	$\delta_{C=S}$	$\upsilon_{NH2\ thiosemi}$	υ_{M-S}	υ_{M-N}	υ_{M-O}	υ_{C-O}
LH	3200– 3300	1560	1420	1340	3130–3420,1480	-	-	-	-
1	3200– 3300 3500– 3600	1530	1400	1220	3130–3420,1480	540	450	620	1230
2	3200– 3300 3500– 3600	1540	1400	1230	3130–3420,1480	550	450	660	1230
3	3200– 3300 3500– 3600	1550	1400	1220	3130–3420,1480	600	460	640	1230
4	3200– 3300 3500– 3600	1545	1400	1230	3130–3420,1480	600	450	650	1230
5	3200– 3300 3500– 3600	1550	1400	1220	3130–3420,1480	590	475	660	1230
6	3200– 3300 3500– 3600	1545	1400	1220	3130–3420,1480	610	460	680	1230
7	3200– 3300 3500– 3600	1540	1410	1230	3130–3420,1480	600	500	670	1230
8	3200– 3300 3500– 3600	1550	1410	1220	3130–3420,1480	570	490	620	1230
9	3200– 3300	1540	-	-	3130–3420,1480	610	420	680	1230
10	3200– 3300	1540	-	-	3130–3420,1480	580	490	650	1230
11	3200– 3300	1540	-	-	3130–3420,1480	560	495	650	1230
12	3200– 3300	1545	-	-	3130–3420,1480	575	500	670	1230
13	3200– 3300	1550	-	-	3130–3420,1480	590	485	660	1230
14	3200– 3300	1540	-	-	3130–3420,1480	600	500	675	1230
15	3200– 3300	1530	-	-	3130–3420,1480	600	470	680	1230
16	3200– 3300	1540	-	-	3130–3420,1480	550	440	660	1230

Table 3: Electronic spectral data of Co(II) and Ni(II) complexes

No	υ_1	υ_2	υ_3	C.T	B	$10D_q$	D_q/B	β	υ_3/υ_2	υ_3/υ_1	C.F.S.E.
1.	9555	15800	18865	23750	400	9130	2.32	0.41	1.19	1.97	7448.0
2.	10529	14593	20500	28800	233	10524	4.51	0.22	1.40	1.94	12634.8
3.	9535	16403	18816	25770	440	9281	2.10	0.45	1.14	1.97	7424.8
4.	10537	15058	19875	28615	221	10537	4.76	0.21	1.13	1.88	12644.4
5.	9501	16161	18834	28125	432	4333	2.16	0.44	1.13	1.98	7466.4
6.	10160	14365	20321	28200	280	10160	3.62	0.27	1.41	2.00	12192.0
7.	9521	16111	19000	29560	436	4479	2.17	0.44	1.17	1.99	7583.2
8	10118	14694	19923	28900	284	10118	3.56	0.27	1.35	1.96	12141.6
9	9495	15922	18911	29100	427	9416	2.20	0.43	1.18	1.99	7532.8
10.	10089	15750	19570	28700	270	10089	3.73	0.26	1.24	1.97	7424.8
11.	9517	15873	18798	28910	408	9281	2.27	0.42	1.24	1.97	7424.8
12.	10204	14897	19127	28500	227	10204	4.49	0.21	1.28	1.87	12244.8
13.	9498	15899	18518	29400	394	9020	2.28	0.40	1.16	1.94	7216.0
14.	10105	15623	19419	28500	314	10975	3.49	0.30	1.24	1.92	13170.0
15.	9523	16007	18350	28000	372	8827	2.37	0.38	1.14	1.92	7061.16
16.	10541	14999	19809	28500	269	10541	3.91	0.26	1.32	1.87	12649.2

Table 4: The antibacterial activities of some complexes against different bacteria

No. LH	*S. aureus* R	*S. pyogenes* MS	*B. subtillis* R	*E. coli* MS	*Ps. aruginosa* R	*Pr. Vulgaris* S	*S. typhi* MS
3	S	R	S	R	S	MS	MS
5	S	S	MS	S	S	S	S
7	MS	S	S	MS	MS	S	R
9	R	MS	R	S	S	MS	S
10	MS	R	S	MS	MS	R	MS
12	S	MS	S	R	R	S	S
14	R	S	MS	S	S	MS	S
16	S	S	R	MS	S	MS	S

S= sensitive, zone diameter nor more than 6mm less than control
MS= moderately sensitive, zone diameter of 6–12mm less than control
R= resistant, zone diameter of 12mm or less than control.

Table 5: Minimum inhibitory concentrations (μg/ml) of the complexes

No.	*S. aureus*	*S. pyogenes*	*B. subtillis*	*E. coli*	*Ps. aruginosa*	*Pr. Vulgaris*	*S. typhi*
3	62.50	–	62.50	–	62.50	500	6250
5	31.25	62.50	500	62.50	62.50	62.50	500
7	500	62.50	62.50	62.50	500	62.50	–
9	–	500	–	62.50	62.50	500	31.25
10	500	–	62.50	500	500	–	500
12	62.50	500	31.25	–	–	62.50	62.50
14	–	62.50	500	–	31.25	500	62.50
16	31.25	62.50	–	500	62.50	500	62.50

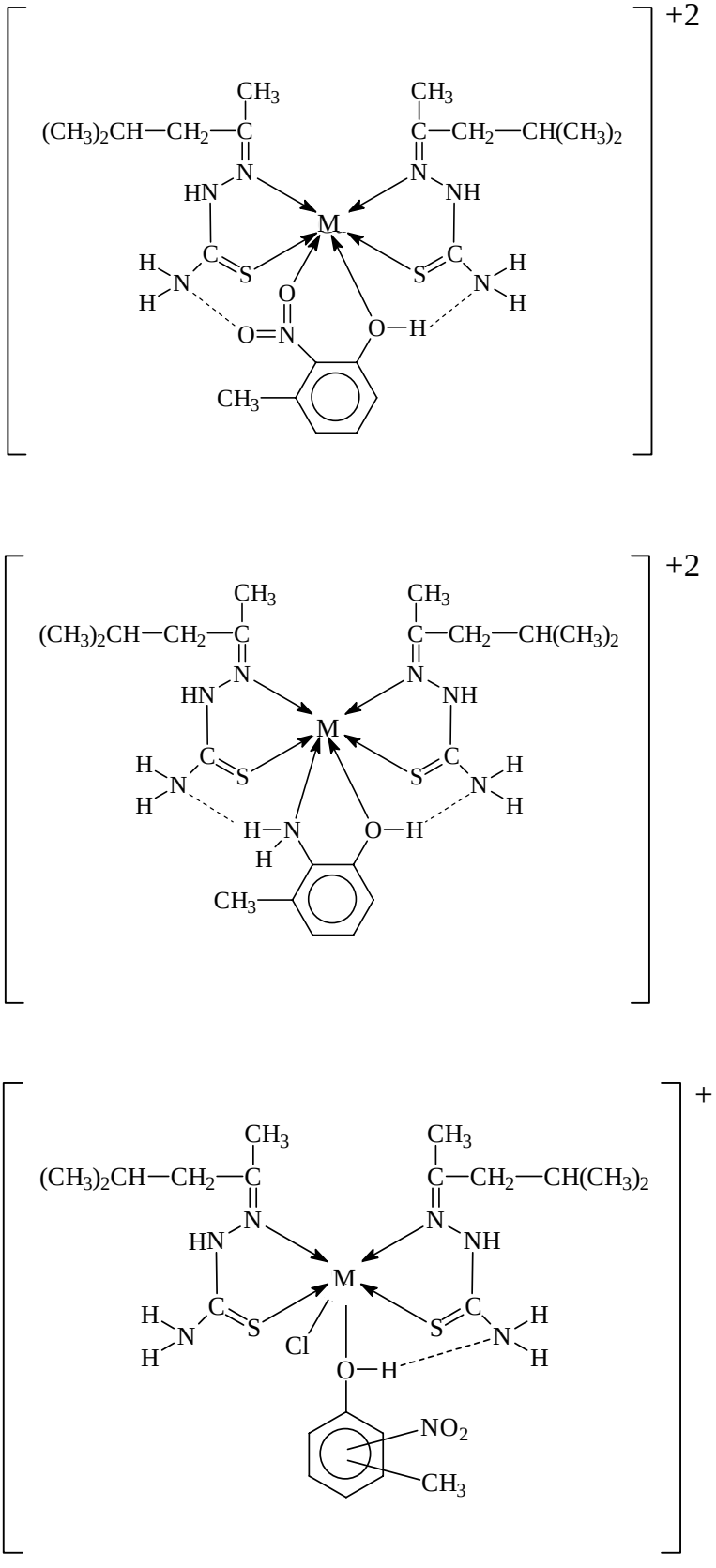


Figure (2) Structures of the complexes in neutral medium

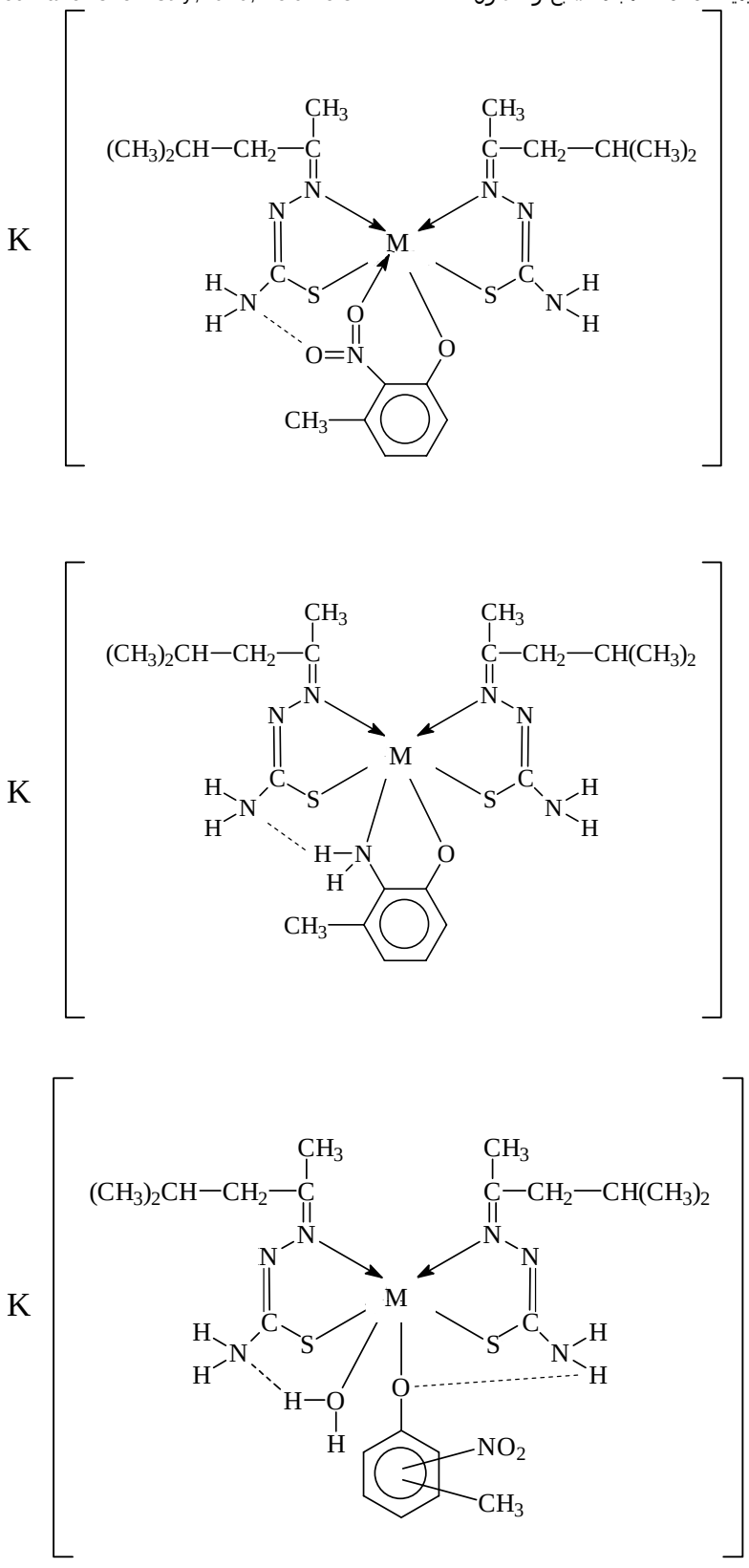


Figure (3) Structures of the complexes in basic medium

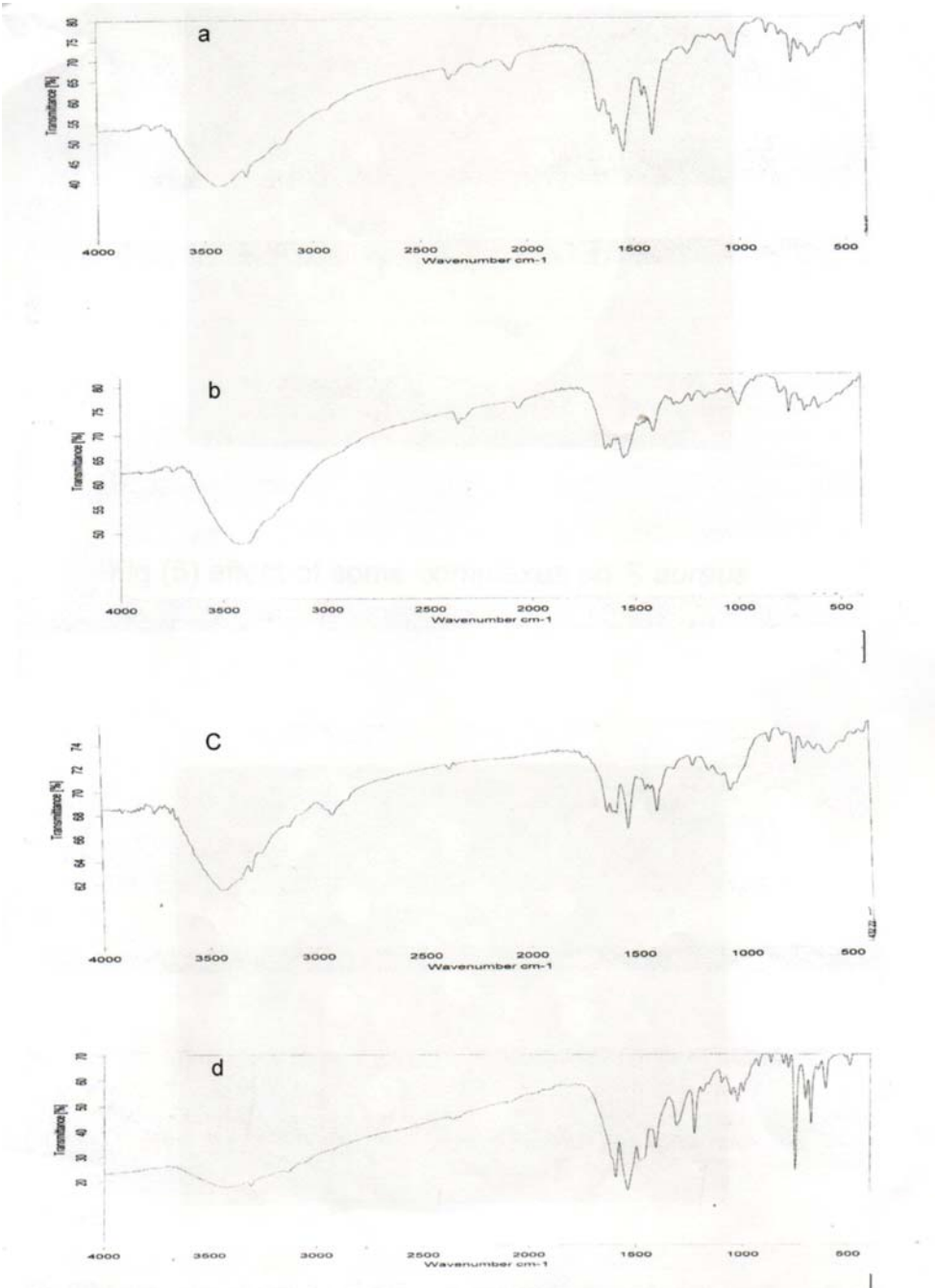


Fig (4):infrared spectra of (a) L_2H ligand,(b)LH ligand,© complex 2 (d) complex 9

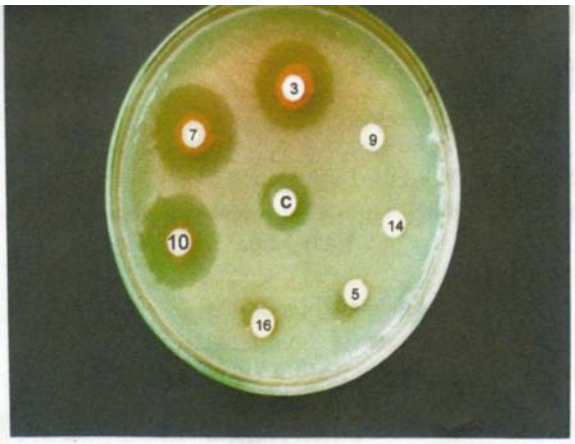
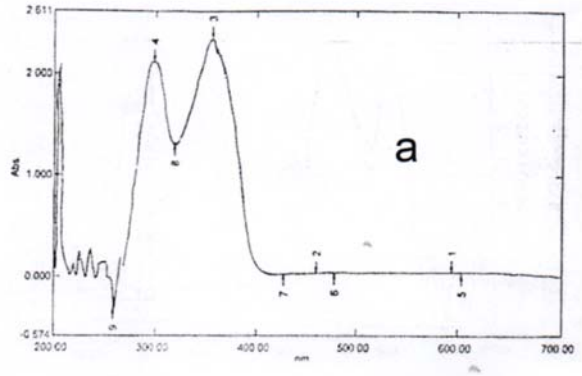
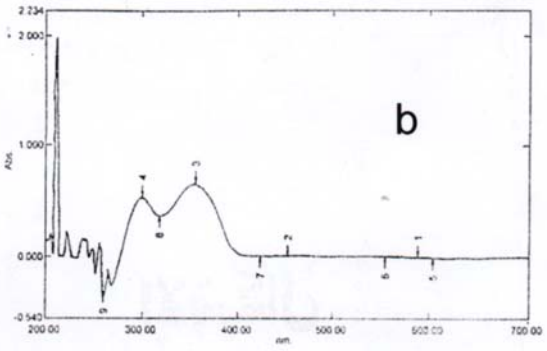


Fig (5) effect of some complexes on *S.aureus*

Fig (6) Antibacterial activity of different concentration of complex 5 on *E.coli*





Fig(7): electronic spectra of (a) complex(1) & (b) complex (3)

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