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Synthesis, Spectral and Antibacterial studies of some transition metal complexes with Schiff base derived from thiosemicarbazide and Phthalaldyde

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ABSTRACT Three transition metal Cr(III), Ni(II) and Cu(II) complexes of the Schiff base N,N′-((*1Z,1′E*)1,2-phenylenebis(methanelylidene)-bis(hydrazinecarbothioamide) have been synthesised and investigated using several techniques: physical characteristics, UV-Vis, mass spectra, molar conductivity, magnetic moment measurements, micro-analytical data (CHNS), ^{1}HNMR and IR. The micro-analytical data indicate the formation of a 1:1[M:L] ratio. The molar conductance measurement showed that the $[NiL(Cl)_2]$ complex has non-electrolytic nature. The molar conductance measurements of the complexes $[Cr(H_2L)(Cl)_2$ and $[Cu(H_2L)(H_2O)_2]Cl_2$ were electrolyte in nature and $[Ni(H_2O)Cl_2]$ was non-electrolyte in nature. The infrared spectral data displayed the main coordination sites of N,N′-((*1Z,1′E*)1,2-phenylenebis (methane lylidene)-bis(hydrazinecarbothioamide) towards Cr(III), Ni(II) and Cu(II) ions. The electronic spectral results exhibited the presence of the electronic transitions in the free Schiff base and its complexes, the data suggested that the Cr(III), Ni(II) and Cu(II) complexes have an octahedral geometrical structure.
Keywords: Schiff base; metal complexes; Spectroscopic study and Biological activity.

تحضير ودراسة طيفية ودراسة ضد البكتيريا لبعض متراكبات العناصر الانتقالية مع قاعدة شيف المشتقة من Thiosemicarbazide and Phthalaldyde

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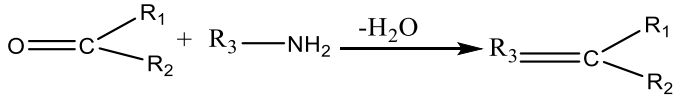
الملخص ثلاثة متراكبات لعناصر الكروم الثلاثي والنيكل الثنائي والنحاس الثنائي مع قاعدة شيف تم تحضيرها وتشخيصها بالستخدام عدة تقنيات منها دراسة الخواص الفيزيائية ،طيف الامتصاص الالكتروني UV-vis وطيف الكتلة والموصلية المولارية والشدة المغناطيسية والتحليل العنصري[CHNS] وطيف الرنين النووي المغناطيسي H NMR وكذلك طيف الاشعة تحت الحمراء IR. تبين من خلال نتائج تحليل العناصر تكون المتراكبات بنسبة [1:1] [M:L] كما اثبت نتائج الموصلية المولارية ان متراكب النيكل الثنائي دو طبيعة غير الكتروتية اما متراكبات الكروم الثلاثي والنحاس الثنائي كانت لهما طبيعة الكتروتية واثبتت نتائج اطياف الاشعة تحت الحمراء ان الترابط بين قاعدة شيف والعناصر الانتقالية كان من خلال ذرات النيتروجين كما اثبتت نتائج الامتصاصات الإلكترونية وجود انتقالات في قاعدة شيف الحرة ومتراكباتها، النتائج اثبتت ان المتراكبات لها تشكل هندسي تماني السطوح.
الكلمات المفتاحية: قاعدة شيف، متراكبات العناصر، الدراسة الطيفية، النشاط البيولوجي.

Introduction:

Schiff bases are very important structures for synthetic organic chemistry. They were discovered by a German chemist, Nobel Prize winner, Hugo Schiff in 1864 [1]. Compounds that containing an azomethine group (-CH=N-), known as Schiff bases formed by the condensation of a primary amines with a carbonyl compounds such as aldehyde and ketone (Scheme 1.) [2,3]. Where R_1, R_2 and R_3 alkyl or aryl



Aldehyde/ Ketone **Schiff base(imine)**

Scheme 1: Formation of Schiff base

Many Schiff bases were prepared by the condensation reaction of certain aromatic amines with aromatic aldehydes derivatives, then the fluorescence properties of these Schiff bases were examined in acidic and basic media. These compounds can be used for spectrofluorimetric monitoring of small pH changes [4]. The synthesized Schiff bases were characterized by spectral techniques and the Schiff bases were yellow color solid and having sharp melting point and insoluble in organic solvents [5]. Thirty-two Cu(II), Ni(II) and Zn(II) complexes with Phthalaldyde, thiosemicarbazones, were synthesized. All ligands and their metal complexes were tested as inhibitors of human leukemia (HL-60) cells growth and antibacterial and antifungal

activities [6]. Three Ru(III) complexes of general formula Na[RuL2] (where L = dibasic tridentate thiosemicarbazone ligands) have been synthesized by reacting $RuCl_3$ salt with thiosemicarbazidebased ligands of ONS donors [7]. Thiosemicarbazones have been the subject of studies not only for coordination chemistry reasons, but for pharmacological as well, due to their good complexing properties and significant biological activity [8]. Schiff base complexes of 4-(2-hydroxbenzhlodeamino)-3-hydroxnaphtaalene-1-sulfonic acid have been synthesized and investigated by (CHNS) elemental analysis, magnetic moment and molar conductivity, IR and electronic spectroscopies[9]. The aim of the study to prepare and cauterization of Schiff base and its complexes and tested to bacteria.

MATERIALS AND METHODS:

All chemicals and solvents used in this work were of analar Grade (Aldrich, BDH) [10]. They include; Phthalaldyde, thiosemicarbazid**e** and some metal salts, ($CrCl_3.6H_2O$, $NiCl_2.6H_2O$ and $CuCl_2.2H_2O$), absolute ethanol, glacial acetic acid, ammonia solution, ether and dimethylforamide (DMF). These solvents were either spectroscopically pure, then tested for their spectral [11].

Synthesis of Schiff base

N,N′-((*1Z,1′E*)1,2-phn-ylenebis(methaneylylidene))bis(hydrazinecarbothio amide) (H_2L) was synthesized by adding (6.725 g, 0.05 mole) of Phthalaldyde in 30 mL ethanol dropwise to thiosemicarbazide (4.557 g, 0.05 mole) in 50 mL of absolute ethanol. The reaction mixture was refluxed for three hours. The obtained product was allowed to cool at ambient temperature, filtered and recrystallized from ethanol, and then dried under vacuum to get brown precipitate yielded 72%.

Synthesis of the complexes

These complexes were synthesized by adding the Schiff base H_2L (2.804 g; 0.01 mole) in 50 mL absolute ethanol to 0.01 mole of the metal salts of $CrCl_3.6H_2O$, $NiCl_2.6H_2O$ and $CuCl_2.2H_2O$, (2.665 g, 0.01mole), (2.377 gm, 0.01 mole) and (1.705 g, 0.01mole) respectively, in the same amount of the absolute ethanol. The reaction mixtures were refluxed for three hours and the isolated solid complexes were filtered, recrystallized from hot ethanol and finally kept in a desiccator over silica gel.

Bacteria assay

The Schiff base and its complexes with Cr(III), Ni(II) and Cu(II) ions were added separately to the mixture of DMF and H_2O solvents (1:1). The obtained mixtures were further purified and filtrated by using Whitman filter paper No 1. Then stock solutions of extracts were sterilized by filtration using a Millipore membrane filter of 0.2 µm pore-size [12]. The sterile mixture resulted from each compound was stored at 40°C for further uses, and the stock mixtures of the compounds were tested against four pathogenic bacteria species (*Escherichia coli, Proteus Sp, Pseudomonas aeruginosa and Staphylococcus aureus*). Antibac,ameter) diffusion method. Petridishes containing Mueller Hinton agar medium were seeded with a 24 hrs. The culture of the bacterial were growth on nutrient agar. Each well was filed with 50µl of the compound. Solvents were used as a negative control. Inoculated plates were incubated at 37 °C for 24 hrs. The assessment of antibacterial activity was based on measurement of the diameter of inhibition formed around the well [13].

RESULTS AND DISCUSSION:

Microanalysis of the Schiff base and its complexes

The Schiff base ((*2Z,2′E*)-2,2′-(1,2-phenylenebis(methaneylylidene) bis (hydrazine-1-carbothioamide) H_2L was synthesized by the reaction of Phthalaldyde which was added dropwise to thiosemicarbazide. The Schiff base was subjected to: CHNS elemental analyses, mass spectra, IR, UV-Vis and proton nuclear magnetic resonance. The values of the molar conductance of the complexes obtained in solvent DMF in the range of 14 - 128 $ohm^{-1}\ cm^2\ mol^{-1}$ indicating that the Ni(II) complex is non-electrolytic, whereas, the complexes of Cr(III) and Cu(II) are electrolytic behavior[14]. Some physical properties of the Schiff base and its complexes under investigation are listed in Table 1. The obtained CHNS data were in a good agreement between the calculated and experimental values.

Table (1): CHNS elemental analyses and some physical properties of the Schiff base H_2L and its complexes

Ligands/ complexes	M.Wt.	Color	%Calc. **(Found)** C%	H%	N%	S%	**Λ (µs)**	BM	**BM**
L ($C_{10}H_{12}N_6S_2$)	280.37	brown	42.84 **(42.22)**	4.31 **(3.79)**	29.98 **(29.13)**	22.87 **(23.32)**	-	-	-
$[CrLCl_2]Cl$	438.73	green	27.38 **(28.13)**	2.76 **(2.98)**	19.16 **(18.71)**	14.62 **(15.09)**	87	3.68	3.68
$[NiL(Cl_2)]$	409.96	Brown	29.30 **(28.78)**	2.92 **(3.11)**	20.50 **(19.68)**	15.64 **(16.33)**	14	2.71	2.71
$[CuL(H_2O_2)]Cl_2$	450.84	Light green	26.64 **(27.48)**	3.58 **(2.98)**	18.64 **(19.79)**	14.22 **(14.98)**	128	1.79	1.79

Mass spectra of the Schiff base

The formulation of the Schiff base H_2L ($C_{10}H_{12}N_6S_2$) (Fig.**1)** is clearly supported from the presence of intense molecular ion peak 280 in the mass spectrum [15]. (Figure 1).

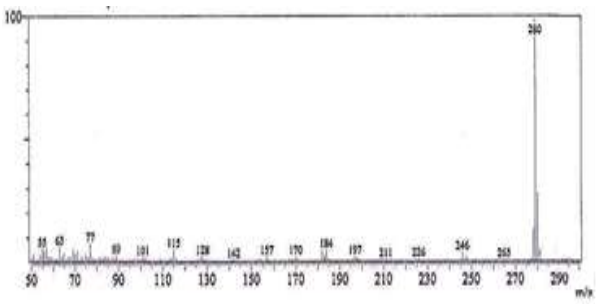


Figure 1: Mass spectrum of $H_2L(C_{10}H_{12}N_6S_2)$

Proton nuclear magnetic resonance spectra of the Schiff base (L)

^{1}HNMR Spectral Studies:

The ^{1}HNMR spectrum of the ligand (H_2L) was recorded in DMSO–d6 solvent Fig.2. The ligand (L), shows amide proton [-NH] at δ 11.810 (s, 1H) as a singlet. The azomethine proton [-N=CH-] has appeared atδ 9,846 (s, 1H) as a singlet, nine aromatic protons have resonated in the region δ 8.138 – 8.305 (m, 4H) as multiplet. The signal at δ 7.479(s, 2H, -NH_2) is due to the proton of –NH[16,17].

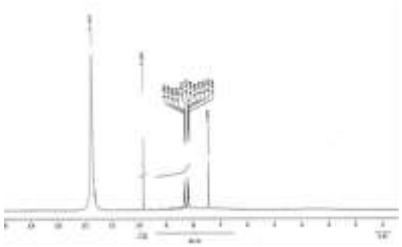


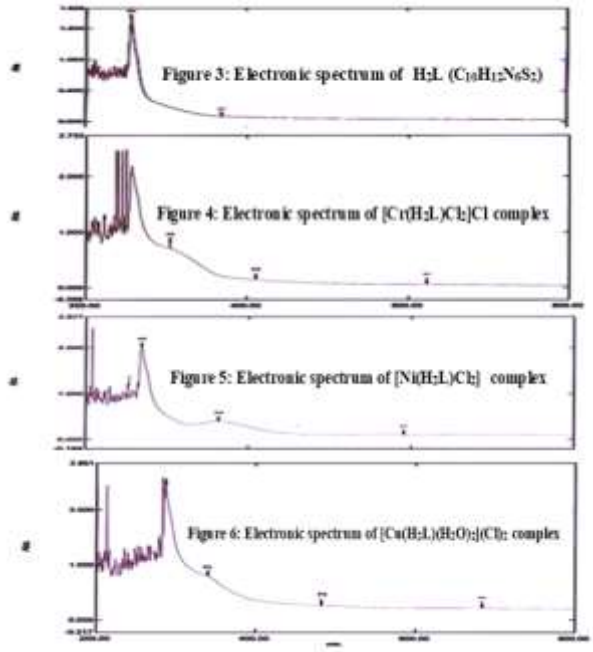
Figure 2: ^{1}HNMR spectrum of H_2L ($C_{10}H_{12}N_6S_2$)

Electronic spectra

The electronic spectral data of the Schiff base (H_2L) and its complexes with Cr(III), Ni(II) and Cu(II) ions are summarized in Table (2) and figures (3 – 6). The electronic spectral data of the Schiff base (L) display two absorptions bands at 37037 and 28169 cm^{-1} corresponding to π→π* and n→π* transitions, respectively [18,19]. The spectral data of the Cr(III) complex show three absorptions bands laying at 16129, 24096 and 32258 cm^{-1}corresponding to $^4A_{2g(F)} \rightarrow {}^4T_{2g(F)}$, $^4A_{2(F)} \rightarrow {}^4T_{1g(p)}$ and CT transitions, respectively [20]. While, three absorptions bands were observed for Ni(II)L at 16949, 17391 and 37735 cm^{-1} showing $^4A_{2g(f)} \rightarrow {}^4A_{1g(p)}$, $^4A_{2g(f)} \rightarrow {}^4A_{1g(f)}$ and CT transition respectively [21]. The electronic spectrum of Cu(II)L shows three absorptions bands at 14388, 21052 and 30769 cm^{-1} which are due $^2B_{1g} \rightarrow {}^2A1g$, $^2Eg \rightarrow {}^2T_{2g}$ and CT charge transfer absorption [22].

Table (2): Infrared and electronic spectral data of the Schiff base (H_2L) and its complexes

Ligand/ Complexes	IR (cm^{-1})					UV – Vis
	νNH	νNH_2	νC=N	νC=S	νM-N	λ_{max} (cm^{-1})
H_2L ($C_{10}H_{12}N_6S_2$)	3410	3266	1609	757	-	(37037) π→π*, (28169) n→π*
$[Cr(H_2L)Cl_2]Cl$	3406	-	1617	762	472	(16129) $^4A_{2g(F)} \rightarrow {}^4T_{2g(F)}$, (24096) $^4A_{2(F)} \rightarrow {}^4T_{1g(p)}$ and (32258)CT
$[Ni(H_2L)(Cl_2)]$	3420	-	1612	760	574	(16949)$^4A_{2g(F)} \rightarrow {}^4A_{1g(P)}$, (17391)$^4A_{2g(F)} \rightarrow {}^4A_{1g(F)}$ and (37735)CT
$[Cu(H_2L)\ (H_2O_2)]Cl_2$	3403	-	1614	760	468	(14388)$^2B_{1g} \rightarrow {}^2A1g$, (21052)$^2Eg \rightarrow {}^2T_{2g}$ and (30769) CT



Infrared spectra of the Schiff base and its complexes:

The infrared spectral data of the Schiff base and its complexes are summarized in table 2 and their spectra are presented in Figures 7–10. The infrared spectral data of the Schiff base show a band at 3410 cm^{-1} which is assigned to υ(NH) group and two bands at 3266 cm^{-1} and 3141 cm^{-1} which attributed to υ(NH_2) group [23,24]. In addition a band at757cm^{-1} assigned to υ(C=S) vibration[25-28]. Also one band at1609 cm^{-1} attributed to υ(HC=N) group. In the complexes the band at 1609 cm^{-1} was shifted to 1617, 1612 and 1641cm^{-1} for complexes $[Cr(H_2L)(Cl)_2]Cl$, $[Ni(H_2L)(Cl)_2]$ and $Cr(H_2L)(H_2O)_2]Cl_2$ respectively [29,30], suggesting a coordination of metal ions through the nitrogen atom of the azomethine group HC=N [31]. New low intensity bands which are not present in the spectrum of free Schiff base appeared at 532-574 cm^{-1} are attributed to υM-N stretching vibrations for Cr(III), Ni(II) and Cu(II) complexes, respectively. The appearance of M-N vibrations support the involvement of nitrogen

atoms in chelation with the metal ions under investigation [32,33].

Figure 7: IR spectrum of the free ligand (H_2L) ($C_{10}H_{12}N_6S_2$)

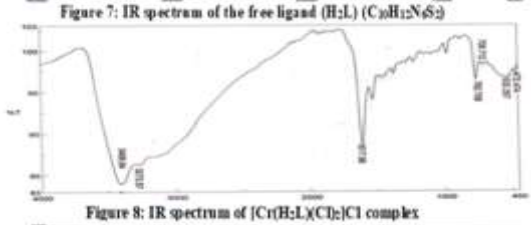


Figure 8: IR spectrum of $[Cr(H_2L)Cl_2]Cl$ complex

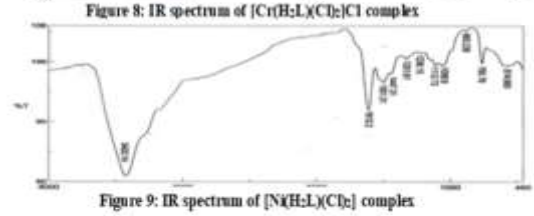


Figure 9: IR spectrum of $[Ni(H_2L)(Cl_2]$ complex

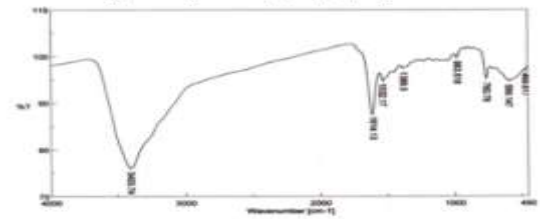


Figure 10: IR spectrum of $[Cu(H_2L)(H_2O)_2]Cl_2$ complex

Antibacterial activity

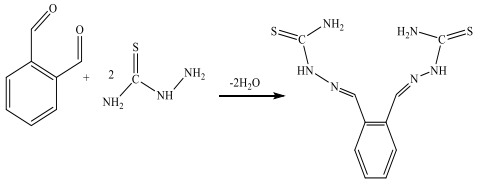
Antibacterial activities of the Schiff base and its complexes were screened against *Proteus sp.*, *Pseudomonas aeruginosa*, *Staphylococcus aureus* and *Bacillus substilis* species. The Schiff base and its complexes show inhibitory activity against all used bacteria species, it was 07-09 mm for the free Schiff base and 09-17 mm for the complexes. The antibacterial results (mm) are presented in Table 3. The antibacterial activity results of the Schiff base and its complexes show a weak to good activity when compared to the control (DMF)[34,35]. It is evident from the above data that the antibacterial significantly increases on coordination. This enhancement in the activity may be rationalized on the basis of their structures mainly possessing an additional azomethine bond. It has been suggested that the Schiff base with nitrogen and oxygen donor systems inhibit enzyme activity [36,37]. Coordination reduces the polarity of the metal ion mainly because of the partial sharing of its positive charge with the donor groups within the complexes ring system[38,39].

Table 3: Inhibition zone of Schiff base and its complexes in mm

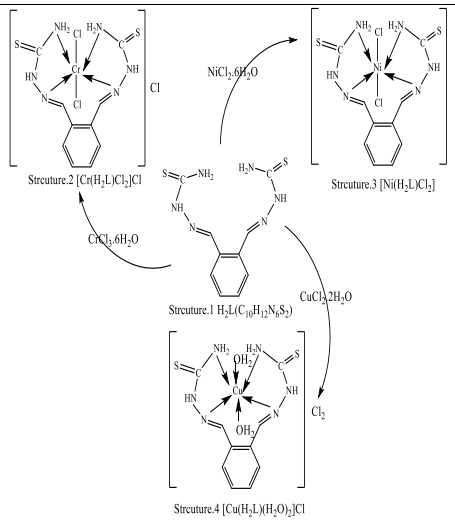
Ligand/ Complexes	*Bacillus subtilis*	*Staphylococcus aureus*	*Pseudomonas aeruginosa*	*Proteus sp.*
H_2L; ($C_{22}H_{20}N_2$)	80	07	09	08
$[Cr(H_2L)Cl_2]Cl$	10	11	12	11
$[Ni(H_2L)\ Cl_2]$	09	12	17	09
$[Cu(H_2L)(H_2O)_2]Cl_2$	11	10	11	10

Conclusion:

On the basis of the elemental composition, electronic and IR spectral studies, the following structures (1-4) **Scheme 3** are proposed for the synthesized Schiff base and its complexes. The overall reaction of Phthalaldyde and thiosemicarbazide to produce a ligand; ((2*Z*,2′*E*)-2,2′-(1,2 phenylenebis (methaneylylidene)-bis(hydrazine-1-carbothioamide) HL is shown below (**Scheme 2**).



Scheme -2: Synthesis of Schiff base (H_2L)



Scheme 3: Synthesis of the complexes

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