

Synthesis , Characterization and Biological Activity of Chromium(III), Manganese,Cobalt,Nickel,Copper and Zinc(II) Complexes with New Schiff base derived from 6-methylquinoxaline-2,3(1H,4H)-dione and Diethylenetriamine

Abstract: The chromium(III),manganese,cobalt,nickel,copper and zinc(II) complexes were prepared by direct reactions of their metal chlorides with new Schiff base(3Z)-3,3'-((azanediylbis(ethane-2,1-diyl))bis(azanylylidene))bis(6-methyl-3,4-dihydroquinoxalin-2(1H)-one) and fully characterized with help of GC-mass,H,¹³C NMR spectra as well as elemental analyses.The magnetic susceptibility of solid complexes and their molar conductance revealed the octahedral geometry for all complexes with [ML(H₂O)₂]Cl₂,M=Mn,Co,Ni,Cu,Zn(II) and [CrL(H₂O)Cl]Cl₂ formula.Further more the antimicrobial activity of the complexes were scanned against two bacteria and fungi and it is found that chromium(III) complex showed the highest inhibition zone towards the selected micro-organisms while the other complexes exhibited medium activity with relative to dimethylfoxide standard and *Amoxyciline* contro.

Key words: Poly dentate N2O2 ligands,Schiff bases of quinoxalines,transition metal chelates,antimicrobial activity.

1.Introduction

Heterocyclic compounds containing nitrogen atom attract a great deal of attention in coordination chemistry and homogeneous catalysis [1–3]. The combination of heterocyclic ring and azomethine moiety exerts potential biological and catalytic activities[4–6]. Although extensive studies have been made on the transition metal complexes of various nitrogen heterocyclic Schiff base ligands, comparatively less is reported on transition metal complexes derived from 2-oxo-1,2-dihydroquinoline-3-carbaldehyde[7–9].Sivakumar P. and coworkers have reported the binuclear palladium(II) complexes with Schiff bases derived from 2-oxoquinoxaline-3-carboxaldehyde and investigated their main role in **catalytic** of imidazole synthesis[10]. **As well as, the binuclear Co,Ni,Cu and Zn(II) complexes with polydentate Schiff bases of quinoxaline-2,3-dione have investigated with their antimicrobial activity** [11]. **As well as AL-Jibouri M.N. has investigated the template Zn,Cd,Hg(II) and Zr(IV) chelates with Schiff bases of quinoxaline-2,3(1H,4H)-dione**[12].The extensive studies of biological activity related to metal(II) chelates of benzopyrazine **have been keeping** by scientists especially DNA cleavage[13]. Synthetic quinoxaline nucleus is the part of a number of antibiotics which are known to inhibit the growth of gram positive bacteria and are also active agents to treat tuberculosis .Recent studies has shown that some quinoxaline derivatives inhibit selectively the platelet derived growth factor (PDGF) receptor kinase[14].

2.1. Instrumentation

Elemental analyses(C.H.N.S) of the new Schiff base HL and its metal(II) complexes were determined using Carlo-Erba 1106 Elemental analyzer.The electronic spectra were recorded for solutions of 6,7-dimethyl quinoxalin-2,3-dione(DMQ),HL ligand and its metal metal complexes using Shimadzu spectrometer in the range 200-800 nm in DMF solvent. The ¹H and ¹³C NMR spectra were carried at Al-Yarmook University,Amman- Jordan on Bruker 300 MHz spectrometer in DMSO-*d*₆ solvent. The Fourier transform infrared spectra of the prepared complexes were recorded in KBr and CsI discs on Shimadzu model FT-IR-8400 spectrometer at the Laboratories of Chemistry Department, College of Science, Al-Mustansiriya University,Baghdad-Iraq.The molar conductance measurements were made on an Philips conductivity bridge type CM-82 with a cell having a cell constant of 1.1 cm⁻¹ in DMSO solutions. The percents of metal contents of the complexes were determined by flame atomic absorption on Shimadzu A.A-670 spectrometer at instrumental analyses laboratory-chemistry department-Al-Mustansiriya university-Baghdad-Iraq.

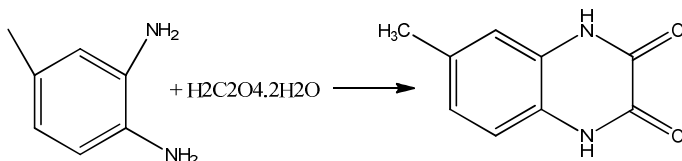
2.2. Materials

The hydrated metal chlorides CrCl₃.6H₂O, MnCl₂.4H₂O, CuCl₂.2H₂O, CoCl₂.6H₂O and NiCl₂.6H₂O and ZnCl₂ were purchased from Sigma-Aldrich company and used without purification. The oxalic acid, 4,5-dimethyl,1,2-phenylenediamine and solvents were supplied from Fluka company in 99% purity. All other chemicals used were of Analar grade.

2.3.Methods

2.2.1. Synthesis of 6-methyl- 2,3-quinoxaline-dione(A)

The 6-methyl- 2,3-quinoxaline-dione was prepared according to the method published in literature[10],Scheme 1.



Scheme 1.synthesis of A compound.

Color:White off.

Yield: 90%.

M.p.: 300 decomposed °C.

FT-IR (KBr, ν , cm^{-1}): 3200-3270 (NH) (Quinoxaline amido),1735, (C=O) (Lactam), 1600 (-C=N-) (Amide pyrazine),1330(-C-N) (Pyrazine ring).

UV/Vis (Methanol, λ_{max} , nm, ϵ): 300 (2.88), 277 (3.492).

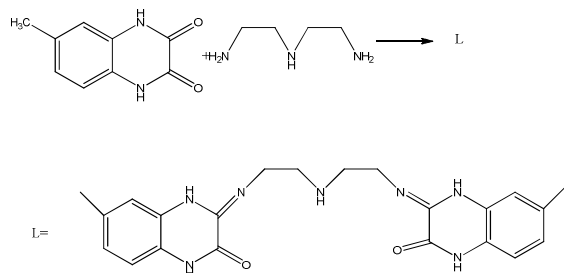
^1H NMR (300 MHz, $\text{DMSO}-d_6$, δ , ppm): 2.50 (s, dms), 3.70 (s, 23H, CH3), 4.66(s, 1H, -NH-CO),8.90(s,1H,-NH-C-N),6.70-7.60(m,3H,Ar-H)

Anal. calcd. for $\text{C}_9\text{H}_8\text{N}_2\text{O}_2$: C, 63.15; H, 5.3; N, 14.73. Found: C, 62.67; H, 4.99; N, 13.67.

MS (EI, m/z (%): 176.055(M^+ , 100).

Synthesis of thiosemicarbazone ligand (3Z)-3,3'-((azanediylbis(ethane-2,1-diyl))bis(azanylylidene))bis(6-methyl-3,4-dihydroquinoxalin-2(1H)-one)

0,01 mole of diethylenetriamine [0.162 g]in 30 ml ethanol was added gradually to 6, -imethyl-quinoxaline 2,3-(1,4H)-dione(A)(1.92 g, 0.01 M). The reaction mixture was refluxed for 8 hours on heating mental and the pale yellow solid separated was filtered, washed several times with ethanol and dried in oven, scheme(2).



Color: Pale yellow

Yield: 78%.

M.p.: 270-272 °C.

FT-IR (KBr, ν , cm^{-1}): 3400-3570 (NH) (Quinoxaline amido),1700-1653 (C=O) (Lactam),16033 (-C=N-,Imine), 1589-1570(-C=N) (Pyrazine ring),2973(C-H,CH3,CH2-).

UV/Vis (Methanol, λ_{max} , nm, ϵ): 350 (2.77), 233 (3.111).

(300 MHz, $\text{DMSO}-d_6$, δ , ppm): 2.5-3.40 (s, 6H, MSO), 1.1-1.2 (s, 3H, CH3), 2.1-2.38(M,4H,-CH2-NH_),3.35(m,4H,CH2-CH2-N=C-),3.35(s,2H,-NH),7.05-7.1(m,6H,Py-H),12.20(s,2H,-NH-pyrazine).

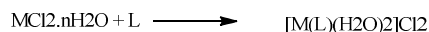
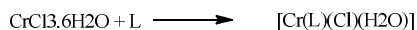
Elemental Analysis: Calc. C, 62.99; H, 6.01; N, 23.37.Found.C,61.90;H, 5.88; N, 22.81.

^{13}C NMR (300 MHz, $\text{DMSO}-d_6$, δ , ppm) of L1 compound : 161.20(C, C=O Amide pyrazine), 154.71 (C,-N Imine), 139.20(C, C-NH-), 127.7(C,C-N- Pyrazine), 31.60-131.50 (C, -C-C-NH), 130.60-129.80 (C,C=N-Ar), 127.70 (C, C=C-Ar), 124.29(C, C-Ar), 114.82(Ar-C=C-CH3),25.90(C, CH2-NH), 21.1 (C,CH2-N=C-),10.50(C,CH3-).

MS (GC-Mass, m/z (%): 420.035(M^+ , 50),342,282,176.

2.4.Synthesis of metal complexes

An ethanolic solution (100 ml) of metal(II) chloride [$\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$ (0.221g, $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ (0.237 g, 0.001 M), $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$ (0.237 g,0.01 M), $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ (0.170 g, 0.01 M) , ZnCl_2 (0.135 g, 0.001 M)] and $\text{CrCl}_3 \cdot 6\text{H}_2\text{O}$ (0.1370g) was added with stirring to an ethanol solution of the ligand [L (0.420 g, 0.001 M)] and refluxed on water bath for 6–8 hours. So obtained solid complex was separated by filtration under suction, washed with hot ethanol and dried in vacuo,scheme(3).



M=Mn, Co, Ni, Cu and Zn(II), n=4, 6 and 2 for Mn, Co, Ni and Cu(II) respectively. **3.2. Antibacterial activity**

Primary screening

The antibacterial activities of the newly synthesized complexes were evaluated by the Agar Well Diffusion Assay Technique against two gram-positive and negative bacteria, *Staphylococcus aureus* and, *Escherichia coli*, two gram-negative and positive fungi, *Aspergillus fumigatus* and *Candida albicans*. The bacterial cultures were maintained on the nutrient agar media by sub-culturing them on fresh slants after every 4-6 weeks and incubating them at the appropriate temperature for 24 h. All stock cultures were stored at 5 °C. For the evaluation of antimicrobial activity of the synthesized complexes, a suspension of each test microorganism was prepared. The turbidity of each suspension was adjusted to 0.5 McFarland units by suspending the cultures in sterile distilled water. The size of final inoculums was adjusted to 6×10^8 CFU mL⁻¹. Volume of 20 mL of agar media was poured into each Petri plate and the plates were swabbed with broth cultures of the respective micro-organisms and kept for 15 min for adsorption to occur. Using a punch, ≈8 mm diameter wells were bored in the seeded agar plates and a 100 µL volume of each test compound reconstituted in DMSO was added into the wells. DMSO was used as the control for all the test complexes. After holding the plates at room temperature for 2 h to allow diffusion of the compounds into the agar, the plates were incubated at 37 °C for 24 hours [15]. The antibacterial activity was determined by measuring the diameter of the inhibition zone. The entire tests were performed in triplicate and the mean of the diameter of inhibition was calculated. The antimicrobial activities of the complexes were compared against standard drugs.

Minimum inhibitory concentration (MIC)

Nutrient broth was adjusted to pH=7.0 for the determination of the MIC of synthesized metal complexes. The minimum inhibitory concentration (MIC) is the lowest concentration of an antimicrobial agent that prevents the development of visible growth of microorganism after overnight incubation. The inoculum of the test micro organisms was prepared using 20 h-old cultures adjusted by reference to the 0.8 McFarland standards (11 cells mL⁻¹). These cultures were further diluted up to 5-fold with nutrient broth to obtain an inoculums size of 1.5×10^6 CFU mL⁻¹. A positive control (containing inoculums but no complex) and a negative control (containing complex but no inoculums) were also prepared. A stock solution of 5 mg mL⁻¹ of each compound was prepared in DMSO and further appropriately diluted to obtain final concentrations ranging from 100 to 3.0 µg mL⁻¹. The requisite quantity of antifungal drug (*Gentamycine*) was added to the broth to obtain its desirable final concentration of 250 µg mL⁻¹. Separate flasks were taken for each test dilution. To each flask was added 100 µL of inoculums. Then the appropriately diluted test sample was added to each flask having broth and microbial inoculums. The contents of the flask were mixed and incubated for 48 h at 37 °C. The test bacterial cultures were spotted in a predefined pattern by aseptically transferring 10 µL of each bacterial culture onto the surface of solidified agar-agar plates and incubated at 37 °C for 24 h for determining the M.I.C. value.

Antifungal study

Potato dextrose medium (PDA) was prepared in a flask and sterilized. To check the growth of bacterial culture in the medium, the requisite quantity of the standard antibiotic (ampicillin) was added to obtain the desired final concentration of 100 µg mL⁻¹ of the medium. Test samples were prepared in different concentrations (10 and 50 µg mL⁻¹) in dimethyl sulphoxide and 100 µL of each sample was spread on the PDA media contained in sterilized Petri plates. Mycelial discs taken from the standard cultures of fungi (*Aspergillus fumigatus* and *Candida albicans*) were grown on the PDA medium for 2-4 days. These cultures were used for the aseptic inoculation in the sterilized Petri dish. Standard cultures inoculated at 25±1 °C were also used as the control. The efficacy of each sample was determined by measuring the radial mycelial growth. The radial growth of the colony was measured in two directions at right angle to each other and the average of two replicates was recorded in each case.

The data are expressed as percent inhibition over the control obtained from the size of colonies. The percent inhibition (ϕ) was calculated using the formula:

$$\phi = 100 \frac{C - T}{C} \quad (1)$$

where

C is the diameter of fungus colony in the control plate after incubation for 48 hours and

T is the diameter of the fungus colony in the tested plate after the same incubation period [16].

Table 4. Antimicrobial activity of the prepared metal complexes

Compound	Inhibition			
	<i>E. coli</i>	<i>S. aurea</i>	<i>Fungi</i>	
	G ⁺	G ⁺	Asp.	Cand.
[CrL(H ₂ O)Cl]Cl	17	23	10	17
[MnL(H ₂ O) ₂]Cl ₂	15.2	27	11	15
[CoL(H ₂ O) ₂]Cl ₂	22	30	29	20
[NiL(H ₂ O) ₂]Cl ₂	12	11.8	17	13
[CuL(H ₂ O) ₂]Cl ₂	9	11	12	10
<i>Amoxycilline</i>	33	38	22	19

3.Results and Discussion

The new metal complexes prepared in this course were non-hygroscopic (stable at the room temperature) and in the form of amorphous solids. These are soluble easily in DMSO, DMF and sparingly in ethanol and methanol whereas they are insoluble in chlorinated hydrocarbons. The elemental analysis data of the ligand and its metal complexes along with their physical properties are shown in Table 1.

Table 1. The physical properties and elemental analysis of the prepared metal complexes.

Compound	Color	M.p. °C*	Molecular Formula	C% Cald. (found)	H% Cald. (found)	N% Cald. (found)	M% Cald. (found)
[CrL(H ₂ O)Cl]Cl ₂	Pale green	309 ^D	595.52	47.43 (46.18)	3.34 (2.92)	18.07 (19.22)	8.75 (8.26)
[MnL(H ₂ O) ₂]Cl ₂	Pink	326 ^D	580.68	-	-	-	9.44 (8.87)
[CoL(H ₂ O) ₂]Cl ₂	Olive	317 ^D	582.89	-	-	-	10.30 (9.828)
[NiL(H ₂ O) ₂]Cl ₂	Green	322 ^D	583.09	45.16 (44.81)	5.14 (4.65)	16.75 (17.53)	10.03 (9.56)
[CuL(H ₂ O) ₂]Cl ₂	Dark green	370 ^D	616.04	43.83 (42.50)	2.63 (2.10)	17.04 (17.99)	15.15 (14.97)
[ZnL(H ₂ O) ₂]Cl ₂	Off white	360 ^D	586.20	47.55 (46.84)	4.11 (4.00)	19.22 (20.44)	11.06 (10.86)

*D=decomposed.

3.1.GC-Mass spectra

The figure 1.shows the absorption that ascribes molecular weight of A compound with $m/e=176$ of 20% intensity due to formation of base peak at 131 up on fragmentation of $-OH$ and $-CH_3$ groups simultaneously.However,the peaks at 175,157 and 147 may be assigned to $M-H$, $M-OH$ and $M-(2OH,H)$ respectively[16]. The gas-chromatography mass spectrum of L1 ligand is shown in figure 1.The molecular ion peaks at 421 and 422 indicate the molecular weight of $C_{22}H_{25}N_7O_2$ formula with effect of deuterium and carbon isotopes. The fragments at 405,386,375 and 246 peaks confirms the departure of $-CH_3$ group and breaking of weak points at $-HN-CH_2-CH_2-$ respectively[17],figures 1-2.

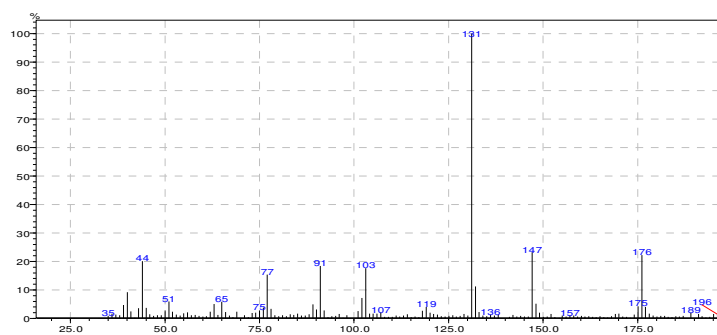


Figure 1.Mass spectrum of A compound.

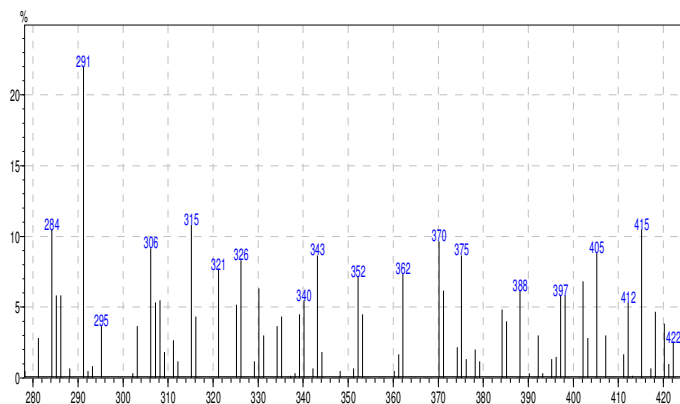
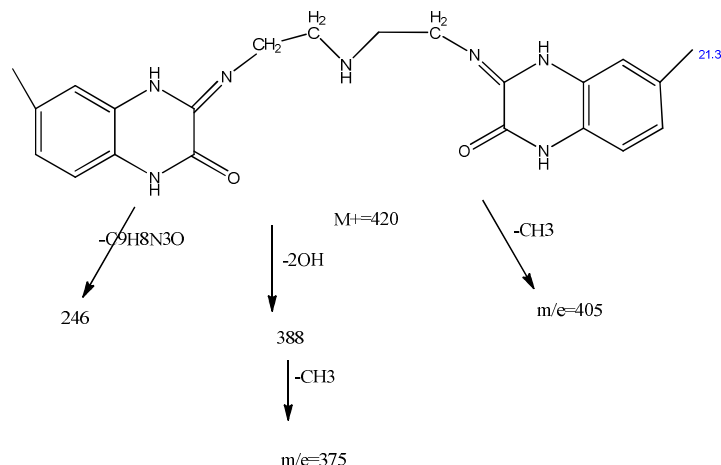


Figure 2.Mass spectrum of L ligand.



Scheme 4.proposed mechanism of L1 fragmentatiions via GC-mass spectra.

3.2. Molar conductivity measurements

The molar conductance values of the complexes measured at room temperature in DMF. solution with 0.001 mol/dm³ concentration fall in the range 140–200/ohm .cm²/mol indicating the electrolytic nature of the complexes[18],due to presence of chloride ions as a counter ions thus supports the $[M(L)(H_2O)_2]Cl_2$ formula. However the chromium(III) complex in DMF solution shows molar conductance value of 80 ohm-1.cm².mol.⁻¹. That reveals one chloride ion in the outer shere of complex structure,Table 4.

3.3.IR spectra

The IR spectra of metal complexes are presented in (Table 2.) The appearance of strong absorption band in the region 1747-16383 cm⁻¹ corresponds to $\nu C=O$ stretching frequency supporting the carbonyl of pyrazine ring that it has forwardshift up on imine prepared due to ring strain. A medium absorption recorded in the regions 3103 cm⁻¹ may be ascribed to $\nu N-H$ [13,19-23].As well as the new shar peak at 1638 cm⁻¹ reveals the imine $-C=N-$ bond of Schiff base. The lowering in the values of wave numbers of $\nu(C=N)$ and $\nu(C=O)$ may be explained on the basis of drift of lone pair density of imine nitrogen and carbonyl oxygen towards metal atoms[20]. The broad absorptions in the spectra of all complexes in the regions 3196-3600 cm⁻¹ assigning to $\nu(O-H)$ of coordinated water molecules in the inner sphere of metal complexes. The bands present in the range ~1299-1349 cm⁻¹ are assigned to $\nu(C-N)$ vibration. Furthermore, the weak to medium bands of M-O and M-N are shown in the 422-460 and 453-550 cm⁻¹[23].Only chromium(III) complex exhibits far-infrared spectrum in the region 277-370 cm⁻¹ confirming Cr-Cl bond.

Table 2. FT-IR absorptions of the quinoxalin-2,3-dione and its template metal complexes in cm^{-1} .

Complex	νNH , $\nu\text{C}=\text{N}$ -	$\nu\text{C}=\text{O}$	$\nu\text{Ar}-\text{CH}$,	$\nu\text{C}-\text{N}$	$\nu\text{M}-\text{N}$	$\nu\text{M}-\text{O}$	Other bands
L	3103(m),1638(s)	1747-1683(s)	3050, 2962(m)	1370-1340(s)	-	-	707-750 ^a
[CrL(H ₂ O)Cl]Cl ₂	3400(br.), 1578(s)	1657(s)	3060 (w) 2870(m)	1325(m)	453(w)	429(w)	780-810(w) ^a ,277-370 ^b
[MnL(H ₂ O) ₂]Cl ₂	3600(br.), 1590(s)	1677(s)	3080	1299(m)	550	460	766(m)
[CoL(H ₂ O) ₂]Cl ₂	3483 , 1580(s)	1701-1656(s)	3060(w), 2870(m)	1326(s)	453(m),	429(w)	727
[NiL(H ₂ O) ₂]Cl ₂	3169, 1530(s)	1640(s)	3010 (w)	1350(m)	500(w)	433(m)	770-790(w) ^a
[CuL(H ₂ O) ₂]Cl ₂	3418(br.),1610	1683-1651	3064(w)	1349	543(m)	455(m)	
[ZnL(H ₂ O) ₂]Cl ₂	3562-3471(br.),1581(s)	1691(s)	3090(w)	1340	501(m)	422(w)	722(w) ^a

S=strong,m=medium,br.=broad,w=weak, a =rocking of methylene -CH₂ moiety and b= far ir of Cr-Cl bond.

3.4. Electronic spectra and magnetic moments

The magnetic and electronic spectral data is in relevance with proposed geometry of complexes. The free ligand solution displays high intensity peaks in the UV regions at 233 and 350 nm,Table 3 which are assigned to electronic transitions for chromophores - C=N and C=C respectively [13,24]. The green solution of Cr(III) complex in DMF shows two weak absorptions at 600 and 460 nm assigning to ${}^4\text{A}_{2g}(\text{F}) \rightarrow {}^4\text{T}_{2g}$ and ${}^4\text{A}_{2g}(\text{F}) \rightarrow {}^4\text{T}_{1g}(\text{F})$ respectively [25]. The disappearance of third transitions in the near UV region may be attributed to overlapping the MLCT with it,thus confirms the octahedral environment around chromium(III) ion[22]. The electronic transitions from the ${}^6\text{A}_1$ ground term of Mn(II) to higher energy terms are spin-forbidden. However, the band appearing in the region 430-350 nm in the electronic spectrum of Mn(II) complex is assigned to ${}^6\text{A}_{1g} \rightarrow {}^4\text{T}_{1g}(\text{G})$ and is consistent with octahedral geometry. Furthermore, the olive solution of cobalt(II) complex, the peaks at 670 and 550 nm are assigned to the ${}^4\text{T}_{1g} \rightarrow {}^4\text{T}_{2g}(\text{F})$ and ${}^4\text{T}_{1g} \rightarrow {}^4\text{A}_{2g}(\text{F})$ respectively, which is typical for octahedral cobalt(II)complexes. As well as the nickel(II) and copper(II) complexes exhibit weak bands in the visible region and others of high intensity that related to LMCT[23,24].The magnetic moments of Cr(III) complex is 3.45 BM is consistent with expected high spin of d^3 configuration[24,25].However, the Mn(II),Co(II),Ni(II) and Cu(II) complexes recorded expected values of magnetic moments at 5.60,3.80.2.89 and 1.70 BM.respectively [24].The communication of UV-Visible spectra of the prepared complexes with their obtained magnetic properties helped us to adopt the octahedral geometry.

Table 3. The electronic spectra λ (cm^{-1}) and molar conductance of the prepared complexes.

Complex	UV-Visible λ , nm	Λ_m , $\text{S mol}^{-1} \text{cm}^2$	μ BM.	Geometry
[CrL(H ₂ O)Cl]Cl	270, 333	80	3.45	Octahedral
[MnL(H ₂ O) ₂]Cl ₂	290, 360	190	5.60	Octahedral
[CoL(H ₂ O) ₂]Cl ₂	311, 357	167	3.80	Octahedral
[NiL(H ₂ O) ₂]Cl ₂	330, 485	200	2.80	Octahedral
[CuL(H ₂ O) ₂]Cl ₂	325, 396	170	1.70	Octahedral
[ZnL(H ₂ O) ₂]Cl ₂	290,370	140	Diamgnetic	Octahedral

Λ_m = molar conductances were measured in DMF solutions.

3.5. ^1H & ^{13}C NMR study

The proton and carbon-13 nuclear magnetic spectra for L ligand in DMSO-d₆ solution were performed by using tetramethylsilane as internal reference. The figure 3 shows different peaks at 12.20, 7.05-7.60, 3.35, 2.80-2.50 and 1.1-1.20 ppm assigning to resonance of pyrazine -NH, Ar-H, -NH of diethylentriamine, -CH₂-CH₂- and -CH₃ moieties respectively[22]. As well as, the figure 4 exhibits the absorptions of ^{13}C NMR spectra of L in DMSO-d₆ solution where the eleven peaks entirely investigate the presence of 161.20 (C, C=O Amide pyrazine), 154.71 (C, =N Imine), 139.20 (C, C-NH-), 127.7 (C, C-N- Pyrazine), 31.60-131.50 (C, -C-C-NH), 130.60-129.80 (C, C=N-Ar), 127.70 (C, C=C-Ar), 124.29 (C, C-Ar), 114.82 (Ar-C=C-CH₃), 25.90 (C, CH₂-NH), 21.1 (C, CH₂-N=C-), 10.50 (C, CH₃-). The data of carbon-13 NMR reveals the expected numbers of carbon atoms in the structure of L Schiff base, then agree well with H NMR, FTIR, CHN and GC-mass spectra[13,26].

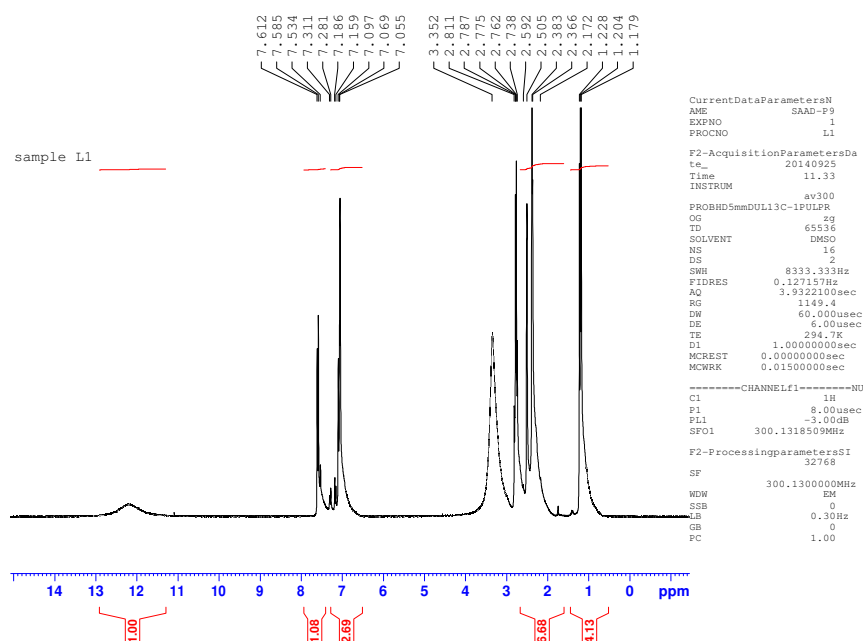
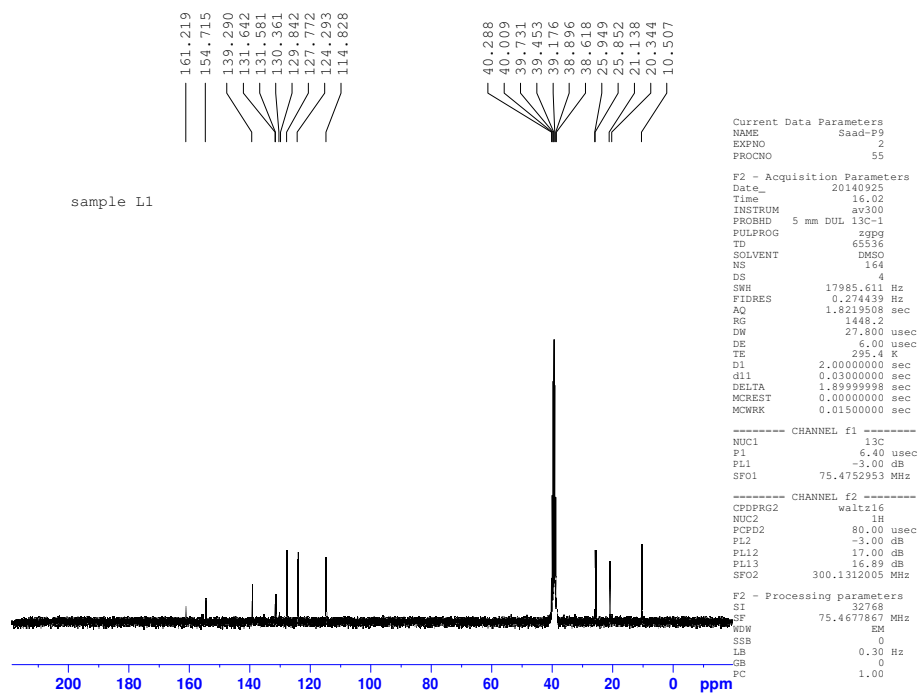


Figure 3. ^1H NMR spectrum of L1 in DMSO-d₆ solution.



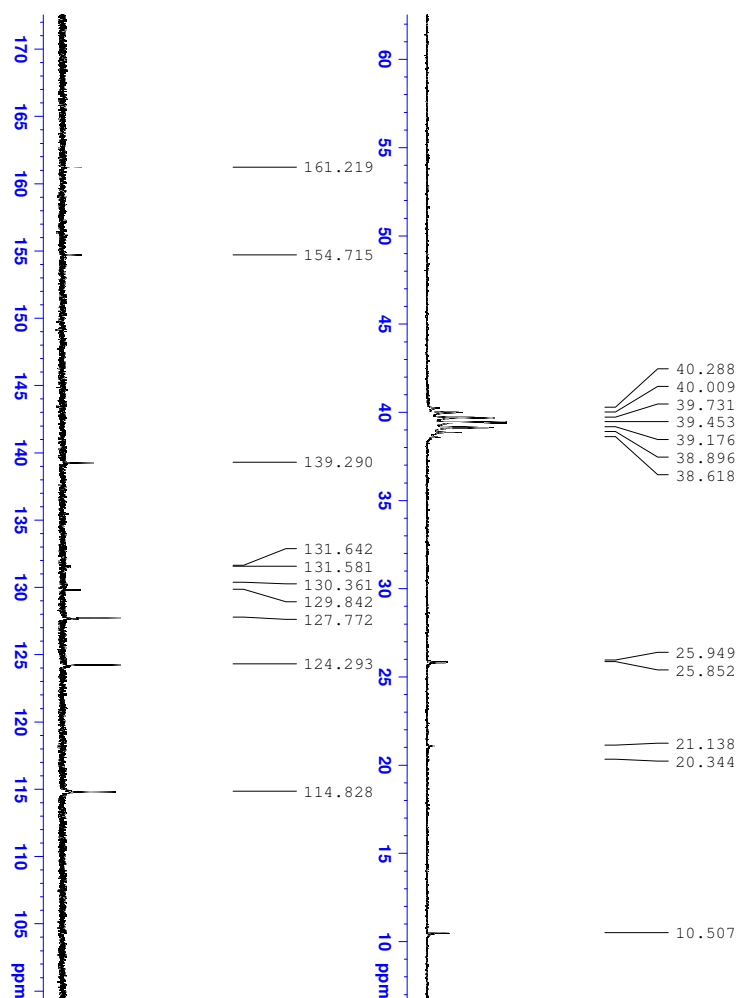


Figure 4. ^{13}C NMR spectrum of L in DMSO- d_6 solution.

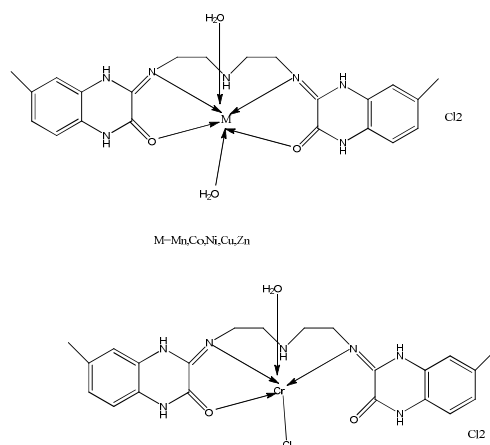
3.6. Antimicrobial studies

In this study, all the chemically synthesized complexes were evaluated against gram-positive and gram-negative bacteria. The MIC values of the synthetic complexes were determined by the method given by Andrews [25].

Standard antibiotic, namely streptomycin was used for comparison with the antibacterial activities exhibited by these complexes. All the complexes of the tested series possessed some antibacterial activity against gram-positive bacteria as well as Gram-negative bacteria, Table 4. In the whole series, cobalt(II) complex solution was found to be most effective against all the tested bacterial strains, showing zone of growth inhibition at 22-30 mm against *bacteria* and 20-29 mm against *fungi* respectively. The other complexes of Cr(III), Mn(II) and Ni(II) exhibited moderate activity against all the tested bacterial strains with a zone of inhibition in the range from 11.0-23 and 5.0-29 mm respectively. This may be interrelated on the basis of chelation theory of antimicrobial activity towards negative and positive gram bacteria [27-].

Conclusions:

The chromium(III), manganese, cobalt, nickel, copper and zinc(II) complexes with tetradentate L ligand of the N₂O₂ donor system were prepared and fully characterized with the help of GC-mass, NMR, FTIR, UV-Visible spectra and elemental analyses. The data of molar conductivity in N,N-dimethylformamide (DMF) reveals the electrolytic behavior of all metal complexes assigning the presence of chloride ions in the outer sphere of complexes formula. The downshift in the carbonyl of pyrazine moiety and terminal imine in the IR spectra for all metal complexes suggests the participation of oxygen and nitrogen atoms of such functional groups in coordination with the central metal ions. The octahedral geometry with [ML(H₂O)₂]Cl₂, M=Mn, Co, Ni, Cu and Zn(II) ions and [CrL(H₂O)Cl]Cl₂ formula were proposed according to data obtained from CHN, NMR, molar conductivity and magnetic moments. As well as the antimicrobial activity of the prepared complexes were screened against some micro-organisms and the obtained inhibition zone data reported the possibility of their applications in treatment of diseases reasoned by these bacteria and fungi.



Scheme 4. Octahedral structure of metal complexes.

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