



Synthesis, Antibacterial Activity and DFT Calculation of Co(II) and Ni(II) Schiff Bases Complexes Derived from Acenaphthenequinone and Phenylenediamine



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Abstract

Tetra-dentate Schiff base ligands. $L_1 = (2Z)-2,2((4\text{-methyl-1,2-phenylene bis (azoneylidene))) \text{ bis (acenaphthylene-1(2H)-one})$, $L_2 = (2Z,2E)-2,2'((4\text{-nitro-1,2-phenylene bis azoneylylidene)) \text{ bis azoneylylidene)) \text{ bis (acenaphthylene-1(2H)-one)}$, $L_3 = 6,6'(((1E,2E)\text{-acenaphthylene -1,2-diylidene)) \text{ bis (azoanylidene)) \text{ bis (3-methylaniline)}$, and $L_4 = 6,6'\text{-}(acenaphthylene-1,2\text{-diylidene)) \text{ bis (azoneylylidene)) \text{ bis (3-nitroanilin)}$, type (N_2O_2) and (N_4) donor atoms obtained from condensation of acenaphthylenequinone with 4-methyl or nitro-*o*-phenylenediamine (2:1) and (1:2) (quinone:amine) molar ratio for the first and second type respectively. These ligands employed used to synthesis complexes in general molecular formula $[ML^nCl_2]$, where $M = \text{Co(II)}$ and Ni(II) and $n = (1-4)$. The ligands and the complexes were characterized by (¹H-NMR, UV-Visible and FTIR) spectroscopy, metal content, elemental analysis C.H.N, magnetic and conductivity measurements. The low conductivity values reveal the non-electrolytic nature for their new complexes. All complexes are hexa co-ordinate with (octahedral geometry). Evaluation biological activity for the compounds tested against Gram(-) bacteria (*Escherichalcoli*, *Pseudoaerogenosa*) and Gram(+) bacteria (*Staphylococcus aureus*, *staphylococcus epidermidis*) and the optimized structure for the compounds carried out by using DFT calculation.

Keywords: Acenaphthenequinone, Schiff base, Phenylenediamine, DFT Calculations.

1. Introduction

Schiff base ligands are still having a great interest by researcher since they have been first synthesized and studied in 1864[1]. They have a board area of various applications from medicine, pharmacology to industry [2]. Tetra-dentate Schiff base derived from acenaphthenequinone and phenylenediamine have been subject of many studies, this is attributed to a variety reason: Generally, tetra-dentate Schiff base distinguished by it's ability to form more stable complexes, where the co-ordination takes place through (N_2O_2) and (N_4) donor [3,4], and these types are more effective as chelating ligand and form 5 or 6 members chelates; such difference in

Schiff base co-ordinated are accountable to great flexibility in the structure of their metal complexes [5,6]. Schiff bases which are derived from diamines such as *o*-phenylenediamine and their complexes have broad important applications in catalysis and co-ordination chemistry[7-9], on the

other hand Schiff bases of acenaphthenequinone are widely synthesized due to their potential applications in various scientific areas[10-13], therefore our work aim to synthesis and characterization of some mononuclear complexes of Co(II) and Ni(II) which contain five membered chelate rings with this type of Schiff base ligands.

2. Experimental:

2.1. Materials and measurement:

In the preparation method; all the reagents and solvents such as: methanol, ethanol, DMF and DMSO, were products from (Fluca or BDH) used as received. The metallic content was estimated spectrophotometrically by Analytic Jena Model Nova AA-350. The prepared compounds were analysed by using the C.H.N elemental analyzer Model 550-carloErba Instrument.

Melting points were registered using SMPLO-Melting Point Apparatus; IR. Spectrum has been recorder on Shimadzu FT-IR-ATR from (400-4000)

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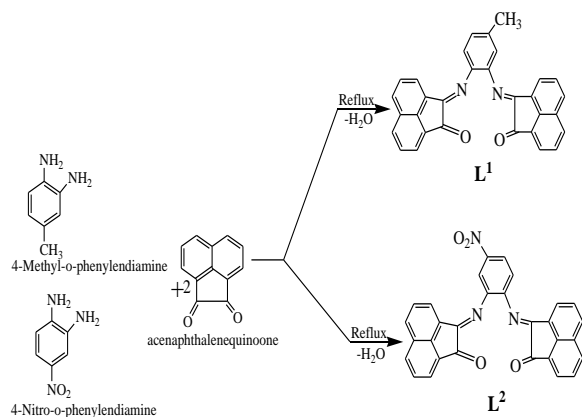
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cm^{-1} using KBr disc. $^1\text{H-NMR}$ spectra were registered at laboratory temperature in DMSO-d_6 using Bruker Bio Spin GMBh (400 MHz), the UV-Vis have been recorded at $25\text{ }^\circ\text{C}$ in DMF (10^{-3} M) solution on Shimadzu UV-1800 UV Spectrophotometers using 1 cm quartz cell from (200-1000) nm. Conductivity –Meter-Model-OAKion-Pc510 used to measure the conductivity of all the prepared complexes, and were carried out in DMF (10^{-3} M) solution at laboratory temperature. The magnetic measurements were carried out on the solid state at 25°C by magnetic susceptibility balance later theoretical studies were executed with Gaussian O3 program to suggested geometric structures of the compounds.

2.2. Preparation of L^1 and L^2 [14,15]

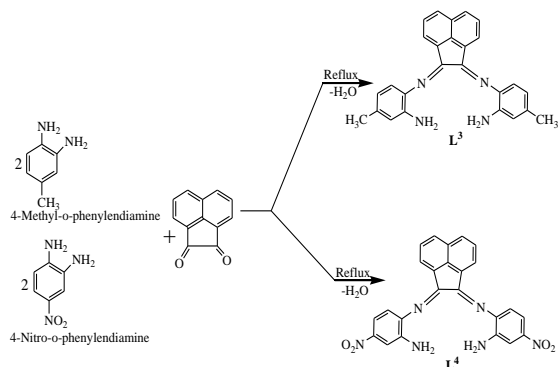
(100 ml) ethanolic mixture of (0.01 mole, 1.221 gm) of 4-methyl-o-phenylenediamine or (0.01 mole, 1.511 gm) of 4-nitro-o-phenylenediamine and (0.02 mole, 3.640 gm) of acenaphthenequinone was reflux in (150 ml) round bottom flask with a magnetic stirrer for (5-6)h, then the coloured solution has been evaporated to the half and, the resulting precipitate, filtered off and then washed several times with (25 ml) of cold ethanol and (25 ml) of ether. The result ligand was dried for $\approx 24\text{h}$. under vacuum, (Scheme 1).



Scheme (1): Synthesis of Schiff base (L^1 and L^2)

2.3. Preparation of the ligands L^3 and L^4

(100 ml) ethanolic mixture of (0.02 mole, 2.44 gm) of 4-methyl-o-phenylenediamine or (0.02 mole, 3.06 gm) of 4-nitro-o-phenylenediamine and (0.01 mole, 1.82 gm) of acenaphthenequinone was reflux in (150 ml) round bottom flask, then complete the reaction steps as mentioned above, (Scheme 2).



Scheme (2): Synthesis of Schiff bases (L^3 and L^4)

2.4. Method of preparation the complexes:

$[\text{ML}^n\text{Cl}_2]$, $\text{M}=\text{Co}(\text{II}), \text{Ni}(\text{II}), n=(1-4)$:

Methanolic solution of (0.01 mole) of metal chloride [$\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ (92.34 gm), $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$ (2.37 gm)] was gradually added to the solution of the ligand which obtained by dissolving (0.01 mole) of: [L^1 (4.51 gm), L^2 (4.81 gm), L^3 (2.37 gm) and L^4 (4.52 gm)] in 100 ml of methanol in (250 ml) round bottom flask. The reaction mixture has been refluxed $\approx (5-6)$ h. then evaporated to half volume, the resulting coloured was cooled, filtered off and washed carefully with cold methanol, ether and then it was dried under vacuum.

2.5. Antibacterial studies

Evaluation of the antibacterial activity for the ligands and their complexes were tested against Gram(+) bacteria (*Staphylococcus aureus* and *Staphylococcus epidermidis*), and Gram(-) bacteria (*Pseudomonas aeruginosa* and *Escherichia coli*) by agar diffusion method [16,17]. The test solution were prepared by dissolving (10 gm) of each tested substance in (1 ml) of DMSO. A (6 mm) diameter blank paper disc were cultivated of ($37\text{ }^\circ\text{C}$) for (24 h), the diameter of the inhibition zone (mm) was determined and the activity was calculated. Standard disk of (Amoxicillin) used as positive control.

3. Result and discussion:

The suggested structure of complexes resulting from direct simple reaction of Schiff bases ligands are shown at (Fig.1). The physical properties of ligands and complexes are tabulated at (Table 1). All prepared complexes are stable in dry air and contain five membered chelate rings, which are often more stable, soluble in most organic solvents, and their melting points at ($195-320^\circ\text{C}$).

Table 1: The physical properties and analytical of the ligands and their complexes.

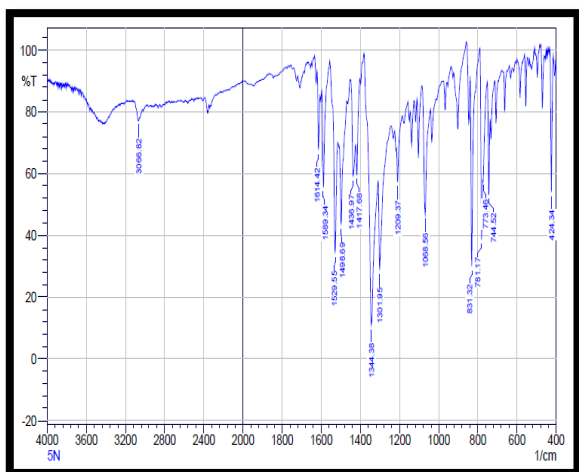
No.	Compound	Colour	m.p.(oC)	AM(ohm ⁻¹ .cm ² .mol ⁻¹)	M% Cal(found)	Analysis calc.(found)%		
						C%	H%	N%
L ¹	C ₃₁ H ₁₉ N ₂ O ₂	Pall yellow	195	----	----	----	----	----
1	[CoL ¹ Cl ₂]	Gray	250	20.3	10.13 (9.50)	64.05 (65.32)	3.29 (3.84)	4.82 (4.90)
2	[NiL ¹ Cl ₂]	Light yellow	255	17.7	----	----	----	----
L ²	C ₃₀ H ₁₅ N ₃ O ₄	Yellow	310	----	----	74.84 (73.52)	3.14 (2.814)	8.73 (9.32)
3	[CoL ² Cl ₂]	Yellow	255	16.1	9.64 (9.83)	----	----	----
4	[NiL ² Cl ₂]	Dark yellow	230	14.6	----	----	----	----
L ³	C ₂₆ N ₄ H ₂₂	Pall yellow	215	----	----	79.97 (80.59)	5.68 (4.86)	14.35 (13.93)
5	[CoL ³ Cl ₂]	Purple	235	22.1	11.32 (10.96)	----	----	----
6	[NiL ³ Cl ₂]	Light yellow	230	13.1	----	----	----	----
L ⁴	C ₂₄ H ₁₆ N ₆ O ₄	Orange	250	----	----	63.71 64.52	3.56 (4.13)	18.58 (19.34)
7	[CoL ⁴ Cl ₂]	Yellowish orange	315	15.1	10.12 (9.90)	----	----	----
8	[NiL ⁴ Cl ₂]	Dark yellow	320	23.5	----	49.53 (50.231)	2.77 (3.17)	14.44 (13.899)

3.2. IR Spectrum:

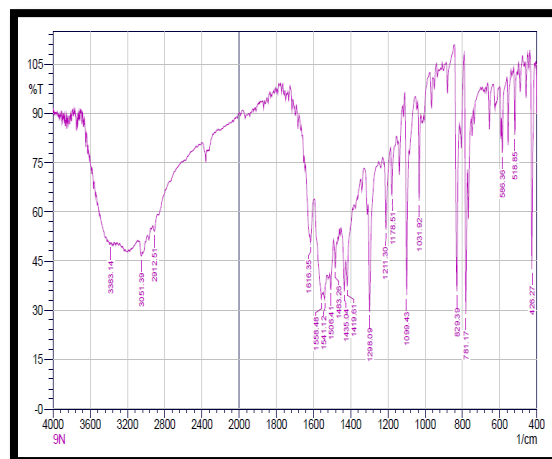
The most important bands of the free ligands and their complexes which are used to demonstrate the structure of the ligands and complexes are tabulated in (Table 2). All ligands show a clear band at (1612-1630) cm⁻¹ due to $\nu(\text{C}=\text{N})$ of azomethine group and this band confirm formation the ligands from phenylenediamine and acenaphthenequinone. In all complexes this band has been shifted towards a lower frequencies and observed at (1590-1616) cm⁻¹ due to formation of the complexes. Another important band appeared at (1720 and 1730) cm⁻¹ due to $\nu(\text{C}=\text{O})$ of terminal carbonyl group in L¹ and L² respectively [21,22], this band has been shifted towards a lower frequencies in complexes(1-4), indicating the coordination of metal ion with oxygen atom of carbonyl group. IR spectra for L³ and L⁴ show broad band at (3410 and 3455) cm⁻¹ respectively due to $\nu(\text{NH}_2)$ [23], in spectra of the complexes (5-8) this band has been shifted towards a lower frequencies indicating that the (NH₂) group shared in coordination [24] on the other hand; new band were appeared at (510-555) cm⁻¹ in the spectrum of complexes (1-4) due to $\nu(\text{M}-\text{O})$ in these complexes, while another new band in the spectrum of all complexes appeared at (435-470) cm⁻¹ due to $\nu(\text{M}-\text{N})$ stretching vibration [25,26]. All complexes not registered band for $\nu(\text{M}-\text{Cl})$ because this band is below to the spectrophotometer limits. The IR. spectrum for the ligands and complexes have been shown in (Fig.3).

Table 2: Characteristic IR spectral data in (cm⁻¹) of ligands and complexes.

No.	$\nu(\text{C}=\text{N})$	$\nu(\text{C}=\text{O})$	$\nu(\text{NH}_2)$	$\nu(\text{M}-\text{N})$	$\nu(\text{M}-\text{O})$
L ¹	1612	17200	----	----	----
1	1590	1702	----	445	510
2	1602	1700	----	450	518
L ²	1614	1730	----	----	----
3	1602	1708	----	465	555
4	1600	1715	----	460	528
L ³	1630	----	3410	----	----
5	1616	----	3383	470	----
6	1612	----	3380	435	----
L ⁴	1630	----	3455	----	----
7	1614	----	3400	450	----
8	1610	----	3390	445	----

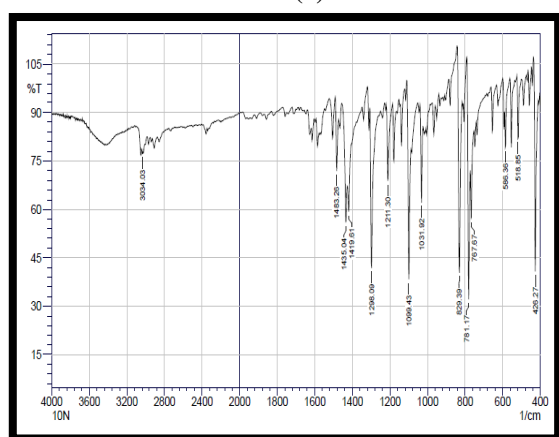


(a)

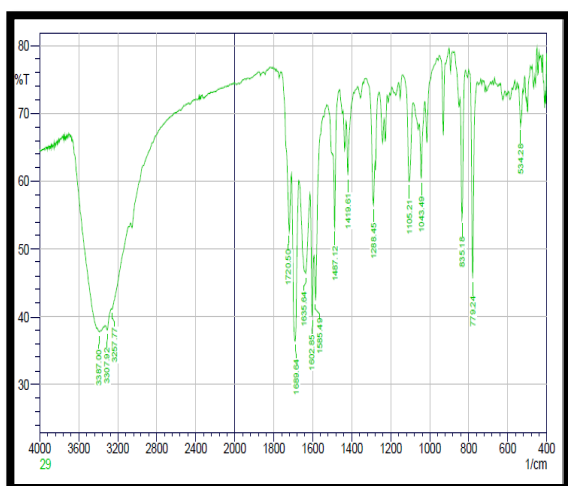


(d)

Fig (3): IR-Spectra of (a) L^2 ; (b) complex 3, (c) L^3 and (d) complex 5, (e) L^4 and (f) complex 7



(b)



(c)

3.3. Electronic spectrum and magnetism:

Electronic spectrum and magnetism values are often very helpful in the evaluation of structural investigation (Table 3) has listed down the electronic spectrum transition and magnetic moments data for the compounds at 25°C. The spectrum of the ligands were characterized by strong bands at (37313 -38461) cm^{-1} and (29411-30487) cm^{-1} assigned to $\pi \rightarrow \pi^*$ and $n \rightarrow \pi^*$ transition respectively of the non-bonding electrons which present at SP^2 hybrid orbital on the nitrogen atom of (C=N) group, these bands are also found in spectrum for the complexes but they are shifted to confirm the coordination between the ligand and the central ion [27].

Band of the charge transfer transition appeared as a new band in spectra of all complexes at (25445-27700) cm^{-1} [28].

Co(II) complexes (1,3,5,7) show two absorption bands at (13333-15873) cm^{-1} and (18181-20833) cm^{-1} assigned to $\nu_2(^4T_{1g(F)} \rightarrow ^4A_{2g(F)})$ and $\nu_3(^4T_{1g(F)} \rightarrow ^4T_{1g(P)})$ transitions and the magnetic moment values for these complexes at (4.78-5.08) B.M, these observation indicates the Co(II) has an (octahedral) environment [29]. The electronic spectrum of Ni(II) complexes (2,4,6,8), exhibited two bands at (16666-19607) cm^{-1} and (22831-26315) cm^{-1} assigned to $\nu_2(^3A_{2g(F)} \rightarrow ^3T_{1g(F)})$ and $\nu_3(^3A_{2g(F)} \rightarrow ^3T_{1g(P)})$ transitions respectively and the obtained magnetic moment values for the same complexes has been found refers to the range (2.93-3.13) B.M, which indicating an octahedral geometry around the (Ni(II) ions [30,31].

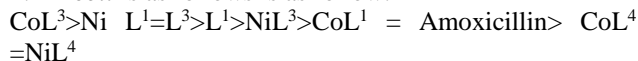
Table 3: Magnetic moments and electronic spectral values for the compounds

No.	μ_{eff} (B.M)	Assignment	Band maxima λ (cm ⁻¹)	Structure
L ¹	----	$\rightarrow n \rightarrow * \pi, \pi \quad * \pi$	30303, 38461	----
1	5.08	$\rightarrow \overset{4}{T}_{1g(F)} \quad \overset{4}{A}_{2g(F)}$ $\rightarrow \overset{4}{T}_{1g(F)} \quad \overset{4}{T}_{1g(P)}, C.T$	14285, 20833, 26315	Oh
2	2.93	$\rightarrow \overset{3}{A}_{2g(F)} \quad \overset{3}{T}_{1g(F)}$ $\rightarrow \overset{3}{A}_{2g(F)} \quad \overset{3}{T}_{1g(P)}, C.T$	17241, 23809, 27329	Oh
L ²	----	$\rightarrow n \rightarrow * \pi, \pi \quad * \pi$	30487, 37313	----
3	4.78	$\rightarrow \overset{4}{T}_{1g(F)} \quad \overset{4}{A}_{2g(F)}$ $\rightarrow \overset{4}{T}_{1g(F)} \quad \overset{4}{T}_{1g(P)}, C.T$	13333, 18181, 25681	Oh
4	3.03	$\rightarrow \overset{3}{A}_{2g(F)} \quad \overset{3}{T}_{1g(F)}$ $\rightarrow \overset{3}{A}_{2g(F)} \quad \overset{3}{T}_{1g(P)}, C.T$	19607, 22831, 25972	Oh
L ³	----	$\rightarrow n \rightarrow * \pi, \pi \quad * \pi$	29411, 37735	----
5	4.88	$\rightarrow \overset{4}{T}_{1g(F)} \quad \overset{4}{A}_{2g(F)}$ $\rightarrow \overset{4}{T}_{1g(F)} \quad \overset{4}{T}_{1g(P)}, C.T$	14705, 20000, 27173	Oh
6	3.03	$\rightarrow \overset{3}{A}_{2g(F)} \quad \overset{3}{T}_{1g(F)}$ $\rightarrow \overset{3}{A}_{2g(F)} \quad \overset{3}{T}_{1g(P)}, C.T$	16666, 25000, 27700	Oh
L ⁴	----	$\rightarrow n \rightarrow * \pi, \pi \quad * \pi$	30303, 38461	----
7	4.78	$\rightarrow \overset{4}{T}_{1g(F)} \quad \overset{4}{A}_{2g(F)}$ $\rightarrow \overset{4}{T}_{1g(F)} \quad \overset{4}{T}_{1g(P)}, C.T$	15873, 19230, 25445	Oh
8	3.13	$\rightarrow \overset{3}{A}_{2g(F)} \quad \overset{3}{T}_{1g(F)}$ $\rightarrow \overset{3}{A}_{2g(F)} \quad \overset{3}{T}_{1g(P)}, C.T$	18867, 26315, 26178	Oh

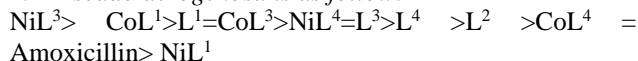
3.4. Antibacterial activity:

Evaluation of biological activity for the ligands and complexes was studied against selected types of G(-) bacteria (*Escherichia coli*, *Pseudo aerogenosa*) and G(+) (*Staphylococcus aureus*, *staphylococcus epidermidis*). The obtained result have been tabulated and shown in (Table 4) and (Fig.4) and compared with standard drug (Amoxicillin), the result evidently show that the ligands and most of the complexes possess different activity against each of above mentioned bacteria, generally the activity order for the prepared compounds against:

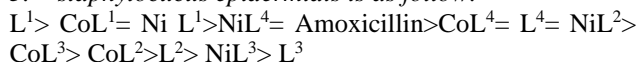
1. *Ecoli* is as follows is as follow:



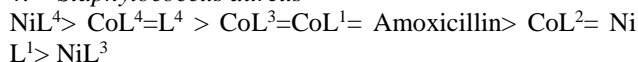
2. *Pseado aerogenosa* is as follow:

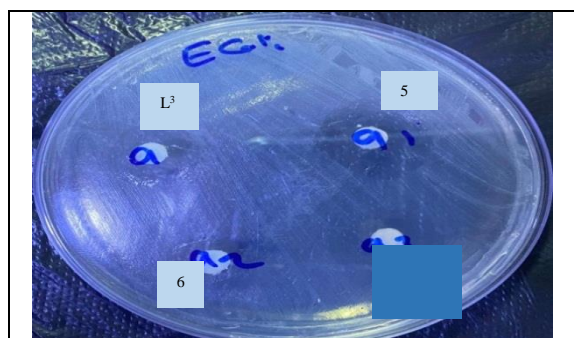


3. *staphylococcus epidermidis* is as follow:

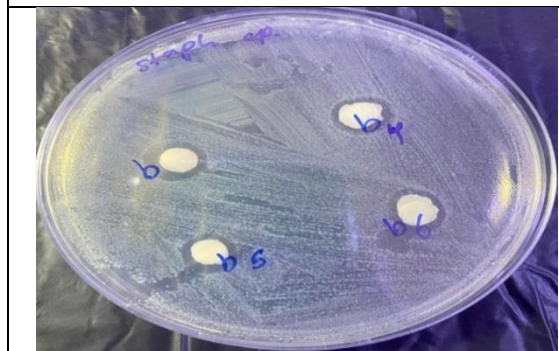


4. *Staphylococcus aureus*

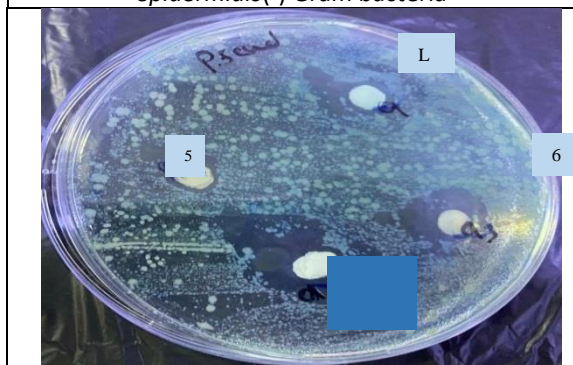




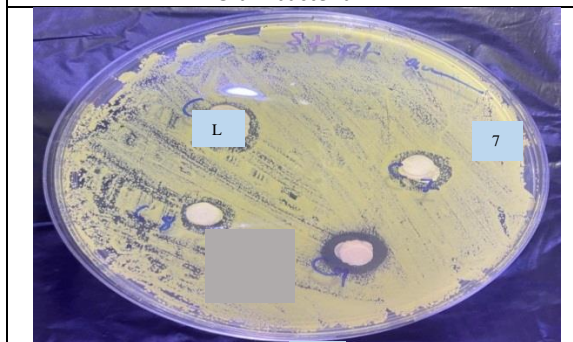
Activity of L³ and complexes (5-6) against *E. coli*(-) Gram bacteria



Activity of L¹ and complexes (1-2) against *staph epidermidis*(-) Gram bacteria



Activity of L³ and complexes (5,6) against *Pseudo*(-) Gram bacteria



Activity of L⁴ and complexes (7-8) against *staph aureus*(-) Gram bacteria

Table 4: The antibacterial activity of ligands and their complexes

4: The antibacterial activity of ligands and their complexes

No.	Compound	Gram (-) bacteria		Gram (+) bacteria	
		<i>E. coli</i>	<i>Pseudo aerogenosa</i>	<i>staphylococcus epidermidis</i>	<i>Staphylococcus aureus</i>
---	L ¹	16	13	13	0
1	[Co L ¹ Cl ₂]	12	15	11	10
2	[Ni L ¹ Cl ₂]	17	2	11	9
---	L ²	0	10	5	0
3	[Co L ² Cl ₂]	0	0	6	9
4	[Ni L ² Cl ₂]	0	13	9	0
---	L ³	17	12	3	0
5	[Co L ³ Cl ₂]	20	13	8	10
6	[Ni L ³ Cl ₂]	14	18	5	3
---	L ⁴	0	11	9	11
7	[Co L ⁴ Cl ₂]	2	8	9	11
8	[Ni L ⁴ Cl ₂]	2	12	10	12

3.5. Density functional theory (DFT) calculation:

Calculation has been carried out with Gaussian 03 program [32]. This computational strategy is used successfully on organic and coordination compounds. Gauss view 4.1 is used to draw the structures, the ligands and their complexes are optimized using B3LYP level with basis set for metal atoms and 6-311G+(d,p) for C,N,O and H atoms[33].

The chemical potential (μ) is defined by DFT as the following equation [34].

$$\mu = \left(\frac{\partial E}{\partial N} \right) \vartheta(r)$$

E=energy; N=number of electron of the system at external pressure $\vartheta(r)$ and chemical hardness (n) is defined as the following equation [34]:

$$n = \frac{1}{2} \left(\frac{\partial \mu}{\partial N} \right) \vartheta(r)$$

But both (μ and n) can be calculated in terms ionized potential (IP) and electron affinity (E) with the following equations:

$$\mu = - \left(\frac{IP + EA}{2} \right) \text{ and } n = \left(\frac{IP - EA}{2} \right)$$

Koopman principle is explained the (IP) and (EA) are related to energies of highest occupied molecular orbital (HOMO) and the lowest unoccupied orbital (LUMO)[35], So-IP= -E_{HOMO}; EA= - E_{LUMO}

$$\mu = \left(\frac{E_{LUMO} - E_{HOMO}}{2} \right) \text{ and } n = \left(\frac{E_{LUMO} + E_{HOMO}}{2} \right)$$

Parr and co-workers propose (ω) as a measure of electrophilicity power [34] and can be written as $\omega = \frac{\mu^2}{2n}$

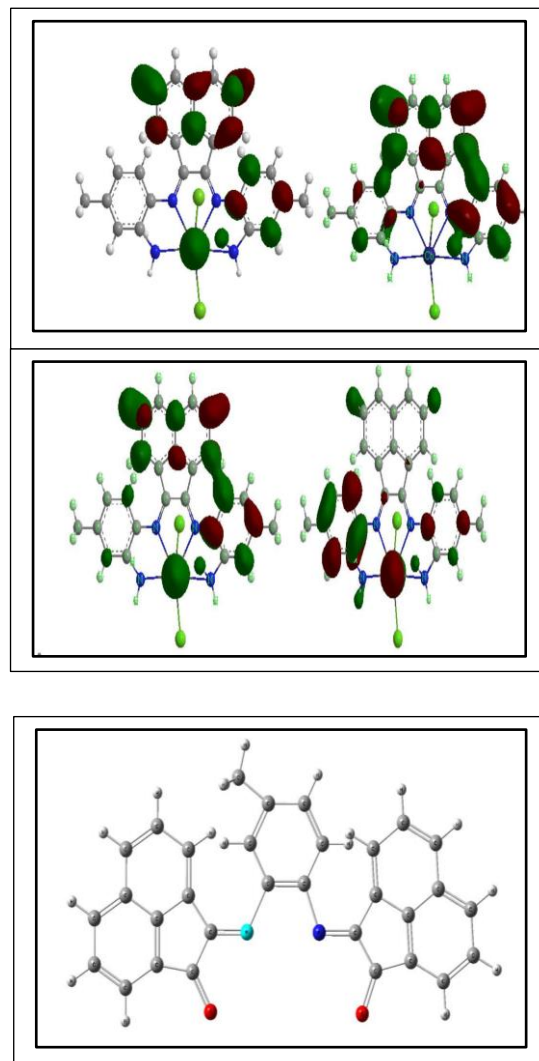
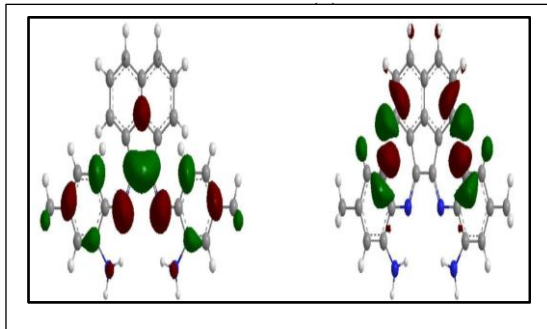
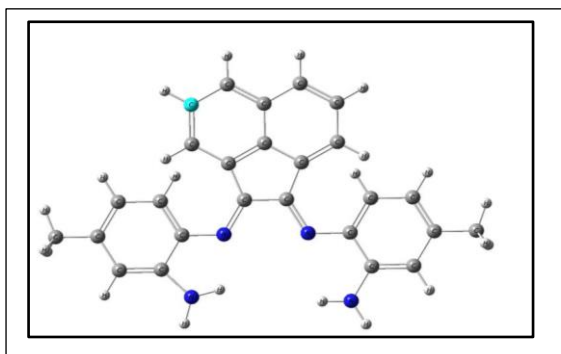
Analysing the frontier molecular orbitals such as (HOMO) and (LUMO) is very important for understanding the most important properties of the molecules such as the chemical reactivity, chemical, hardness, softness and kinetic stability [36]. The reactivity of the compound can be determined by considering the minimum electrophilicity principle, when the compound has a minimum electrophilicity it has a maximum stability. The optimized structure of L^1 and L^3 and their complexes shown in (Fig.5) and values of (μ, η, ω) which calculated from (HOMO) and (LUMO) values are tabulated in (Table 5), generally the reactivity order of L^1, L^2 and their preparation is as follows: $L^1Co > L^1 > NiL^1$; $L^3 > NiL^3 = L^3Co$

4. Conclusion:

Co(II) and Ni(II) complexes with N_2O_2 and N_4 donor Schiff base ligand have been synthesized and characterized by different physicochemical techniques. The data revealed that all the complexes in this work has been classified as mononuclear with octahedral structure. The ligands and their complexes were examined against Gram (+) and Gram (-) bacteria, the test result were compared with standard drug (Amoxicillin).

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We thank Dep. of Chemistry, College of Science, University of Mosul, and Al-Rabeeh Laboratory for supporting this work.



(b)

Fig.5: Optimize structure for (a) L^1 and (b) L^2 and limit molecular orbital (HOMO) and (LUMO) for L^3 and their complexes

Table 5: Calculate of some quantum chemical parameters of L^1 (a) and L^3 (b)

Compound	HOMO	LUMO	η	μ	W
Ligand	-9.475	-9.348	0.0635	-9.4115	2.812299
L1Co	-7.827	-7.545	0.141	-7.686	4.164759
L1Ni	-8.236	-8.2	0.018	-8.218	0.60782

(a)

Compounds	HOMO	LUMO	η	μ	W
L3	-8.483	-7.968	0.2575	-8.2255	8.711077
L3Co	-4.056	-3.253	0.4015	-3.6545	2.681091
L3Ni	-4.072	-3.268	0.402	-3.670	2.707249

(b)

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