

Preparation and Characterization of some Transition Metal Complexes with di - 2- Quinolinone Dibenzylidene Ligand and their Adducts with 1,10 – Phenanthroline

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ABSTRACT

The present work includes the preparation with complexes of the general formula $[M_2L_2] Cl_4$ and $[M_2L_2(1,10\text{-phen})_2] Cl_4$ were

M= Mn (II), Fe (II), Co (II), Ni (II), Cu (II) and Zn (II)

L = di -2- quinolinone dibenzylidene

1,10-phen = 1,10 phenanthroline

The ligand was prepared by condensation reaction of N-aminoquinoline-2-one with terphaldehyde (2:1) and characterized by infrared and proton NMR.

The complexes number (1-6) prepared into (2:2) (M: L) molar ratio respectively while complex number (7-12) prepared into (2:2:2) (M: L: 1,10-phen) molar ratio respectively.

The I.R spectra data suggest the involvement of nitrogen atom of azomethine group and oxygen atom of carbonyl group of the ligand in coordination to central metal ion.

The synthesized complexes were characterized by elemental analysis, molar conductance, magnetic susceptibility, and spectral measurements (UV – Vis, IR).

Elemental analysis data proposed 2:2 (metal: ligand) and 2:2:2 (1,10-phen). The molar conductance measurements showed 1:4 electrolytes complexes, Further more magnetic moment measurements and electronic spectra indicated that the complexes of the type $[M_2L_2] Cl_4$ possess a tetrahedral geometry while complexes of the type $[M_2L_2(1,10\text{-phen})_2] Cl_4$ possess an octahedral geometry.

Keywords: di-2-quinolinonedibenzylidene, metal, complexes, 1,10 phenathroline, dinuclear.

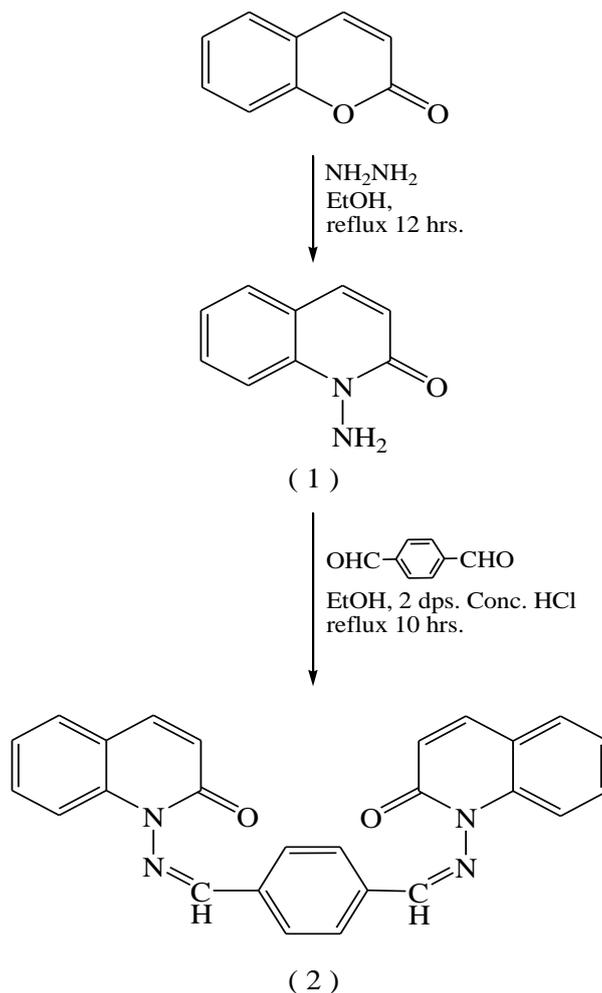
INTRODUCTION

Ligands like Schiff bases with nitrogen or oxygen donor atoms are conceded to be a good category of organic compounds, that have the ability to bind with different metal ions that possess an existing non-medical and medical properties, which was very common in the last few years (Liu *et al.*, 2018; Lashanizadegan *et al.*, 2016). The synthesis of these ligands was conducted easily by the condensation of primary amines with ketones or aldehydes (Gerloch *et al.*, 1968) Schiff base complexes with transition metals received a good attention due to their wide application in different areas since they have diverse role in metalo-organic, inorganic and biochemistry (Ganguly *et al.*, 2008; Golcu *et al.*, 2005). These compounds considered as a chelating ligands in the transition metals coordination chemistry; since they are stable in reductive and oxidative conditions (Al Zoubi and Ko., 2017).

These ligands interact with metal ions to give complexes with different geometries that possess more biological activities (Da Silva *et al.*, 2011) like antibacterial, anticancer, antimalarial, antipyretic, antifungal, anti-inflammatory and antiviral properties (Afradi *et al.*, 2017; Anitha *et al.*, 2012; Raman *et al.*, 2001).

A series of quinolones possess a wide spectrum of activity and bio availability (Zhang *et al.*, 2018; Kocsis *et al.*, 2016) which make them good candidates for the treatment of infectious diseases with different locations like: urinary, gastrointestinal, cutaneous, respiratory, and bone (Dhiman *et al.*, 2019). The antibacterial mechanism of quinolones came from its ability to inhibit DNA replication and transcription in bacteria (Nagaraja *et al.*, 2017; Charrier, 2017). A new trend in complex combination field of quinolones was represented by its chelation with lanthanide metal ion (Ln^{+3}) (Măciucă *et al.*, 2020). Quinolone is a heterocyclic aromatic compound which is considered to be a cyclic amide that incorporated with the structure of different pharmaceuticals (Gümüş *et al.*, 2010). Derivatives of 2-quinolone have a different biological activities like anti-depressant (Oshiro *et al.*, 2000) antimalarial (Xiao *et al.*, 2001) antiulcer (Banno *et al.*, 1988) and antioxidant activity (Al-Omar *et al.*, 2006). Furthermore, quinoline-2-one derivatives were found to be of great importance in chemotherapy as antitumor drugs (Abhishek *et al.*, 2014). Synthesis of ligands that contains different donors is increased continuously and their importance increased when they possess biological activity (Govind and Rajesh, 1992; Kong *et al.*, 2003). Quinolones studies revealed a metal-drug interaction. Ciprofloxacin, for instance, showed a decrease in absorption when co administered with magnesium, aluminum antacid (Reese *et al.*, 2000). A dinuclear complexes for Cu (II), Ni (II), Co (II) and Zn (II) were prepared, and their antifungal and antibacterial properties against *Escherichia coli* were tested (Mruthyunjayaswamy *et al.*, 2005). Furthermore the dinuclear Zn (II) complexes revealed higher activity against bacterial strains (Krstic *et al.*, 2019). In addition to the above observations, dinuclear complexes of platinum and copper showed an anticancer activity (Broomhead *et al.*, 1993; Ahmad *et al.*, 2021). Finally, complexes of Cu (II) and azomethine were screened for their fungicidal, antibacterial and protistocidal activities against *colpoda steinii*, *Escherichia coli* 078, *Staphylococcus aureus* P-209 and *Penicillium italicum* (Vlasenko *et al.*, 2021).

In the present work, N-aminoquinoline-2-one has been synthesized through the reaction of coumarone and hydrazine hydrate. This compound was allowed to react with terphthalaldehyde in order to produce the ligand that reacted with different metal salts to give the dinuclear complexes General Scheme.



General Scheme : Synthesis of di-2-quinolinone dibenzylidene (2)

EXPERIMENTAL

Instrumentation

All chemicals used were reagent grade from B.D.H. of Fluka accompanies, used as supplied, Melting point were determined on a Stuart SMP30 advanced Digital Melting Point apparatus (UK), and they were uncorrected.

Infrared spectra were recorded on Shimadzu as KBr disk ($400-4000\text{ cm}^{-1}$) and Bruker Alpha FT-IR spectrophotometer (Germany).

The UV – Vis spectra (200 – 1100 nm) were recorded on a Shimadzu UV-1650 pc spectrophotometer using DMF as solvent.

Molar conductance was measured at room temperature using laboratory conductivity meter con 720 WTW/Germany (2004).

The magnetic susceptibilities were measured at $25\text{ }^{\circ}\text{C}$ on the solid by Faraday method using Bruker BM6 instrument. Diamagnetic corrections were calculated using Pascal's constants.

Proton NMR spectra were recorded on Bruker Biospin 400 MHz, Germany, using TMS as internal reference, and DMSO - d_6 as a solvent.

The C.H.N. elemental analyses were performed on euro EA 300 Instrument, The metal content of complexes were determined spectrophotometrically [Mn (II), Fe (II), Co (II) and Cu (II)] using Shimadzu AA670 atomic absorption spectrophotometer. zinc and nickel were determined volumetrically using standard EDTA and murexide as indicator.

Synthesis of compound

1. Synthesis of N-amino quinoline-2-one (1): (Al-Bayati and Radi, 2010).

A mixture of coumarin (1.46g,0.01mol) and excess of hydrazine hydrate (99%) (3.2g,0.1mol) in absolute ethanol(25ml) was refluxed for 12 hrs., the mixture was then cooled and solid that formed was collected and recrystallized from chloroform, m.p (131-133 °C).

2. Synthesis of ligand (di-2-quinolinone dibenzylidene) (2):

Terphthaldehyde (0.96g,0.01mol) was dissolved in (10ml) of absolute ethanol, (1-2 drops) of concentrated HCl was added to the solution. N-amino quinoline 2-one (3.2g, 0.02mol) was dissolved in (10-15ml) of absolute ethanol and this was added to the latter solution. The mixture refluxed for (9-10) hrs., and the orange precipitate was filtered and washed then dried under vacuum.

3. Preparation of complexes [M₂L₂] Cl₄:

(1.672g,0.002mol) of ligand dissolved in (20ml) of absolute ethanol, then added to (0.002mol) of metal salt (MnCl₂.5H₂O, FeCl₂.4H₂O, CoCl₂.6H₂O, NiCl₂.6H₂O, CuCl₂.2H₂O, ZnCl₂). The mixture was heated under reflux for 6 hrs. The precipitates were filtered and dried under vacuum.

4. Preparation of adducts [M₂L₂(1,10-phen)₂] Cl₄:

(0.36g,0.002mol) of 1,10-phenanthroline was added to a solution consist of (1.672g, 0.002mol) of ligand dissolved in (20ml) of absolute ethanol and (0.002 mol) of metal salts. The mixture refluxed for 6 hrs., filtered and the precipitate dried under vacuum.

RESULTS AND DISCUSSION

Elemental analysis

The experimental percentage and the calculated one for C.H.N. and M% for the prepared compounds were listed in (Table 2). We also recorded colors, melting points, percentage yield and molar conductivity in (Table 1).

Molar conductivity

Molar conductivity measurements are useful in the investigation of the existence of moving electrolytic species (ions) in the complex's solutions in DMF at room temperature (25-30C⁰). Table (1) showed that the metal complexes possess high molar conductivity values in the range (242.5-336.5) ohm⁻¹.cm².mol⁻¹ which indicating 1:4 electrolytic nature of the inorganic compound (Geary,1971).

Table 1: Physical data for ligand and it's complexes

No.	Formula	Color	m.p. °C or Decomposition	Yield %	Λ _M (DMF)
L	(L) C ₂₆ H ₁₈ N ₄ O ₂	Dark orange	*280	74.4	
1.	[Mn ₂ L ₂]Cl ₄	Orange	*270	58.63	333
2.	[Fe ₂ L ₂]Cl ₄	Dark red	> 300	60.5	287
3.	[Co ₂ L ₂]Cl ₄	Green	*265	76.6	287.5
4.	[Ni ₂ L ₂]Cl ₄	Dark yellow mustard	> 300	57.4	330.5
5.	[Cu ₂ L ₂]Cl ₄	Dark brown	*262	70.4	296.5
6.	[Zn ₂ L ₂]Cl ₄	Dark orange	> 300	63.8	242.5
7.	[Mn ₂ L ₂ (1,10-phen) ₂]Cl ₄	Orange	*250	59.5	286.5
8.	[Fe ₂ L ₂ (1,10-phen) ₂]Cl ₄	Dark red	> 300	70.3	255.5
9.	[Co ₂ L ₂ (1,10-phen) ₂]Cl ₄	Green	*265	63.8	252.5
10.	[Ni ₂ L ₂ (1,10-phen) ₂]Cl ₄	Orange	> 300	59.5	336.5
11.	[Cu ₂ L ₂ (1,10-phen) ₂]Cl ₄	Green	*245	78	328.5
12.	[Zn ₂ L ₂ (1,10-phen) ₂]Cl ₄	Dark orange	> 300	53.1	281.8

* = decomposition

Table 2: Elemental micro analysis % for ligand and it's complexes

No.	C calculated (found)	H calculated (found)	N calculated (found)	M calculated (found)
L	74.64 (74.241)	4.30 (4.19)	13.39 (13.70)	----
1.	57.3 (57.00)	3.30 (2.70)	10.2 (9.90)	10.11 (9.40)
2.	57.2 (56.80)	3.30 (2.70)	10.2 (10.00)	10.2 (9.70)
3.	56.9 (56.50)	3.28 (3.80)	10.21 (10.20)	10.7 (10.00)
4.	56.9 (56.00)	3.2 (3.00)	10.2 (9.50)	10.7 (10.00)
5.	56.6 (55.50)	3.25 (3.90)	10.13 (10.70)	11.4 (10.80)
6.	56.3 (56.10)	3.24 (3.30)	10.1 (10.00)	11.7 (11.00)
7.	43.0 (43.10)	2.48 (1.80)	7.73 (6.80)	7.5 (7.10)
8.	43.0 (43.50)	2.48 (2.80)	7.72 (7.60)	7.72 (6.80)
9.	42.8 (41.50)	2.47 (2.00)	7.69 (6.00)	8.1 (7.90)
10.	42.8 (41.90)	2.47 (2.60)	7.69 (7.66)	8.0 (7.10)
11.	42.8 (41.90)	2.47 (3.00)	7.69 (7.66)	8.66 (8.40)
12.	42.5 (41.70)	2.45 (1.80)	7.62 (7.00)	8.89 (8.70)

Electronic spectra and magnetic moment

The electronic spectra of the ligand showed band at (28248 cm^{-1}) which assigned to $n \rightarrow \pi^*$ transition, and (37878 cm^{-1}) for $\pi \rightarrow \pi^*$ transition. These bands were shifted to lower wave length in complexes at the regions ($23148\text{-}27624\text{ cm}^{-1}$) and ($32654\text{-}36593\text{ cm}^{-1}$) respectively suggesting the complexation of ligand with the metal. A new strong band for all complexes (1-12) appeared at the region ($26315 - 31645\text{ cm}^{-1}$) that related to the charge transfer.

Mn (II) complex (1) was paramagnetic and gave magnetic moment (5.5 B.M) which in correspondence with five unpaired electron. No band in electronic spectra for Mn (II) shown due to orbital forbidden and spin forbidden. Thus, magnetic moment suggests a tetrahedral structure (Lever,1984). While complex (7) gave magnetic moment (5 B.M) and electronic spectra of Mn (II) in octahedral geometry (Cotton,1999). (High-spin) are spin forbidden and orbital forbidden.

The magnetic moment of Fe (II) complex (2) was (4.7 B.M) indicative for a tetrahedral geometry and the electronic spectra showed two absorption bands at (11363 and 12345 cm^{-1}) respectively which correspond to ${}^5E \rightarrow {}^5T_2$ transition in a tetrahedral geometry (Nicholls, 2013), while complex (8) of Fe (II) gave magnetic moment (4.9 B.M) and showed broad absorption band at (21459 cm^{-1}) which were assigned to $5T_2g(D) \rightarrow 5Eg(D)$ in octahedral (high-spin) geometry (Guzzi and El Alagi,2013)

The magnetic moment of Co(II) complex (3) was (4.20 B.M) and the electronic spectra showed a band at (14836 cm^{-1}) assigned to ${}^4A_2(F) \rightarrow {}^4T_1(P)$ transition, ν_1, ν_2 did not appear to fall outside the range of the device used for measurement these values suggest a tetrahedral geometry (Rakhi and Shelly, 2011; Saad E. Al-Mukhtar, 2017), while in complex (9), the magnetic moment was (4.82 B.M) and electronic spectra of this complex showed two bands at (14970 and 17667 cm^{-1}) which assigned to

${}^4T_{1g}(F) \rightarrow {}^4A_{2g}(F)\nu_2$, ${}^4T_{1g}(F) \rightarrow {}^4T_{1g}(P)\nu_3$ that related to ν_2 and ν_3 respectively, these values suggest (high-spin) octahedral geometry (Sönmez *et al.*, 2003).

The magnetic moment of Ni (II) complex (4) was (3.3 B.M) and the electronic spectra showed band at (13,698 cm^{-1}) assigned to ${}^3T_1(F) \rightarrow {}^3T_1(P)\nu_3$ this values suggests a tetrahedral geometry, while in complex (10) the magnetic moment (3.1 B.M) and the electronic spectra of complex showed three bands at (12468, 14513 and 24752 cm^{-1}) corresponded to three spin allowed transition ${}^3A_{2g} \rightarrow {}^3T_{2g}(\nu_1)$, ${}^3A_{2g}(F) \rightarrow {}^3T_{1g}(F)(\nu_2)$ and ${}^3A_{2g}(F) \rightarrow {}^3T_{1g}(P)(\nu_3)$. This transition suggested octahedral geometry (Seleem *et al.*, 2011).

The magnetic moment of Cu (II) complex (5) has been found (2.0 B.M), and the electronic spectra of this complex showed band at (11574 cm^{-1}) which assigned to ${}^2T_{2g}(D) \rightarrow {}^2E_g(D)$ transition in tetrahedral environment, while the magnetic moment of complex (11) was found (2.0 B.M), and electronic spectra of this complex showed a broad band at (19011 cm^{-1}) assigned to ${}^2B_{1g} \rightarrow {}^2B_{2g}$, because of the influence of Jan Teller, ${}^2B_{1g} \rightarrow {}^2E_g$ transition in distorted octahedral structure around Cu (II) ions (Kulkarni *et al.*, 2012; Mostafa, 2011).

Zn (II) complexes (6,12) were diamagnetic and the electronic spectra gives only charge transfer transition in (26315, 31318 cm^{-1}) respectively (Nabeel and Farah, 2012).

All of the above data were listed in (Table 3).

Table 3: Magnetic susceptibility and electronic spectra of ligand and their complexes

No.	μ_{eff} B.M.	Electronic transition	Absorbtion band cm^{-1}
L	----	$n \rightarrow \pi^*$, $\pi \rightarrow \pi^*$	28248,37878
1.	5.5		31645
2.	4.7	${}^5E \rightarrow {}^5T_2$	11363,12345
3.	4.2	${}^4A_2(F) \rightarrow {}^4T_1(P)$	14836
4.	3.3	${}^3T_1(F) \rightarrow {}^3T_2(P)$	13698
5.	2.0	${}^2T_{2g} \rightarrow {}^2E_g$	11574
6.	----	$n \rightarrow \pi^*$, $\pi \rightarrow \pi^*$	23584,34211
7.	5.0		37593
8.	4.9	${}^5T_{2g} \rightarrow {}^5E_g$	21459
9.	4.82	${}^4T_{1g}(F) \rightarrow {}^4A_{2g}(F)$ ${}^4T_{1g}(F) \rightarrow {}^4T_{1g}(P)$	14970 17667
10.	3.1	${}^3A_{2g} \rightarrow {}^3T_{2g}$ ${}^3A_{2g}(F) \rightarrow {}^3T_{1g}(F)$ ${}^3A_{2g}(F) \rightarrow {}^3T_{1g}(P)$	12468 14513 24752
11.	2.0	${}^2B_{1g} \rightarrow {}^2B_{2g}$ ${}^2B_{1g} \rightarrow {}^2E_g$	19011
12.	----	$n \rightarrow \pi^*$, $\pi \rightarrow \pi^*$	26315, 31318

Infrared spectra

The most important IR absorption bands for the ligand and its complexes are listed in (Table 4). The IR spectra for ligand showed characteristic absorption bands at (1685 cm^{-1}) that assigned for carbonyl group $\nu(\text{C}=\text{O})$, while the azomethine group $\nu(\text{C}=\text{N})$ gave a band at (1602 cm^{-1}), furthermore the $\nu(\text{N}-\text{N})$ group gave a band at (947 cm^{-1}) and finally a band at (1213 cm^{-1}) that attributed to the $\nu(\text{C}-\text{N})$ group. (Parikh, 1974). Fig. (1).

This spectra gave an indication for the formation of the complexes since there is a drop in both of carbonyl and azomethine frequencies which appears at (1600-1620 cm^{-1}) and (1491-1510 cm^{-1}) respectively that confirm complication through both of $\nu(\text{C}=\text{O})$ and $\nu(\text{C}=\text{N})$ (Despaigne *et al.*, 2009; Prasad and Agarwal, 2006).

Furthermore, the $\nu(\text{N}-\text{N})$ absorption frequencies for these complexes appear at higher values (950-958 cm^{-1}) then it was in the ligand and this will give further support for the coordination via azomethine nitrogen atom (Maurya *et al.*, 2002). Figs. (2,3).

Moreover, complexes were also showed a band at (1211-1233) for $\nu(\text{C-N})$ function. It is worth noticing that, the $\nu(\text{M-O})$ and $\nu(\text{M-N})$ vibrational frequencies appear at low values, in which the $\nu(\text{M-O})$ showed absorption bands at (490-526 cm^{-1}) (Agarwal *et al.*, 2006), while the $\nu(\text{M-N})$ band at (432-500 cm^{-1})_for azomethine group (Abd Al-Razaq *et al.*, 2016), all of the above values indicate the formation of a tetrahedral complexes via the coordination of metal with the synthesized tetra dentate ligand, On the other hand complexes (7-12) showed another absorption band at (452-516 cm^{-1}) for $\nu(\text{M-N})$ function confirm the formation of octahedral complexes due to the coordination of the metal with the synthesized tetra dentate ligand and 1,10 phenathroline (Raya *et al.*, 2006). Figs. (4,5).

Table 4: Infrared Spectra for ligand and its complexes

No.	IR ν (cm^{-1})						
	$\nu(\text{C=O})$	$\nu(\text{C=N})$	$\nu(\text{C-N})$	$\nu(\text{N-N})$	$\nu(\text{M-O})$	$\nu(\text{M-N})$ with izomethin	$\nu(\text{M-N})$ with 1,10 phen.
L	1685	1602	1213	947	---	---	---
1.	1618	1510	1211	958	526	440	---
2.	1611	1509	1216	955	520	439	---
3.	1600	1498	1216	953	516	470	---
4.	1607	1499	1233	952	523	432	---
5.	1607	1491	1222	951	518	440	---
6.	1618	1500	1220	950	520	500	---
7.	1620	1500	1211	958	490	454	510
8.	1618	1510	1211	958	526	439	476
9.	1620	1509	1216	955	520	470	516
10.	1608	1497	1215	956	523	424	477
11.	1604	1492	1222	950	518	490	452
12.	1610	1502	1215	951	520	500	470

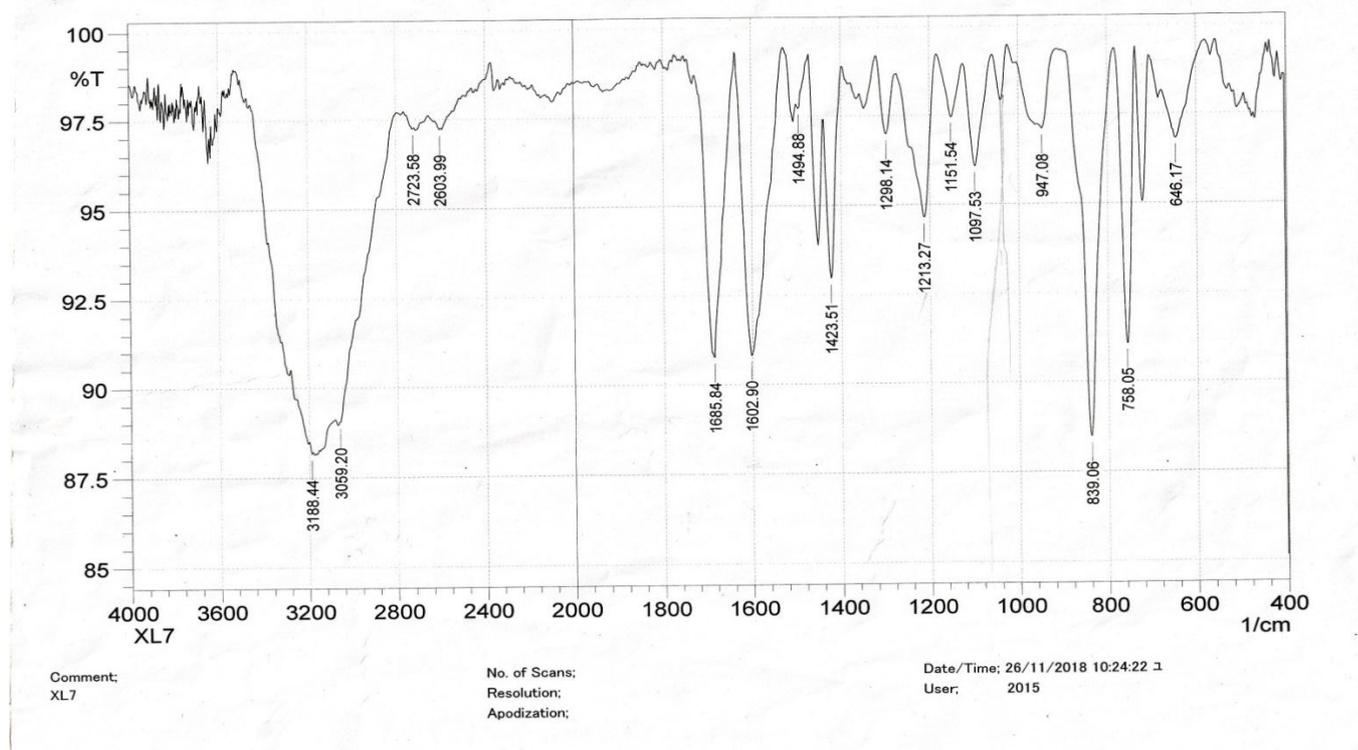


Fig. 1: IR Spectra for ligand

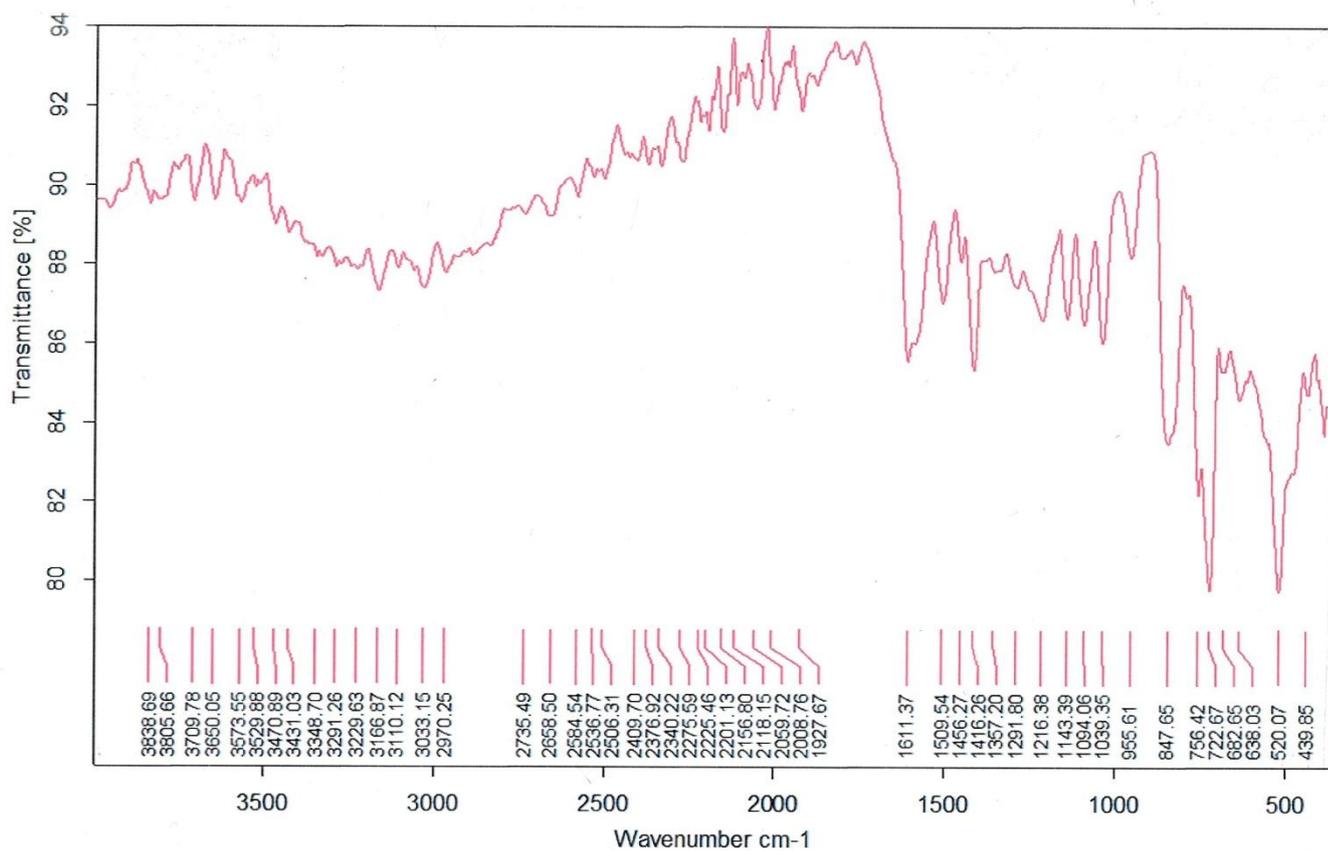


Fig. 2: IR Spectra for complex 2

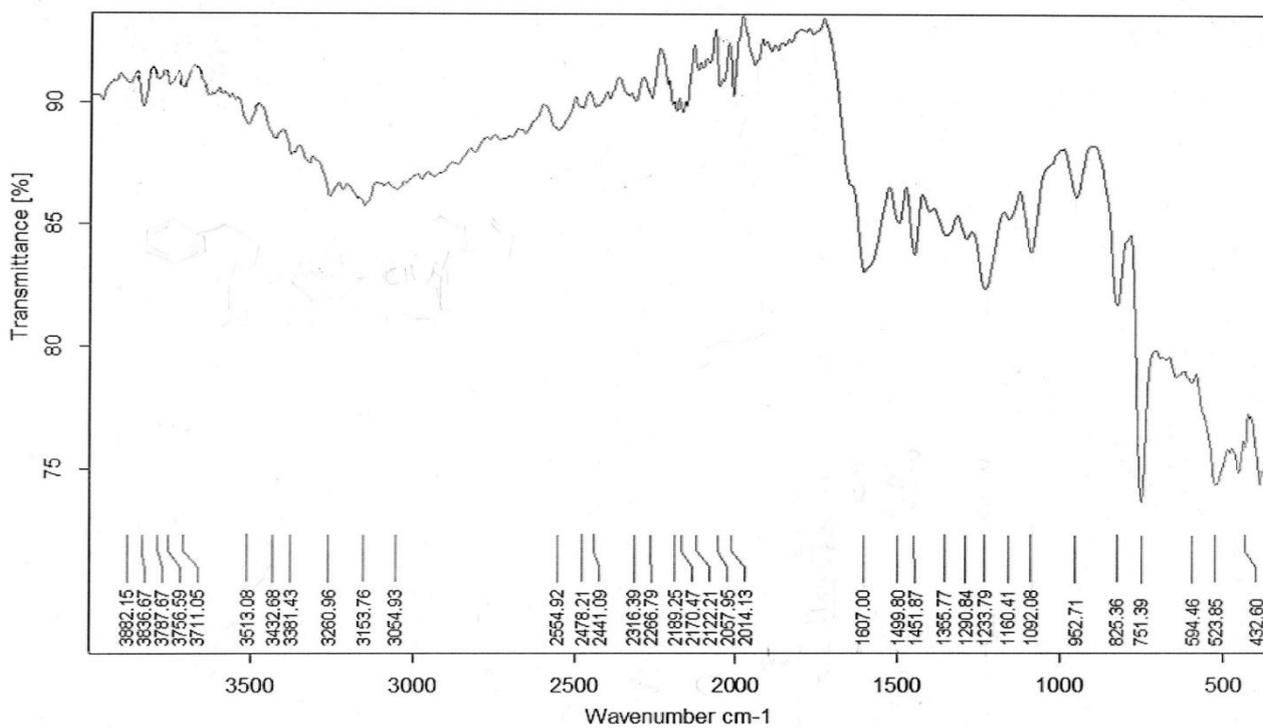


Fig. 3: IR Spectra for complex 4

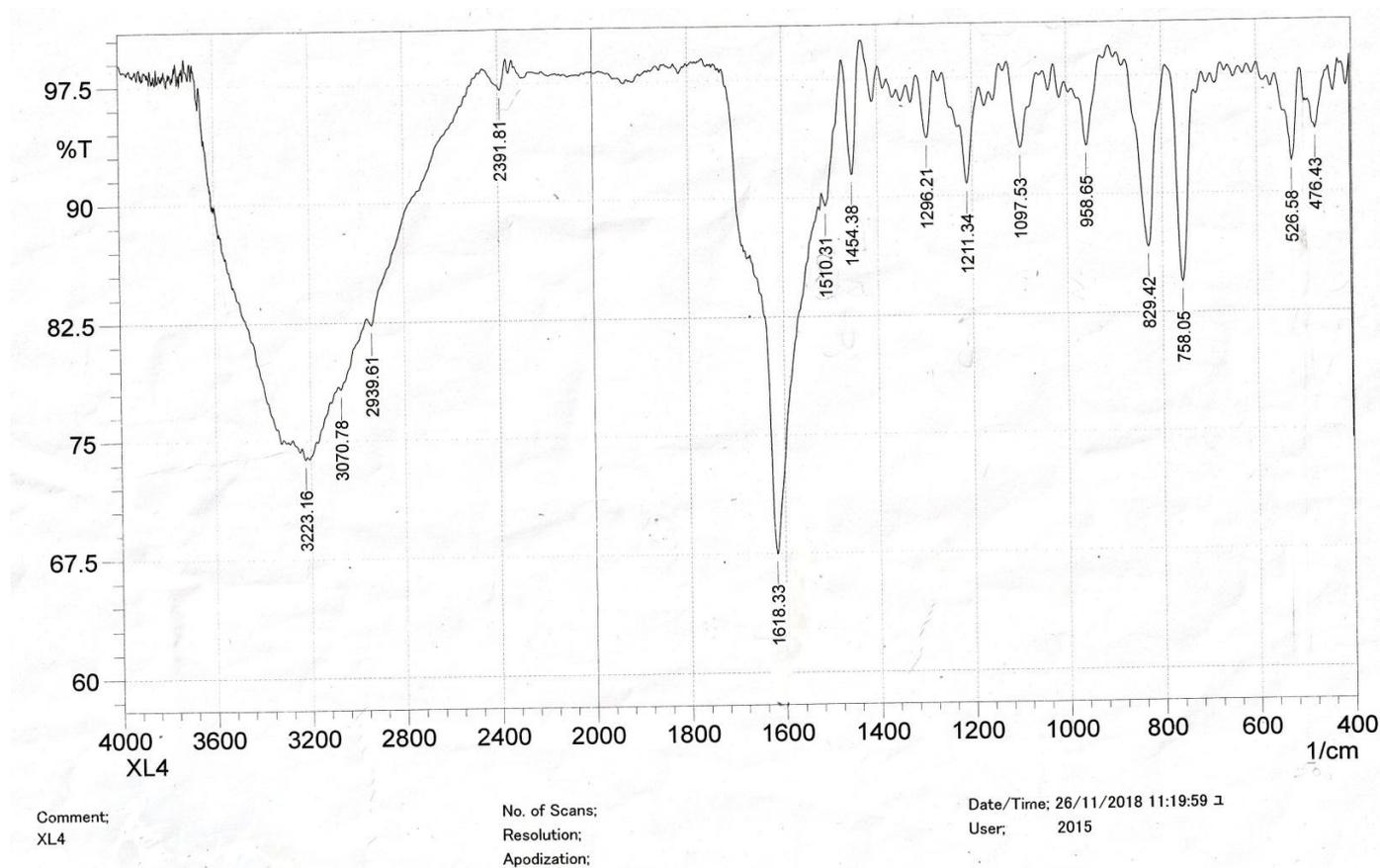


Fig. 4: IR Spectra for complex 8

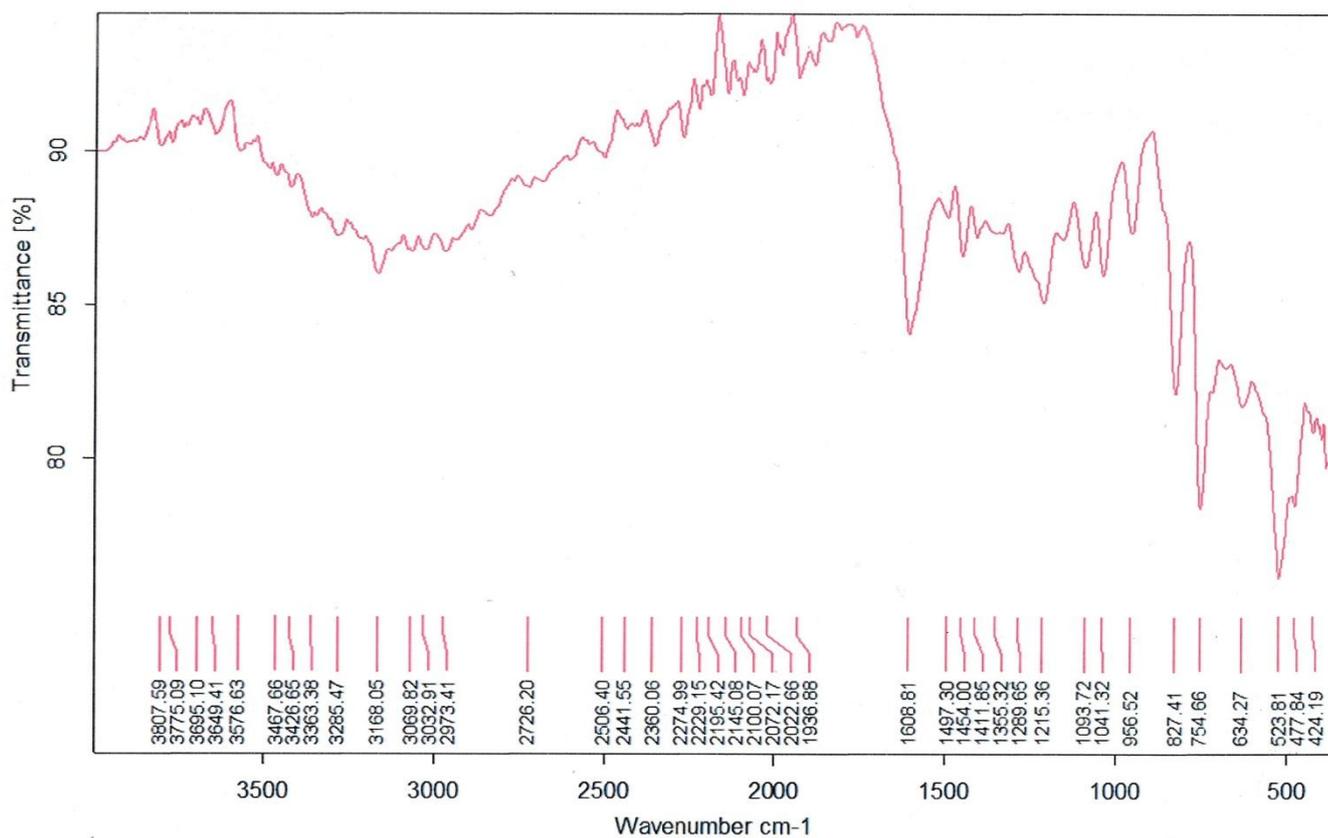


Fig. 5: IR Spectra for complex 10

Proton NMR:

$^1\text{H-NMR}$ spectra were recorded using $\text{d}_6\text{-DMSO}$ as a solvent and TMS as an internal reference. Chemical shifts were reported in (ppm) and coupling constant were given in hertz (Hz). Only $^1\text{H-NMR}$ spectrum for ligand was recorded due to the paramagnetic properties of the metal complexes the spectrum of the ligand showed characteristic bands as the following:

- δ (6.7 – 8.13) ppm (m, 16 H, Aromatic).
- δ (8.16) ppm (s, 2H, azomethine)
- Fig. 6 (a,b)

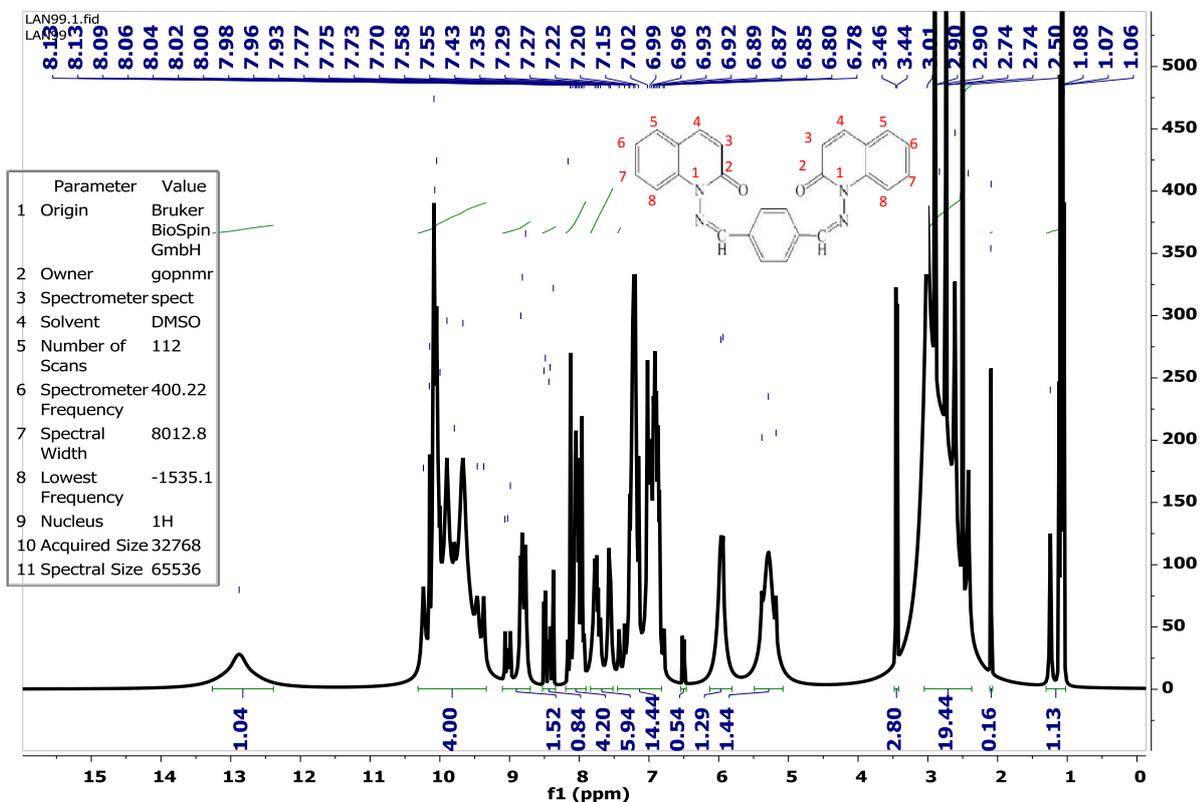


Fig. 6-a

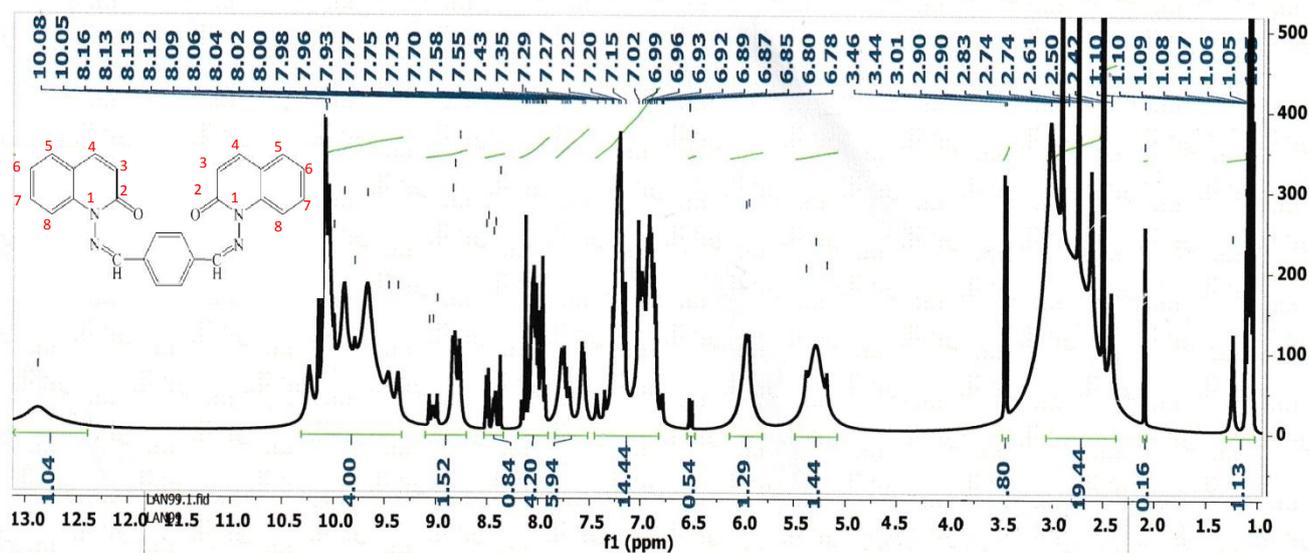


Fig. 6-b

NMR Spectra for ligand (di-2-quinolinone dibenzylidene)

Conclusion Structure of Complexes:

On the basis of the elemental analysis, conductivity measurements, magnetic moment data, spectroscopic studies, the possible structure of complexes: Figs.(7,8)

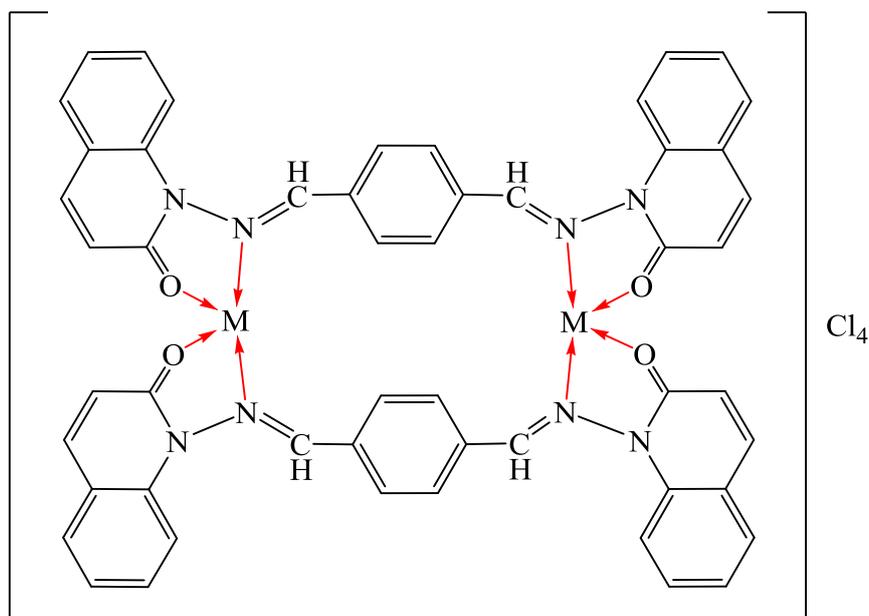


Fig. 7: complexes of the type $[M_2L_2] Cl_4$
M=Mn (II), Fe (II), Co (II), Ni (II), Cu (II), Zn (II)

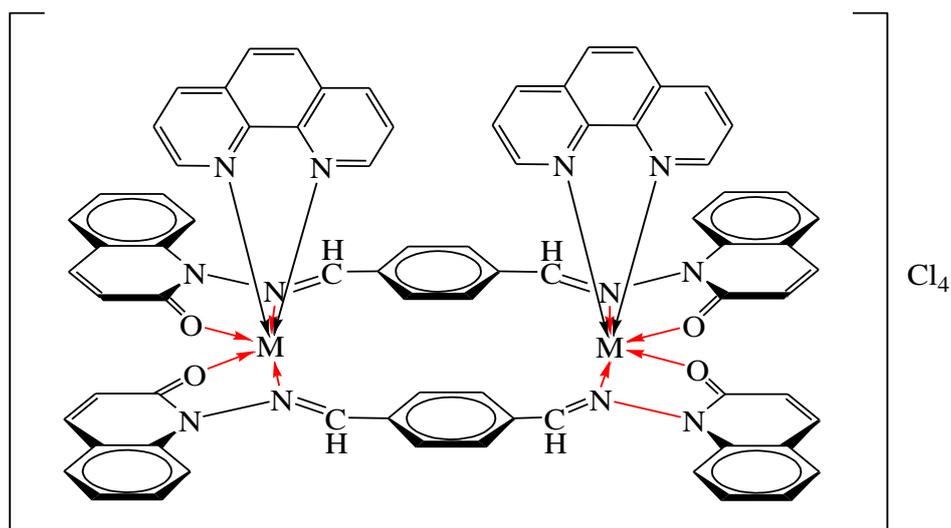


Fig. 8: complexes of the type $[M_2L_2(1,10 phen_2)] Cl_4$
M=Mn (II), Fe (II), Co (II), Ni (II), Cu (II), Zn (II)

REFERENCES

- Abd Al-Razaq, E.; Buttrus, N.; Mohammed, E. H.; Jbarah, A. A. (2016). Synthesis and DFT calculations of dinuclear complex of Co^{2+} , Ni^{2+} and Cu^{2+} with macrocyclic Schiff base Ligands. *Orient. J. Chem.*, **32**(1), 137.
- Abhishek, K.; Fernandes, J.; Pankaj, K. (2014). Synthesis, antimicrobial and anti-inflammatory studies of some novel Schiff base derivatives. *Int. J. Drug Develop. Res.*, **6**, 165-7.

- Afradi, M.; Foroughifar, N.; Pasdar, H.; Moghanian, H.; Foroughifar, N. (2017). Facile green one-pot synthesis of novel thiazolo [3, 2- a] pyrimidine derivatives using Fe₃O₄ 1- arginine and their biological investigation as potent antimicrobial agents. *Appl. Organomet. Chem.*, **31**(9), e3683.
- Agarwal, R. K.; Sharma, D.; Singh, L.; Agarwal, H. (2006). Synthesis, biological, spectral, and thermal investigations of cobalt (II) and nickel (II) complexes of N-isonicotinamido-2', 4'-dichlorobenzalaldimine. *Bioinorg. Chem. and Appl.*, **2006**, Article ID 029234.
- Ahmad, M.S.; Khalid, M.; Khan, M.S.; Shahid, M.; Ahmad, M.; Saeed, H.; Ashafaq, M. (2021). Tuning biological activity in dinuclear Cu (II) complexes derived from pyrazine ligands: Structure, magnetism, catecholase, antimicrobial, antibiofilm, and antibreast cancer activity. *Appl. Organomet. Chem.*, e6221.
- Al-Mukhtar, S.E.; Alkatib, H.F.; Alnuaimy, L.A. (2017). Preparation and Characterization of some transition metal complexes with Crotyl xanthate ligand and their adducts with Nitrogen bases. *Raf. J., Sci.*, **26**(1), 49-55.
- Al Zoubi, W.; Ko, Y.G. (2017). Schiff base complexes and their versatile applications as catalysts in oxidation of organic compounds: part I. *Appl. Organomet. Chem.*, **31**(3), e3574.
- Al-Bayati, R.I.; Radi, M.F. (2010). Synthesis of novel 2-quinolone derivatives. *Afr. J. Pure Appl. Chem.*, **4**(10), 228-232.
- Al-Omar, M.A.; El-Azab, A.S.; El-Obeid, H.A.; Abdel Hamide, S. G. (2006). Synthesis of some new 4-(3H)-quinazoline analogs as potential antioxidant agents. *J. Saudi. Chem. Soc.*, **10**(1), 113-130.
- Anitha, C.; Sheela, C.D.; Tharmaraj, P.; Sumathi, S. (2012). Spectroscopic studies and biological evaluation of some transition metal complexes of azo Schiff-base ligand derived from (1-phenyl-2, 3-dimethyl-4-aminopyrazol-5-one) and 5-((4-chlorophenyl) diazenyl)-2-hydroxybenzaldehyde. *Spectrochimica Acta Part A: Molec. and Biomol Spectrosc.*, **96**, 493-500.
- Banno, K.; Fujioka, T.; Kikuchi, T.; Oshiro, Y.; Hiyama, T.; Nakagawa, K. (1988). Studies on 2 (1H)-quinolinone derivatives as neuroleptic agents. I. Synthesis and biological activities of (4-Phenyl-1-piperaziny1)-propoxy-2 (1H)-quinolinone derivatives. *Chem. and Pharmaceut. Bulle.*, **36**(11), 4377-4388.
- Broomhead, J.A.; Rendina, L.M.; Webster, L.K. (1993). Dinuclear complexes of platinum having anticancer properties. DNA-binding studies and biological activity of Bis (4, 4'-dipyrazolylmethane-N, N')-bis [dichloroplatinum (II) and related complexes. *J. Inorg. Biochem.*, **49**(3), 221-234.
- Charrier, C.D.; Salisbury, A.M.; Savage, V.J.; Duffy, T.; Moyo, E.C.; Malam, N.; Ooi, N.; Newman, R.; Cheung, J.; Metzger, R. and others (2017). Novel bacterial topoisomerase inhibitors with potent broad-spectrum activity against drug-resistant bacteria. *Antimicrobial Agents and Chemother.*, **61**(5).
- Cotton, F.A. (1999). 'Wilkinson; Murillo, CA; Bochmann, M'. Advanced Inorganic Chemistry: A Comprehensive Text; Wiley, New York. pp. 652-674.
- Da Silva, C.M.; da Silva, D.L.; Modolo, L.V.; Alves, R.B.; de Resende, M.A.; Martins, C.V.; de Fátima, Â. (2011). Schiff bases: A short review of their antimicrobial activities. *J. Adv. Res.*, **2**(1), 1-8.
- Despaigne, A.A.R.; da Silva, J.G.; do Carmo, A.C.M.; Sives, F.; Piro, O.E.; Castellano, E.E.; Beraldo, H. (2009). Copper (II) and zinc (II) complexes with 2-formylpyridine-derived hydrazones. *Polyhedron*, **28**(17), 3797-3803.

- Dhiman, P.; Arora, N.; Thanikachalam, P.V.; Monga, V. (2019). Recent advances in the synthetic and medicinal perspective of quinolones: A review. *Bioorg. Chem.*, **92**, 103291.
- Ganguly, R.; Sreenivasulu, B.; Vittal, J.J. (2008). Amino acid-containing reduced Schiff bases as the building blocks for metallasupramolecular structures. *Coordinat. Chem. Rev.*, **252**(8-9), 1027-1050.
- Geary, W.J. (1971). The use of conductivity measurements in organic solvents for the characterisation of coordination compounds. *Coord. Chem. Rev.*, **7**(1), 81-122.
- Gerloch, M.; Lewis, J.; Mabbs, F.E. (1968). The preparation and magnetic properties of some Schiff base-iron (III) halide complexes. *J. Chem. Soci. A: Inorganic, Physical, Theoretical*, 112-116.
- Golcu, A.; Tumer, M.; Demirelli, H.; Wheatley, R.A. (2005). Cd (II) and Cu (II) complexes of polydentate Schiff base ligands: synthesis, characterization, properties and biological activity. *Inorg. Chim. Acta*, **358**(6), 1785-1797.
- Govind, M.; Rajesh, N. (1992). Synthesis and anti-inflammatory activity of N-pyridinobenzamide-2-carboxylic acid and its metal chelates. *Ind. J. Pharm.*, **24**(4), 207.
- Gümüş, A.; Karadeniz, Ş.; Uğraş, H. İ.; Bulut, M.; Çakır, Ü.; Gören, A. C. (2010). Synthesis, complexation, and biological activity studies of 4-aminomethyl-7, 8-dihydroxy coumarines and their crown ether derivatives. *J. Hetero. Chem.*, **47**(5), 1127-1133.
- Guzzi, S. A.; El Alagi, H. S. (2013). Synthesis and characterization of Fe (II) and Co (II) complexes of Schiff base derived from 3, 3'-diaminobenzidine and salicylaldehyde. *J. Chem. and Pharm. Res.*, **5**(10), 10-14.
- Kocsis, B.; Domokos, J.; Szabo, D. (2016). Chemical structure and pharmacokinetics of novel quinolone agents represented by avarofloxacin, delafloxacin, finafloxacin, zabofloxacin and nemonoxacin. *Ann. Clin. Microbiol Antimicrob*, **15**(1), 1-8.
- Kong, D.; Reibenspies, J.; Mao, J.; Clearfield, A.; Martell, A. E. (2003). Novel 30-membered octaazamacrocyclic ligand: synthesis, characterization, thermodynamic stabilities and DNA cleavage activity of homodinuclear copper and nickel complexes. *Inorg. Chim. Acta*, **342**, 158-170.
- Krstic, M.; Petković, B.; Milčić, M.; Mišić, D.; Santibanez, J. F. (2019). Synthesis, characterization and biological study of new dinuclear zinc (II) and nickel (II) octaaza macrocyclic complexes. *Macedonian J. Chem. and Chem. Eng.*, **38**(1), 1-11.
- Kulkarni, P.A.; Habib, S.I.; Saraf, V.D.; Deshpande, M.M. (2012). Synthesis, spectral analysis and antimicrobial activity of some new transition metal complexes derived from 2, 4-dihydroxy acetophenones. *Res. J. Pharm. Biog. Chemical Sci.*, **3**, 107-113.
- Lashanizadegan, M.; Shayegan, S.; Sarkheil, M. (2016). Copper (II) complex of (±) trans-1, 2-cyclohexanediamine azo-linked Schiff base ligand encapsulated in nanocavity of zeolite-Y for the catalytic oxidation of olefins. *J. Serb. Chem. Soc.*, **81**(2), 153-162.
- Lever, A.B.P. (1984). *Inorganic Electronic Spectroscopy. Studies in Physical and Theoretical Chemistry*". Elsevier, 2nd ed., New York (1968). 33p.
- Liu, Y.T.; Sheng, J.; Yin, D.W.; Xin, H.; Yang, X.M.; Qiao, Q.Y.; Yang, Z.J. (2018). Ferrocenyl chalcone-based Schiff bases and their metal complexes: Highly efficient, solvent-free synthesis, characterization, biological research. *J. Organomet. Chem.*, **856**, 27-33.
- Măciucă, A.M.; Munteanu, A.C.; Uivarosi, V. (2020). Quinolone complexes with lanthanide ions: An insight into their analytical applications and biological activity. *Molecules*, **25**(6), 1347.

- Maurya, M.R.; Khurana, S.; Zhang, W.; Rehder, D. (2002). Biomimetic oxo-, dioxo-and oxo-peroxo-hydrazone-vanadium (IV/V) complexes. *J. Chem. Soc., Dalton Trans.*, **15**, 3015-3023.
- Mostafa, M. (2011). Metal chelates of hydrazone ligand chelating tendencies of 2-Carboxyphenylhydrazoneacetanilide (2-Cphaaa) Ligand. *Res. J. Chem. Sci.* ISSN, 2231, 606X.
- Mruthyunjayaswamy, B.H.M.; Ijare, O.B.; Jadegoud, Y. (2005). Synthesis, characterization and biological activity of symmetric dinuclear complexes derived from a novel macrocyclic compartmental ligand. *J. Braz. Chem. Soc.*, **16**(4), 783-789.
- Nabeel, B.H.; Farah, S.T. (2012). Synthesis and structural studies on some transition metal complexes of Bis-(benzimidazole-2-thio) ethane, propane and butane ligands. *Res. J. Chem. Sci.*, ISSN, 2231, 606X.
- Nagaraja, V.; Godbole, A.A.; Henderson, S.R.; Maxwell, A. (2017). DNA topoisomerase I and DNA gyrase as targets for TB therapy. *Drug Discov. Today*, **22**(3), 510-518.
- Nicholls, D. (2013). The chemistry of iron, cobalt and nickel: *Comprehens. Inorg. Chem.*, (24). Elsevier Science.
- Oshiro, Y.; Sakurai, Y.; Sato, S.; Kurahashi, N.; Tanaka, T.; Kikuchi, T.; Tottori, K.; Uwahodo, Y.; Miwa, T.; Nishi, Takao (2000). 3, 4-Dihydro-2 (1 H)-quinolinone as a novel antidepressant drug: synthesis and pharmacology of 1-[3-[4-(3-chlorophenyl)-1-piperazinyl] propyl]-3, 4-dihydro-5-methoxy-2 (1 H)-quinolinone and its derivatives. *J. Med. Chem.*, **43**(2), 177-189.
- Parikh, V.M. (1974). "Absorption Spectroscopy of Organic Molecules". Addison-Wesley Publication Company. Reading, MA, pp. 46-91.
- Prasad, S.; Agarwal, R.K. (2006). Nickel (II) Complexes of hydrazone of isoniazid and their magneto-spectral, electrochemical, thermal and antimicrobial investigations. *Internat. J. Inorg. Chem.*, **2008**, Article ID 350921.
- Rakhi, C.; Shelly, S. (2011). Spectral and Pharmacological Study of Cu (II), Ni (II) and Co (II) Coordination Complexes. *Res. J. Chem. Sci.*, **1**(5), 87-90.
- Raman, N.; Raja, Y.P.; Kulandaisamy, A. (2001). Synthesis and characterization of Cu (II), Ni (II), Mn (II), Zn (II) and VO (II) Schiff base complexes derived from o-phenylenediamine and acetanilide. *J. Chem. Sci.*, **113**(3), 183-189.
- Raya, I.; Baba, I.; Yamin, B. M. (2006). New mixed ligands complexes of samarium (III) with dithiocarbamates and 1, 10-phenanthroline. *Malaysia. J. Analy. Sci.*, **10**(1), 93-98.
- Reese, R. E.; Betts, R. F.; Gumustop, B.; Betts, R.; Gumustop, R.; Betts (2000). "Handbook of Antibiotics". Vol. 601. Philadelphia: Lippincott Williams and Wilkins.
- Seleem, H.S.; Mostafa, M.; Stefan, S. L.; Abdel-Aziz, E. (2011). Structural diversity of 3d complexes of an isatinic quinolyl hydrazone. *Res. J. Chem. Sci.*, ISSN, 2231, 606X.
- Sönmez, M.; Levent, A.; Şekerci, M. (2003). Synthesis and Characterization of Cu (II), Co (II), Ni (II), and Zn (II) Complexes of a Schiff Base Derived from 1- Amino- 5- benzoyl- 4- phenyl- 1H- pyrimidine- 2- one and 3- Hydroxysalicylaldehyde. *Synth. and Reac. in Inorg. and Met. Org. Chem.*, **33**(10), 1747-1761.
- Vlasenko, V.G.; Burlov, A.S.; Koshchienko, Y.V.; Kolodina, A.A.; Chaltsev, B.V.; Zubavichus, Y.V.; Klimenko, A.I. (2021). Synthesis, X-ray structure and biological activity of mono-and

dinuclear copper complexes derived from N-{2-[(2-diethylamino (alkyl) imino)-methyl]-phenyl}-4-methyl-benzenesulfonamide. *Inorg. Chim. Acta.*, **523**, 120408.

Xiao, Z.; Waters, N.C.; Woodard, C. L.; Li, Z.; Li, P.K. (2001). Design and synthesis of Pfmrk inhibitors as potential antimalarial agents. *Bioorg. and Med. Chem. Lett.*, **11**(21), 2875-2878.

Zhang, G.F.; Zhang, S.; Pan, B.; Liu, X.; Feng, L. S. (2018). 4-Quinolone derivatives and their activities against Gram positive pathogens. *Euro. J. Med. Chem.*, **143**, 710-723.

تحضير وتشخيص بعض معقدات عناصر السلسلة الانتقالية الأولى مع ليكاند ثنائي -2- كوينولينون ثنائي بنزاليدين ومشتقاتها مع 10,1 فينانثرولين

الملخص

يتضمن هذا البحث تحضير معقدات ذات الصيغة العامة $[M_2L_2]Cl_4$ و $[M_2L_2(1,10-phen)_2]Cl_4$ حيث أن :
 $Zn(II)$ and $Cu(II)$, $Ni(II)$, $Co(II)$, $Fe(II)$, $Mn(II)$ = M
 = L = ثنائي -2- كوينولينون ثنائي بنزاليدين.

10,1 = 1,10-phen فينانثرولين.

تم تحضير الليكاند من خلال مفاعلة N - أمينوكوينولين -2- ون مع التيريفثالديهايد بنسبة (1:2) وشُخص الليكاند باستخدام طيف الأشعة تحت الحمراء (IR) وطيف الرنين النووي المغناطيسي (1H NMR).

أظهر طيف الأشعة تحت الحمراء تناسق كل من ذرة النتروجين لمجموعة الأزوميثين وذرة الأوكسجين في مجموعة الكاربونيل لليكاند مع الأيون المركزي.

شخصت المعقدات المحضرة بواسطة التحليل الدقيق للعناصر، التوصيلية المولية، الحساسية المغناطيسية والقياسات الطيفية (UV - Vis, IR) أظهرت نتائج التحليل العنصري مطابقة مع الصيغة العامة المقترحة للمعقدات:

(2:2) (فلز : ليكاند) و (2:2:2) (فلز : ليكاند:10,1- فينانثرولين)، أما نتائج التوصيلية المولية فقد أظهرت أن المعقدات تكون بنسبة (4:1) بالإضافة الى ذلك فقد أظهرت نتائج الحساسية المغناطيسية والأطياف الالكترونية بأن المعقدات من نوع $[M_2L_2]Cl_4$ تمتلك بنية رباعي السطوح بينما المعقدات من نوع $[M_2L_2(1,10-phen)_2]Cl_4$ تمتلك بنية ثماني السطوح .

الكلمات الدالة: ثنائي بنزاليدين ثنائي -2- كوينولينون، فلز، معقدات، 1 و 10 فينانثرولين، ثنائي النوى.