# Synthesis and spectroscopic study of transition metal ions (Co(II), Ni(II), Cu(II) ) complexes containing a mixed ligands via salpphen and some of amino acids

# تحضير ودراسة طيفية لمعقدات ايونات عناصر انتقالية Co(II) وNi(II) و Cu(II) ) حاوية مزيج من الليكندات عن طريق السالفين وبعض الاحماض الامينية

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## Abstract

Schiff base ligand salpphen (L) were synthesized by the condensation reaction of *p*-phenylenediamine with salicylaldehyde, Schiff base ligand was used as a primary ligand and some of amino acid (Alanine or Glycine) as secondary ligand to synthesized complexes containing mixed ligands with the metals ions Co(II), Ni(II), Cu(II) . The prepared ligand(L) was characterized on the basis elemental analyses(CHN), FT-IR,$^{1}$HNMR, UV-Visible spectroscopy. While prepared complexes were characterized on the basis elemental analyses, FT-IR, $^{1}$HNMR, UV-Visible spectroscopy, conductivity and magnetic measurements. The morphology of the one of the complexes CuLAla was studied by scanning electron microscopy (SEM). The compounds were subjected to simultaneous thermogravimetric analysis (TGA/DTA) to study their decomposition mechanism and thermal stability. The spectroscopic and magnetic moments shows the suggested geometry of the metal ions complexes are octahedral. The antibacterial activities of the ligand and some of its metal complexes have been screened against bacteria *E. coli and S. aureus*.

*Keywords:* salpphen, mixed ligands, amino acids.

**الخلاصة**

تم تحضير قاعدة شيف السالفين(L) من تفاعل تكثيفي بين البارا- فنلين ثنائي الامين والساليسالديهايد، استخدمت قاعدة شيف المحضرة كاليكاند اولي اما الحامض الاميني(الكلايسين اوالالنين) استخدم كليكاند ثانوي لتحضير معقدات حاوية مزيج من الليكندات مع الايونات الفلزية Co (II) وNi (II) وCu (II) . الليكند المحضر شخص بالاعتماد على تحليل العناصر(CHN) ومطيافية الاشعة تحت الحمراءFT-IR والرنين النووي المغناطيسي للبروتونHNMR$^{1}$ والاشعة المرئية فوق البنفسجيةUV-Vis. بينما المعقدات المحضرة شخصت بالاعتماد على تحليل العناصر(CHN) ومطيافية الاشعة تحت الحمراء والرنين النووي المغناطيسي والاشعة المرئية فوق البنفسجية وقياسات التوصيلية الكهربائية والحساسية المغناطيسية. درست مورفولوجية احد المعقدات(CuLAla) بتقنية المجهر الالكتروني الماسح(SEM). درست الاستقرارية الحرارية وميكانيكيات التفكك الحراري للمركبات باستخدام التحاليل الحرارية الوزنية والتحاليل الحرارية التفاضلية(TGA/DTA). من خلال القياسات الطيفية والمغناطيسية تم اقتراح الشكل الثماني السطوح لجميع المعقدات المحضرة. درست الفعالية البيولوجية لبعض المركبات المحضرة ضد نوعين من البكتريا المرضية( *E. coli* و*S. aureus*. ).

*مفاتيح الكلمات:* السالفين ،مزيج الليكندات ،الاحماض الامينية.

## 1. Introduction

Coordination chemistry of transition metal ions has a wide interdisciplinary relevancy in day to day life. Modified pharmaceuticals are no doubt gift of above mentioned field and indirectly demand to dominate the harmful effects of bacteria, fungi and viruses. It definitely to synthesize like modifier versions. These important properties seem to be a result of chelation behavior among ligands and transition metal ion [1-4]. The coordination chemistry to the transition metal complexes as mixed ligands are of general interest because they can supplying new materials with useful properties like magnetic exchange, nonlinear optical property , electrical conductivity , and antimicrobial activity . The biological significance of mixed ligand complexes is that they are at times more effective than the free ligands. Mixed-ligand complexes containing N and O donor's atom are important because of their antibacterial, antifungal, and anticancer activities [5-7]. Mixed ligand complexes include amino acids as a secondary ligand are of importance as they are potential models for enzyme-metal ion of substrate complexes. The lot of interest is being given to the study of mixed ligand complexes of Schiff bases include salicylaldehyde with amino acids [8-10].

The aim of the study is synthesis ligand contend multi_electronic dentate (Schiff base), synthesis complex for transition metal ion with mixed ligands (Schiff base and selected amino acids), and evaluation of their antibacterial activity.

## 2. Experimental

All the chemicals were of reagent grade, were used as supplied from (Fluka), (B.D.H.), (Himedia), (Merck). Conductivity measurements for $10^{-3}$M solution of the complexes in (DMF) were carried out with on Digital Conductivity Meter - WT-720 – ino Lab (Germany). Infrared spectra were recorded on a FT-IR-8400 S shimadzu (Japan). The UV/Vis spectra were recorded on UV-Visible spectrophotometer – 1800 shimadzu (Japan) for $10^{-3}$ M solution of complexes in DMSO using 1cm quartz cell. Melting points were measured using Stuart Melting points apparatus (England). Magnetic susceptibility was measured by using Magnetic Susceptibility Balance, Johnson Mattey (England). The NMR spectra were recorded on Broker - 400 (Germany). The element analysis was measured by using Thermo Finning Flash EA1112. The Thermogravimetric analyses (TGA/DTA) was measured by using Perkin Elmer STA6000/TGA4000. Scanning Electron Microscopy (SEM) by use Inspect S50 SEM FEI Company.

### 2.1. Synthesis of ligand salpphen (L)

The ligand L was prepared according to the literature [11], to the solution of *p*-phenylenediamine (4.623 mmol ،0.5g) in (20 mL) absolute ethanol, a solution of salicylaldehyde (9.245 mmol ، 1.129g) was add , the mixture was reflexed for two hours , the color of the mixture was change to yellow after filtering. The precipitate was washed with diethyl ether, the yellow precipitate was then recrystallized from toluene, organic precipitate was obtaining. Table 1 is illustrated some of properties to the ligand(L).

**Table (1) Colors, yields%, elemental analyses, and melting points of the prepared compounds**

| Compound | Color | Yield % | m.p.°C | % H | | % N | | % C | |
|---|---|---|---|---|---|---|---|---|---|
| | | | | Cal. | Fou. | Cal. | Fou. | Cal. | Fou. |
| L | Orang | 83 | 218-220 | 5.1 | 5.02 | 8.86 | 8.44 | 75.93 | 75.43 |
| CoLAla | Brown | 63 | 352-354 | 4.31 | 4.26 | 9.21 | 9.1 | 51.33 | 51.11 |
| CoLGly | Black brown | 59 | 354-357 | 3.82 | 3.65 | 9.65 | 9.59 | 49.67 | 49.43 |
| NiLAla | Green | 64 | 251-253 | 4.31 | 4.19 | 9.22 | 9.15 | 51.37 | 51.21 |
| NiLGly | Black | 67 | 288-290 | 3.82 | 3.49 | 9.66 | 9.46 | 49.71 | 49.19 |
| CuLAla | Black | 66 | 296-298 | 4.24 | 4.19 | 9.07 | 8.95 | 50.56 | 50.27 |
| CuLGly | Black | 62 | 320-323 | 3.76 | 3.53 | 9.5 | 9.25 | 48.89 | 48.27 |

### 2.2. Synthesis of complexes from mixed ligands

To the solution of chloride of the metal ion ($CoCl_2.6H_2O$, $NiCl_2.6H_2O$ and $CuCl_2.2H_2O$) (3.1 mmol) in (20 mL) acetone , a solution of the ligand L (1.5 mmol) in (30 mL) acetone, and a solution of amino acid (3.1 mmol) in (3 mL, 0.1M HCL) was add, the pH of the mixture was about (10-11) by add (1M NaOH), the mixture was reflexed to 12 hours. The formed precipitate was filtered and washed with diethyl ether, Table 1 shows Colors, yields %, elemental analyses, and melting points.

### Results and Discussion

**FT-IR:** the most important infrared bands for the ligand and its complexes were listed in Table 2, the υ(C=N) imine stretching band at (1612) $cm^{-1}$ belong to the Schiff base ligand [12] was shifted to higher frequency with complexes [13], the υ(O-H) in the ligand was at (3419)$cm^{-1}$ [14] , the υ($COO^-$) symmetric and asymmetric to the amino acid was at (1410, 1527)$cm^{-1}$ [15] , these bands in the complexes was shifted and changed in intensities, The υ(-$NH_2$) symmetric and asymmetric to the amino acid was at (3167-2975)$cm^{-1}$ [16]. They shift to higher frequencies in the complexes. Further proof for the involvement of both azomethine , the $COO^-$ and $NH_2$ groups in complexation is the appearance of weak bands in the far IR region of complexes at (500–600) and (600–700) due to t(M-N) and t(M-O) respectively which are absent in the free ligands [17,18] . Figures 1(A-G) shows the FT-IR spectra for the ligand and its complexes.

**Table (2) FT-IR frequencies in ($cm^{-1}$) of the prepared compounds**

| compound | υC=N imine | υN-H asym. (aa) | υN-H sym. (aa) | $\upsilon COO^-$ asym. (aa) | $\upsilon COO^-$ sym. (aa) | υO-H $H_2O$ | υM-O | υM-N |
|---|---|---|---|---|---|---|---|---|
| L | 1612 | ……. | ……. | ……. | ……. | 3419 | ……. | ……. |
| CoLAla | 1650 | 3265 | 3166 | 1595 | 1446 | 3558 | 657 | 520 |
| Ni LAla | 1680 | 3335 | 3249 | 1573 | 1419 | 3471 | 650 | 543 |
| Cu LAla | 1610 | 3255 | 3055 | 1525 | 1456 | 3360 | 673 | 549 |
| Co LGly | 1678 | 3331 | 3055 | 1616 | 1454 | 3444 | 611 | 557 |
| Ni LGly | 1652 | 3360 | 3055 | 1614 | 1457 | 3472 | 623 | 532 |
| Cu LGly | 1651 | 3271 | 3050 | 1599 | 1443 | 3333 | 682 | 559 |

(aa) amino acid

**$^1$HNMR:**

The $^1$HNMR spectra of the ligand (L) (Table 3, Fig.2 A) display two signals at 9.04 and 13.09 ppm to protons corresponding to the CH=N- and -OH groups. the aromatic protons display multi signals at (6.23-7.69) ppm [19,20], in the complexes (Fig.2(B-E)) these signals was shifted due to coordination with metal ion. In the complex CuLAla the signal of Ar-OH did not appear. Furthermore the complexes shows signals of protons of $CH_3$, $CH_2$, CH, $NH_2$ to amino acid [21,22], Table 4 shows the most important signal of $^1$HNMR for the ligand and its complexes.

**Table (3) $^1$HNMR in (ppm) for the ligand L and its complexes with amino acids**

| compound | Ar-H | Ar-OH | CH=N | $CH_3$ | CH | $CH_2$ | $NH_2$ |
|---|---|---|---|---|---|---|---|
| L | 6.23-7.69 | 13.09 | 9.04 | ... | .... | .... | .... |
| CoLGly | 6.424-8.003 | 13.728 | 8.874,8.976 | ... | .... | 4.620 | 5.429 |
| NiLAla | 6.598-7.531 | 13.676 | 8.838 | 1.232 | 3.362 | ... | 5.352 |
| NiLGly | 6.604-7.804 | 13.09, 13.712 | 8.858, 9.036 | .... | .... | 3.366 | 5.364 |
| CuLAla | 6.669-8.797 | .... | 9.679-9.683 | 1.238 | 3.558 | .... | 5.005 |

**Thermogravimetric analyses (TGA/DTA):**

Thermogravimetric analyses for the ligand (L) and its some complexes (Figures.3(A-D) with the amino acid (Alanine) have been explained by using the technic of TGA/DTA at heating rang of 30-800°C at 10°C/min , Table 4 shows the percentage of the loss of weight and the suggest compound that loss and suggest compound result after decomposition. The TGA/DTA for the ligand and its complexes showed that the ligand is less stable than its complexes. The complexes show the gradual loss in weight due to decomposition by fragmentation with increasing temperature.

**Table (4 ) Thermogravimetric analysis (TGA/DTA) for some prepared compounds**

| compound | Weight loss % | | Temp. range of decomp. °C | Lost suggest species | Result suggest species |
|---|---|---|---|---|---|
| | Found | calculate | | | |
| L | 36.0 | 37.6 | 27.87-335 | - phOH, - CH=N | ph, phOH, CH=N |
| | 59.0 | 62.0 | 335-790 | ph,- phOH, -CH=N | .... |
| CoLAla | 10.0 | 10.5 | 0-260 | $-4H_2O$ | 2 $C_3H_6NO_2$(Ala)<br>2phO, ph, 2CH=N, 2Co |
| | 14.0 | 13.0 | 260-500 | $-C_3H_6NO_2$(Ala) | $C_3H_6NO_2$(Ala)<br>2phO, ph, 2CH=N, 2Co |
| | 19.0 | 20.9 | 500-655 | $-C_3H_6NO_2$(Ala)<br>-2CH=N | 2phO, ph , 2Co |
| | 5.0 | 4.7 | 655-800 | -2O | 3ph, 2Co |
| NiLAla | 18.0 | 18.5 | 0-110 | -2Cl<br>$-3H_2O$ | 2Ala, 2phOH, ph, 2CH=N, 2Ni |
| | 11.0 | 11.6 | 110-180 | -Ala | Ala, 2phOH, ph, 2CH=N, 2Ni |
| | 11.0 | 11.6 | 180-305 | -Ala | phOH, phO, ph, 2CH=N, 2Ni |
| | 27.0 | 26.9 | 305-492 | -2ph,-2CH=N,-H | 2OH,ph, 2Ni |
| | 8.0 | 9.4 | 492-814 | -ph | 2NiO |
| CuLAla | 33.3 | 34.3 | 0-330 | $-4H_2O$ , -Ala ,<br>-ph | Ala, 2phO, 2CH=N, 2Cu |
| | 13.3 | 13.9 | 330-620 | -Ala | 2phO, 2CH=N, 2Cu |
| | 2.4 | 2.9 | 620-720 | -CH=N | 2phO, CH=N, 2Cu |
| | 2.7 | 3.9 | 720-800 | -CH=N | 2ph, 2CuO |

**Molar conductance:**

The molar conductivity data are summarized in Table 6, the molar conductance values for the complexes CoLAla, NiLGly, CuLAla indicating the nonelectrolyte nature, and for the complexes CoLGly, NiLAla, CuLGly indicating 1:1 electrolytic nature for this complex.

**Magnetic susceptibility**:

The magnetic moment of Co(II) complexes was about (4.13, 4.29)B.M this values suggested octahedral structure[23], and for the complexes of Ni(II) was about (3.16, 2.06)B.M this values suggested octahedral structure[24], and for Cu(II) complexes was about (1.54, 1.41)B.M the lower value suggested octahedral structure[25], the values of magnetic moment listed in Table 6 .

**Electronic spectra:**

The electronic spectra for the ligand L and its complexes shown in Figures(4 A-G), the spectrum of the ligand exhibited band at 250nm for $\pi\rightarrow\pi^*$ transition (phenyl) and band at 375nm for $n\rightarrow\pi^*$ transition (HC=N) [26], the spectra of Co(II) complexes showed bands between 540-690nm correspond to $^4T_1g\rightarrow{}^4A_2g^F$ transition based on the nature of band on octahedral geometry [27], the spectrum of Ni(II) complexes showed band that correspond to $^3A_2g\rightarrow{}^3T_1g^F$, $^3A_2g\rightarrow{}^3T_1g^P$ transition this band back to octahedral geometry [28], and the spectra of Cu(II) complexes showed band between 550-800nm correspond to $^2B_1g\rightarrow{}^2A_1g$, $^2B_1g\rightarrow{}^2B_2g$, $^2B_1g\rightarrow{}^2Eg$ transition this transition belong to distorted octahedral[29].Table 5 shows the molar conductivity, magnetic moment, electronic spectral for the prepared complexes.

**Table (5) Electronic spectral data, magnetic moment and molar conductivity of the complexes in DMSO**

| complexes | λ(nm) | υ ($cm^{-1}$) | transitions | $\mu_{eff}$ BM | conductivity ($S.cm^2.mole^{-1}$) | Suggestion formula |
|---|---|---|---|---|---|---|
| CoLAla | 540-650 | 18518-15384 | $^4T_1\ g \rightarrow {}^4A_2g^F$ | 4.13 | 40.6 | Oh |
| CoLGly | 580-690 | 17241-14492 | $^4T_1\ g \rightarrow {}^4A_2g^F$ | 4.29 | 52.9 | Oh |
| NiLAla | 510-560<br>640-690 | 19607-17857<br>15625-14492 | $^3A_2g \rightarrow {}^3T_1g^P$<br>$^3A_2g \rightarrow {}^3T_1g^F$ | 3.16 | 53.3 | Oh |
| NiLGly | 750<br>690 | 13333<br>14492 | $^3A_2g \rightarrow {}^3T_1g^F$<br>$^3A_2g \rightarrow {}^3T_1g^P$ | 2.06 | 10 | Oh |
| CuLAla | 550-800 | 18181-12500 | $^2B_1g \rightarrow {}^2A_1g$<br>$^2B_1g \rightarrow {}^2B_2g$<br>$^2B_1g \rightarrow {}^2Eg$ | 1.54 | 40.8 | Oh |
| CuLGly | 550-800 | 18181-12500 | $^2B_1g \rightarrow {}^2A_1g$<br>$^2B_1g \rightarrow {}^2B_2g$<br>$^2B_1g \rightarrow {}^2Eg$ | 1.41 | 52.3 | Oh |

**Scanning Electron Microscopy (SEM):**

The SEM use to study the morphology of complexes by many researchers on the field of coordination chemistry [30-32].

The SEM was taken at 30kV accelerating voltage and magnification was fixed according to 98x-21723x, in SEM image macroscopic phase separation dense layer was noticed, by some image that take for the complex CuLAla show the crystalline nature very clearly special on magnification 23120x. The crystalline nature can be describe as linear fibre random for the complex CuLAla with length 0.47µm and broaden 0.2µm by program *MacBiophotonics Image*J, (Figure 5).

**Biological activity:**

Biological activity for the ligand L and some of its complexes was test as inhibition growth active against gram negative bacteria *Escherichia coli* and gram positive bacteria *Staphylococcus aureus*, by method of disc diffusion. The solvent used was DMSO, and the sample concentrations were 500 ppm, The test results obtained are listed in Table 6 and figures 6(A-C).

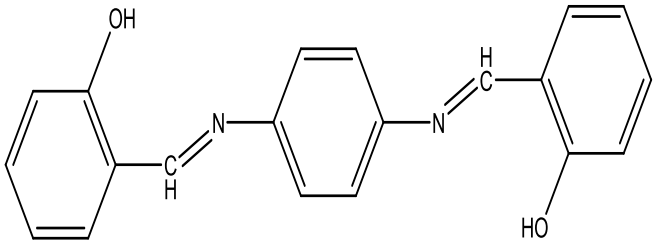
**Table (6) The antibacterial activities of the ligand and some of its metal complexes**

| compound | E.Coli | Staphylococcus aureus |
|---|---|---|
| | Diameter of zone of inhibition (mm) | |
| L | 17 | 22 |
| CoLAla | 14 | 15 |
| CoLGly | 17 | 18 |

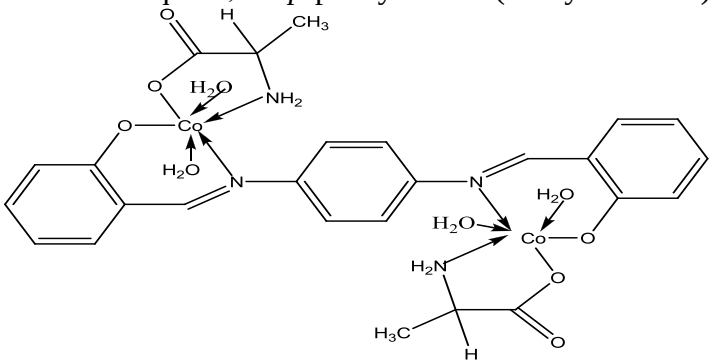
The result show that the inhibition in growth of gram positive do much of gram negative, the ligand inhibition in the growth of bacteria do much of its complex.

**3. Proposed structures for ligand(L) and its complexes with its name:**

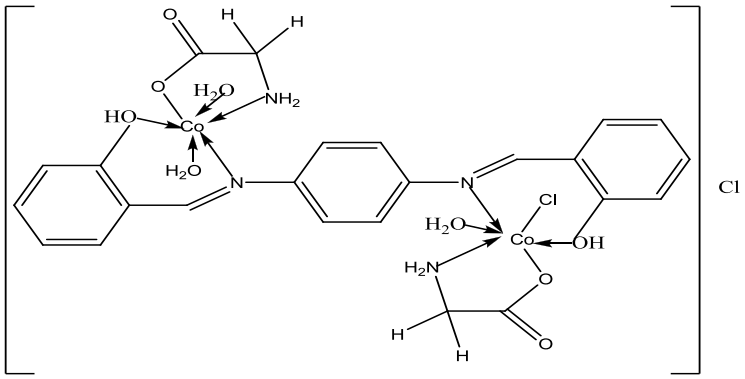
(L) *N,N`* -*p*-phenylene-bis(salicylideimine)



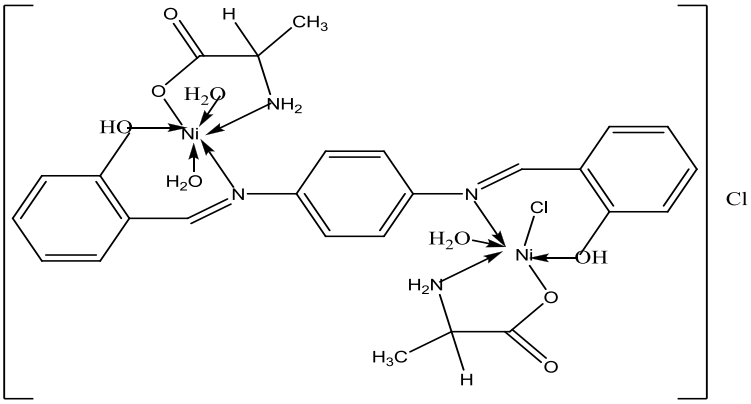

(CoLAla) dialaninatotetraaqua *N,N`* -*p*-phenylene-bis(salicylideimine)atodicobalt(II)



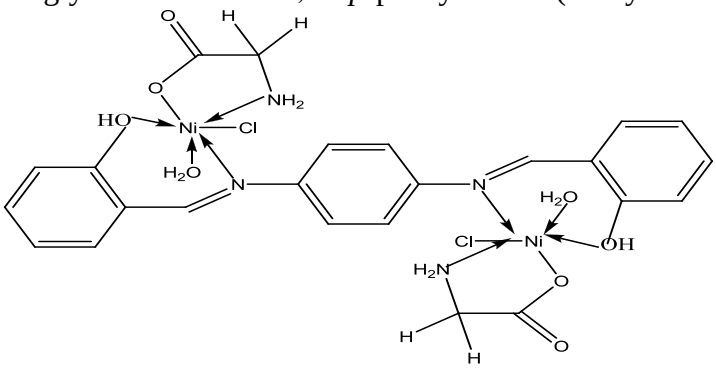
(CoLGly) triaquadiglycinatochloro *N,N`-p*-phenylene-bis(salicylideimine)dicobalt(II) chloride



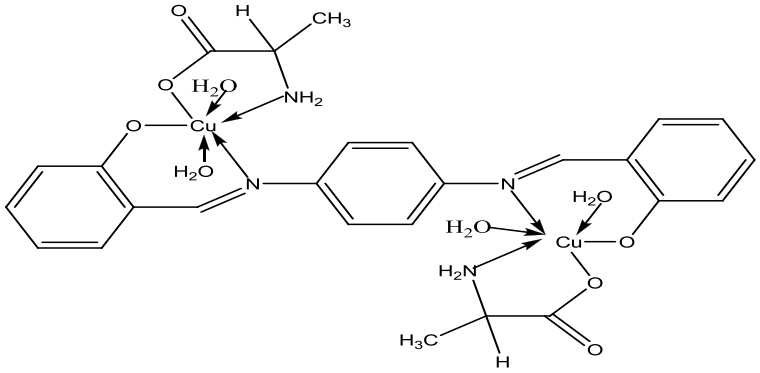
(NiLAla) dialaninatotriaquachloro *N,N`-p*-phenylene-bis(salicylideimine)dinikel(II) chloride



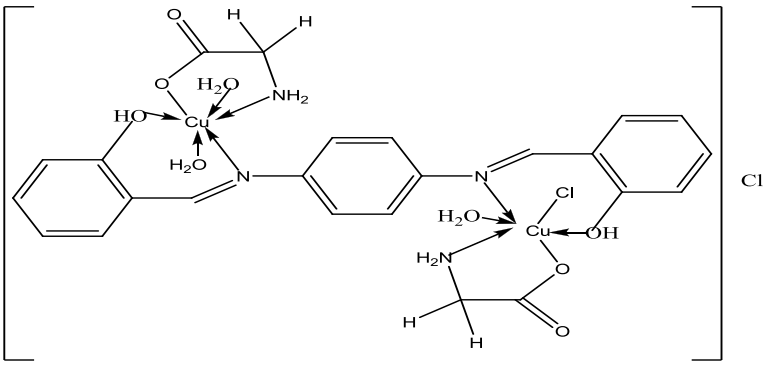
(NiLGly) diaquadiglycinatodichloro *N,N`-p*-phenylene-bis(salicylideimine)dinikel(II)



(CuLAla) dialaninatotetraqua *N,N`-p*-phenylene-bis(salicylideimine)atodicopper(II)



(CuLGly) triaquadiglycinatochloro *N,N` -p*-phenylene-bis(salicylideimine)dicopper(II) chloride



## 4. Conclusion

According to elemental analyses(CHN), $^{1}$HNMR spectra, FT-IR spectra, UV-Vis spectra, and magnetic moment the structural of the ligand and its complexes were proposed, from the spectra of $^{1}$HNMR and FT-IR was clear that the group of –OH in the complexes CoLAla and CuLAla was ionized. The molar conductance values for the complexes CoLAla, NiLGly, CuLAla indicating the nonelectrolyte nature, and for the complexes CoLGly, NiLAla, CuLGly indicating 1:1 electrolytic nature. The morphology of complex CuLAla showed the crystalline nature very clearly. The compounds were subjected to simultaneous thermogravimetric analysis (TGA/DTA) to study their decomposition mechanism and thermal stability. From physical measurements and magnetic moment was clear that the complexes were octahedral geometry.

## References

[1] Mathur, N., Kasana, A., Bargotya, S., Manna B., (2014). Biological Activities of Some New Environmentally Safe 2-Aminobenzothiazole Complexes of Copper (II) Derived Under Microwave Irradiation. *IAAST*, 5,37-42

[2] Quyoom, S., (2014). Studies of Some Mixed ligand Ternary Metal Complexes of N Acetyl Cysteine with Zinc (II) and Nickel (II) Metal ions. *Res. J. Chem. Sci.* 4, 32-35.

[3] El-Ajaily, M. M., Alassbaly, F. S., Etorki, A. M., Maihub, A. A.,(2015). Mixed-Ligand Chelate Formation of Co(II), Ni(II), Cu(II) and Zn(II) Ions with Schiff Base as Main Ligand and Amino Acid as Co-Ligand. *IRJPAC*, 5, 229-237.

[4] Kekare, M., Vaidya, V., Thakur, J., Patil, S., Langi, B., (2012). Synthesis, Characterization and Biological Studies on Some Mixed Ligand La (III) Complexes. *IJSR*.3(11),178-183

[5] Halli, M.B., Patil, V.B., Sumathi, R.B., Mallikarjun, K., (2012), Synthesis, characterization and biological activity of mixed ligand metal (II) complexes derived from benzofuran-2-carbohydrazide Schiff base and malonyldihydrazide. *Der Pharma Chemica*. 4(6), 2360-2367.

[6] Jaafar, Z. N., (2013). Synthesis, Characterization and Biological Activity of Co(II),Ni(II),Cu(II) and Zn(II) Complexes with Mixed Ligands of Alanine and Tributylphosphine. *IJIBC*. 3(4): 70-73.

[7] Hossain, A., Islam, S., Alam, A., Sultan, T.,(2013). Synthesis, Physiochemical Studies And Antimicrobial Screening Of Metal Complexes Of Fe(III) & Au(III) With Amino Acids. *Int. J. Sci. Tech.Res.* 210-217.

[8] Patil, A. R., Donde, K. J., Raut, S. S., Patil, V. R., Lokhande, R. S.,(2012). Synthesis, characterization and biological activity of mixed ligand Co(II) complexes of schiff base 2-amino-4 nitrophenol-n-salicylidene with some amino acids. *J. Chem. Pharm. Res.,* 4, 1413-1425.

[9] Rajasekar, K., Ramchandramoorthy R.T., Paulraj, A., (2012) " Microwave assisted synthesis, structural characterization and biological activities of 4-aminoantipyrine and thiocyanate mixed ligand complexes", *Res.J. Pharmaceutical Sci.*, 1, 22-27.

[10] Abdelkarim, A. T., (2015). Spectroscopic characterization of novel Cu(II) mixed-ligand complexes involving tridentate hydrazone ligand and some amino acids as antibacterial and antioxidant agents. *Int J Pharma Sci.* 5, 839-851.

[11] Pelin, S., Yurdanur, A., Funda, D.,(2007). Synthesis and Spectroscopic Characterization of New $[Cu(tBu\text{salpphen})]_2$, $[MoCl_2\ (\text{salpphen})]_2$ Complexes and $Sn_2Cl_8(tBu\text{salpphen}).2HNEt_3$ Adduct. *Turk J Chem*. 201 – 209.

[12] Uddin, M. N., Salam, M. A., Sultana, J., (2015). Pb(II) complexes of Schiff bases derived from benzoylhydrazine as the antibacterial agents. *Modern Chem.,* 7-14.

[13] Taghreed. H., Amer. J., Hussein, A. O., (2014). Synthesis, Physico-Chemical and Antimicrobial Properties Of Some Metal (II) -Mixed Ligand Complexes Of Tridentate Schiff Base Derives From B-Lactam Antibiotic {(Cephalexin Mono Hydrate)-4-Chlorobenzophenone} And Saccharin. *Int. J. Chemical Process Engineering Res.* 109-120.

[14] Bhatt, N.V., Seshadri, D.T., Phadke, R., (2012).Synthesis and characterization of Dodecylbenzene Sulfonic Acid doped Tetraaniline via Emulsion Polymerization. *E-Journal of Chemistry, 9(3), 1342-1346*.

[15] Shakir, M., Varkey, S.P., (1995). " Synthesis of novel Transition – metal complexes of 13-14-member-ed-Tetraamines and their Transition metal Complexes ", *Polyhedron*, 14, 9, 1117 – 1127 .

[16] Ajay, R.P., Kamini J.D., Sambhaji, S.R., Vishwanath R.P., Ramo S.L., (2012) . Synthesis, characterization and biological activity of mixed ligand Co(II) complexes of Schiff base 2-amino-4-nitrophenol-n-salicylidene with some amino acids . *JCPR,* 4(2):1413-1425.

[17] Kazuo,N., (2009) . Infrared and Raman Spectra of Inorganic and Coordination Compound : Part A :Theory and Applications in Inorganic Chemistry , $6^{th}$ Ed. . John Wiley and Sons, Inc., New York.

[18] Nakamoto,K., (1997). . Infrared and Raman Spectra of Inorganic and Coordination Compound : $5^{th}$ Ed., John Wiley and Sons, Inc., New York .

[19] Bushra, K., Mohammed, A. H., Askar, K. A.,( 2014). Synthesis & Characterization of New Schiff Bases Derived From 2-hydroxybenzaldehye & Amino Acids and Their Vanadyl Complexes. *RJPBCS* 5,1457-1472.

[20] Pattanaik, S., Rout, S.S., Panda, J., Sahu, P.K., Banerjee, M., (2011). Synthesis, Characterisation and Biological Evaluation of Bidentate Ligands (Reduced Schiff's Base) With Metals of Copper, Nickel and Zinc Complexes. *RASAYAN J. Chem.* 4, 136-141

[21] Aghera, V. K., Parasania, P. H., (2009) A cleaner approach for reduction of some symmetric diimines using $NaBH_4$ . *Indian J.chem*. 48B, 438-442 .

[22] Khaleel, A. A., Abdel Aziz A., (2014). Reduced Schiff base zinc complexes as proposed models of the active site of the dinuclear zinc enzyme A. Aminopeptidase. *J. Chem. Pharm. Res.*, 6, 162-171.

[23] Shavaleev, N.M., Mohammad, N.K., (2013). "Redox properties of Cobalt (II) Complexes with Azole- pyridines ", *Inorg. Chim. Acta*, 20, 261- 268 .

[24] Taghreed, H., Jarad, A. J., Hussein, A. O.,(2014). Synthesis, Physico-Chemical And Antimicrobial Properties Of Some Metal (Ii) -Mixed Ligand Complexes Of Tridentate Schiff Base Derives From B-Lactam Antibiotic {(Cephalexin Mono Hydrate)-4-Chlorobenzophenone} And Saccharin. *Int. J. Chem. Process Engineering Res.*, 109-120.

[25] Syed Khalid Aftab, (2010). Studies of the Effect of Transition Metal Ion on the Biological Activity of Some Novel Organic Compounds. Ph. D. Thesis, University Of Sargodha.

[26] Gup, R., Kırkan, B.,(2005). Synthesis and spectroscopic studies of copper(II) and nickel(II) complexes containing hydrazonic ligands and heterocyclic coligand. *Spectrochimica Acta Part A* 62, 1188–1195.

[27] Wilson, C. Y., Velayudham, S., Manickam, S., Deivanayagam, E., (2015). β - Tolyl Alanine Derived Schiff Base Complexes –Synthesis Characterization and Antimicrobial Assessment. *J. Pharm. Sci. & Res*. 7, 25-32.

[28] Mahmoud, W. H., Mohamed, G. G., Maher, M.I., (2014).Synthesis, Characterization and in vitro Biological Activity of Mixed Transition Metal Complexes of Lornoxicam with 1,10-phenanthroline . *Int. J. Electrochem. Sci.*, 1415 – 1438.

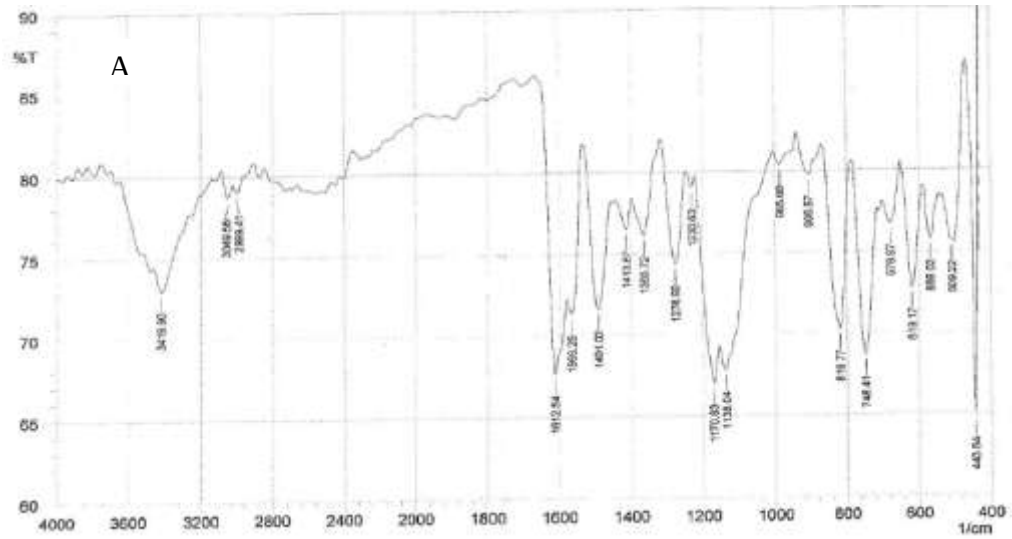
[29] Ahmed, M., (2014). Synthesis and Structural Analysis of Copper (II) Glutathione Complexes via Cu-S Linkage. *World Appl. Sci. J.*, 1357-1362.

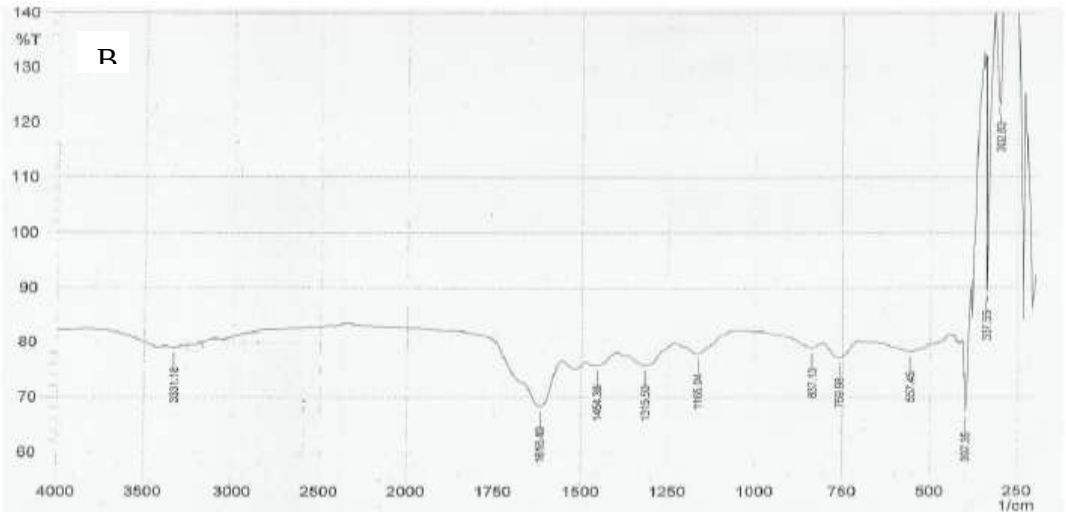
[30] Sravanthi, P., Chandrakala, C., Nagaraja, K., Jeyaraj, B.,(2015). Development of Cobalt Schiff Base Precursors for Nanocrystallinecobalt Oxide Thin Film by Thermal Cvd Method. *Ijpcbs* .112-125.

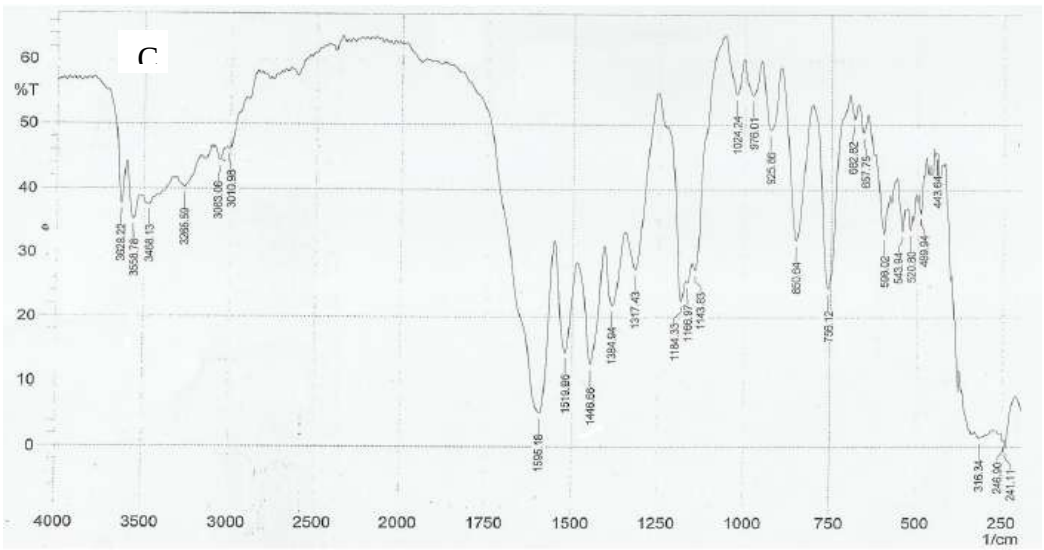
[31] Subbaraj, P., Ramu, A., Raman, N., Dharmaraja, J., (2013). Mixed Ligand Complexes Containing (2-Hydroxy- 4 -Methoxyphenyl) (Phenyl) Methanone and 2-Aminophenol: Synthesis and DNA Cleavage. *IJESE.1(7),* 79-84.

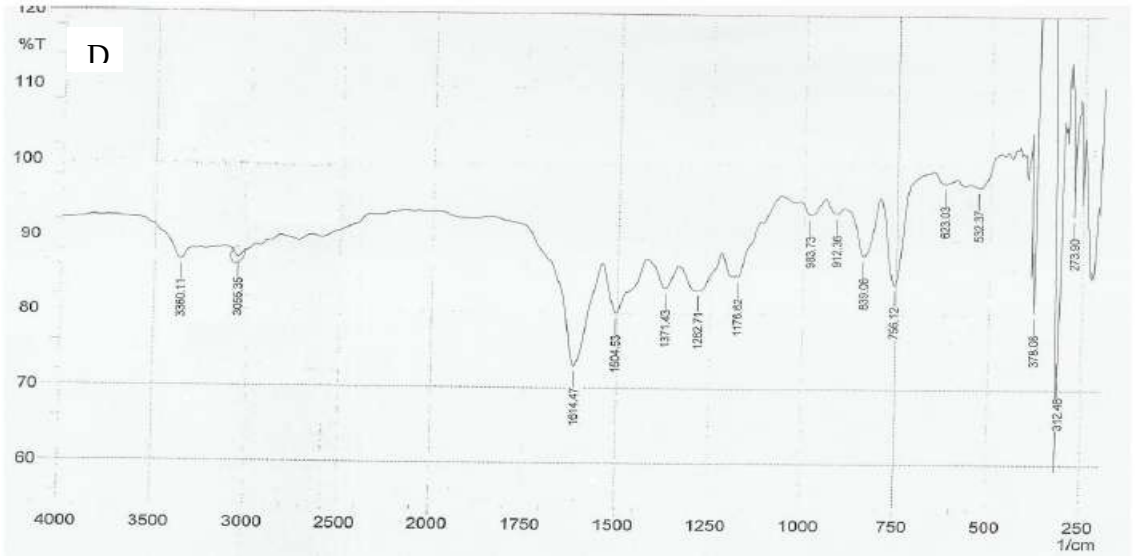
[32] Leelavathy, C., Antony, S. A., (2013). Structural Elucidation And Thermal Studies Of Some Novel Mixed Ligand Schiff Base Metal (II) Complexes. *IJBACS*. 3(4),88-95.

## Figures

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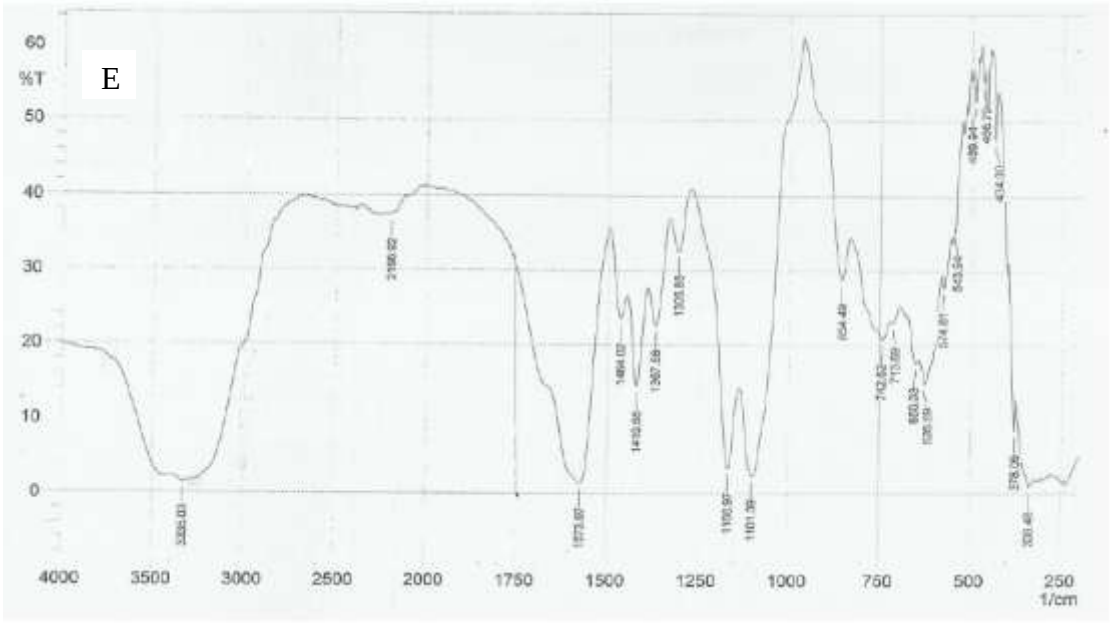
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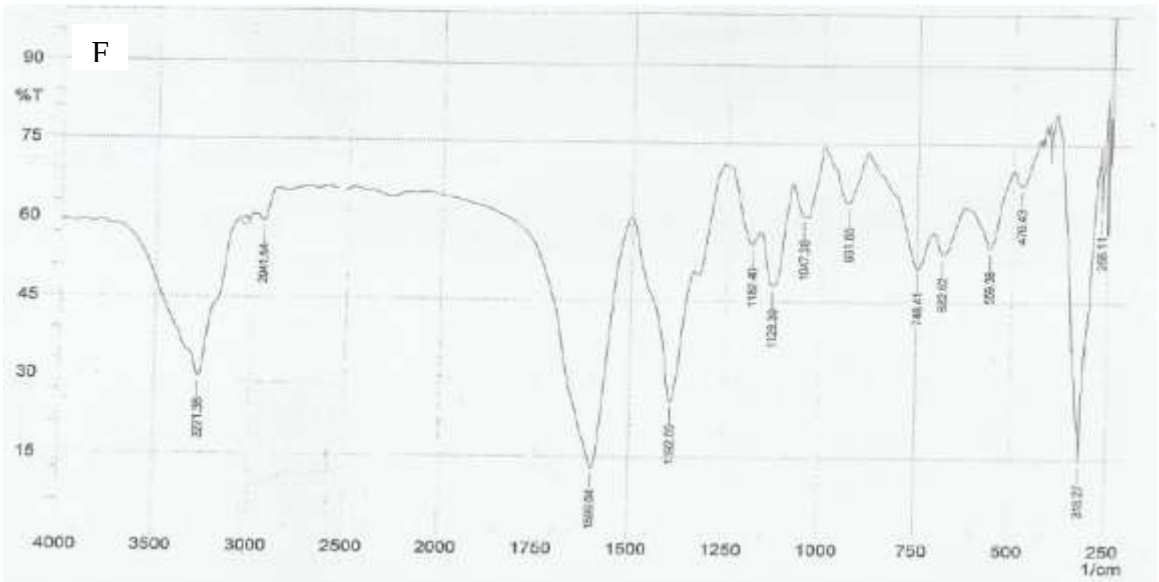
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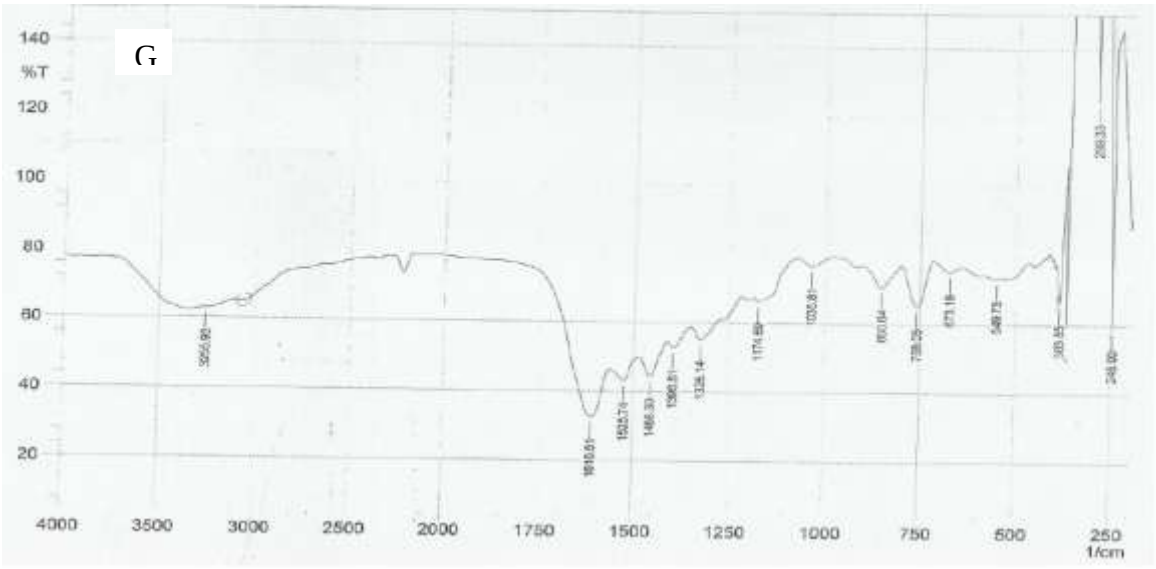
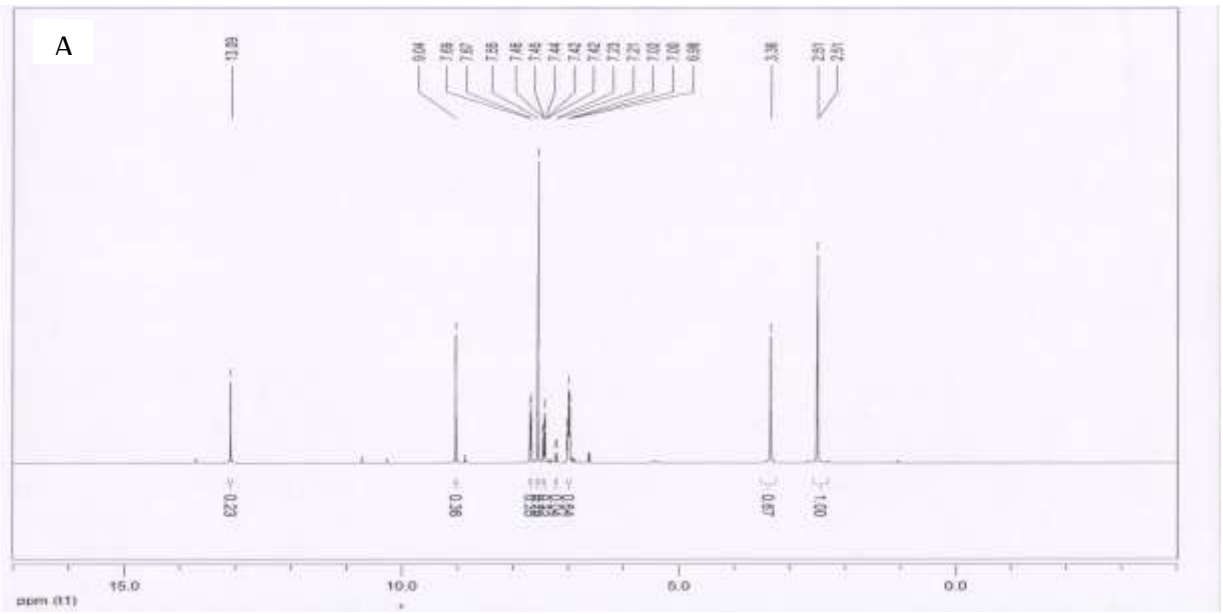
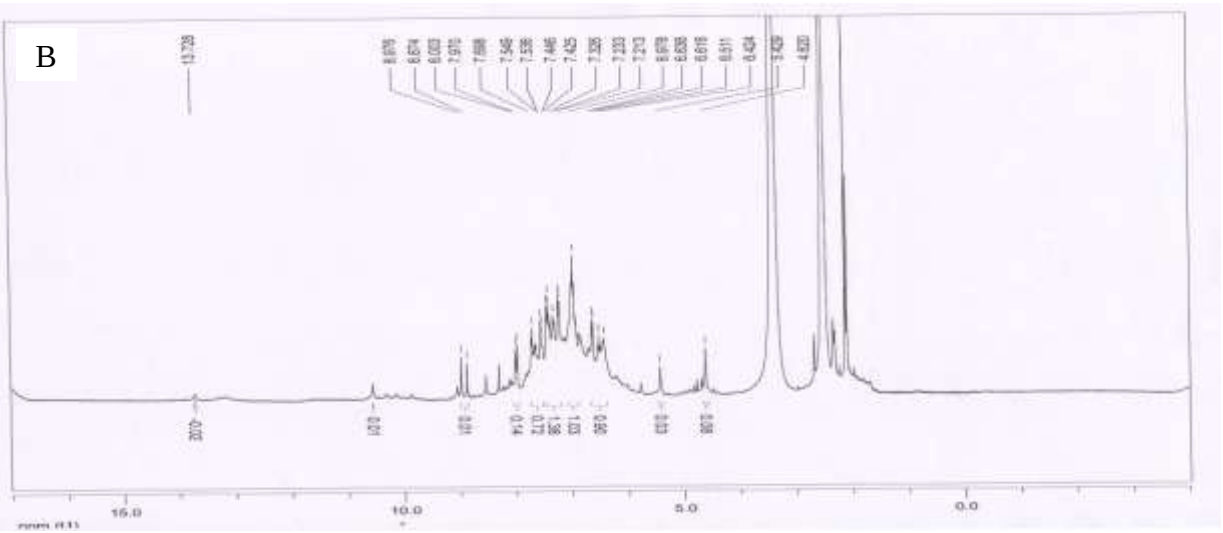
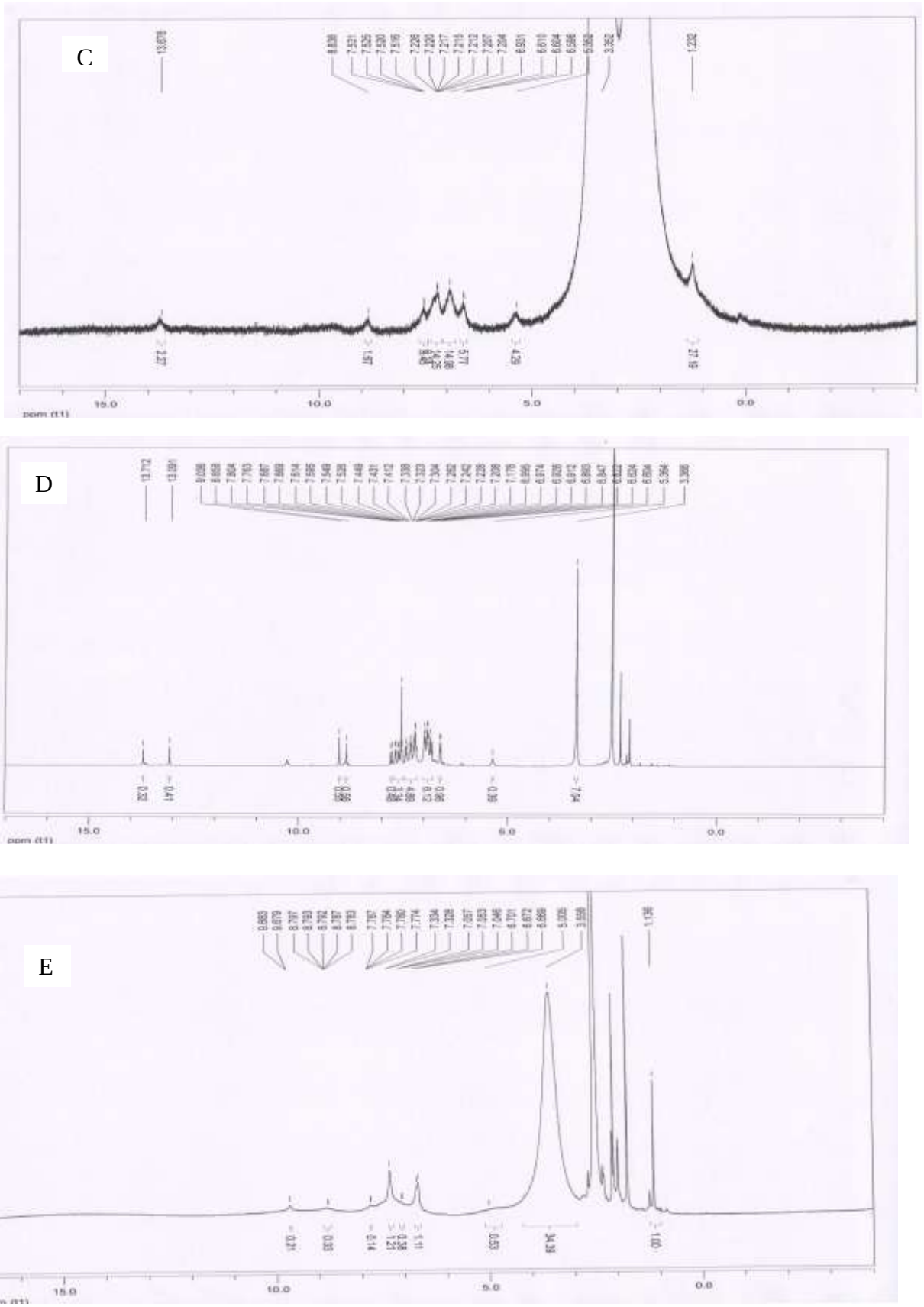


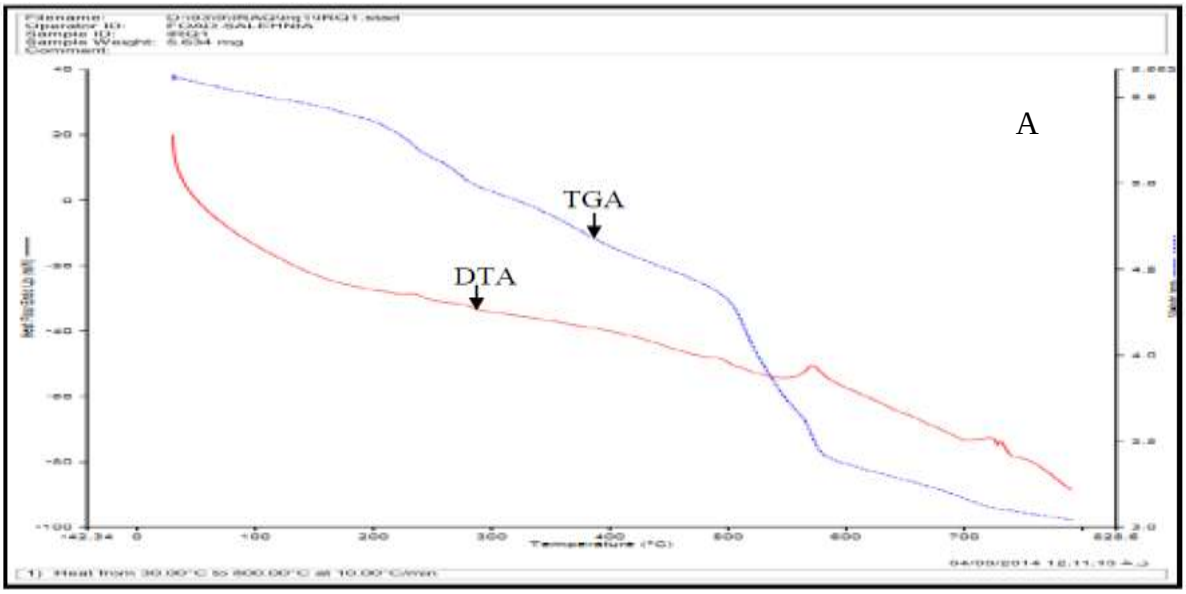

Figure (1) FT-IR spectra for :A( ligand(L)), B (CoLAla), C (Ni LAla), D (Cu LAla),E (Co LGly), F (Ni LGly), G (Cu LGly)






**Figure( 2) [1]HNMRR spectra for :A( ligand(L)), B (CoLGly), C (Ni LAla), D (Ni LGly),E (CuLAla**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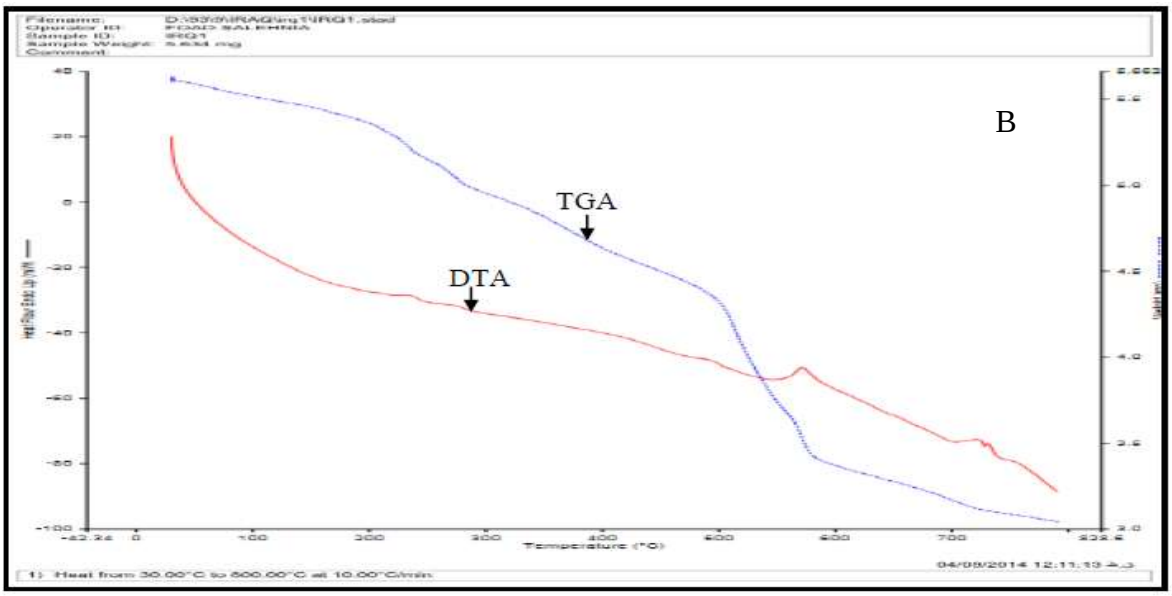
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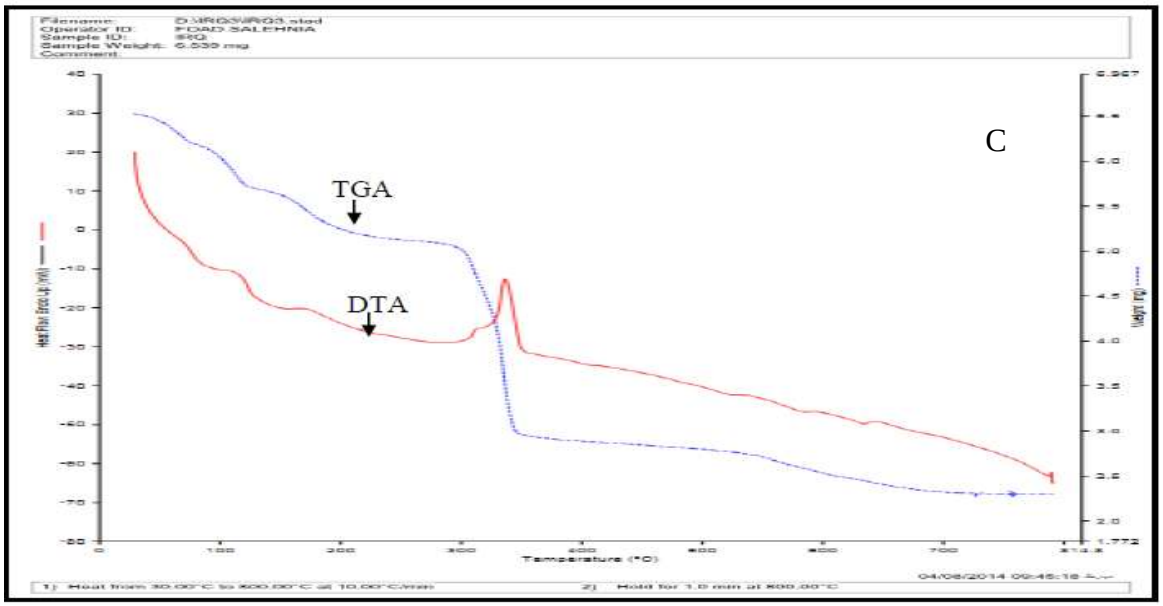
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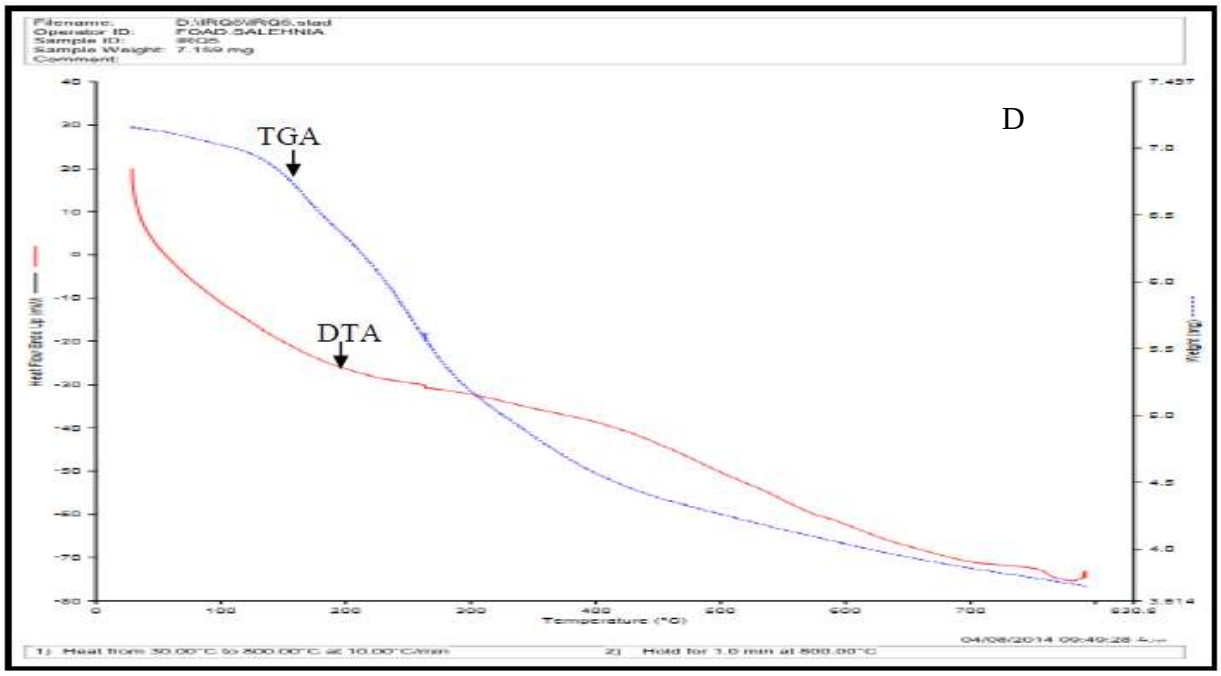
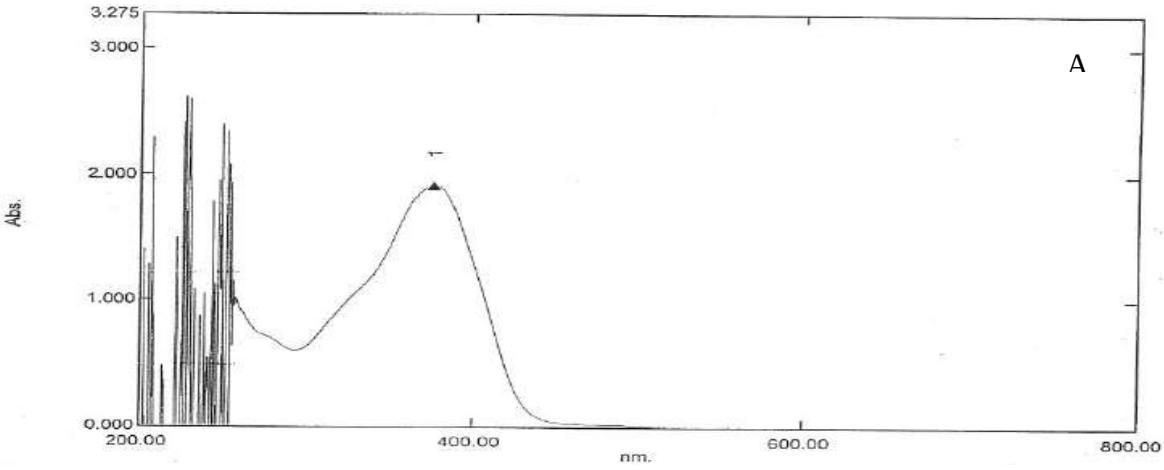
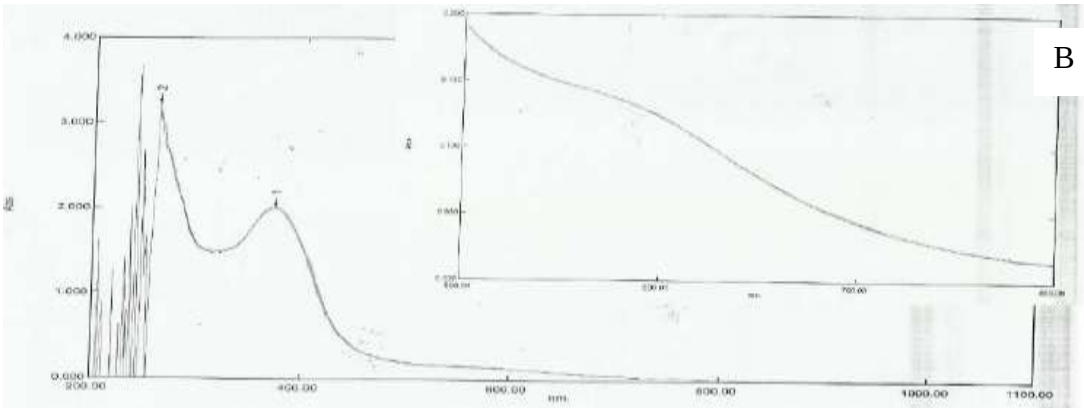
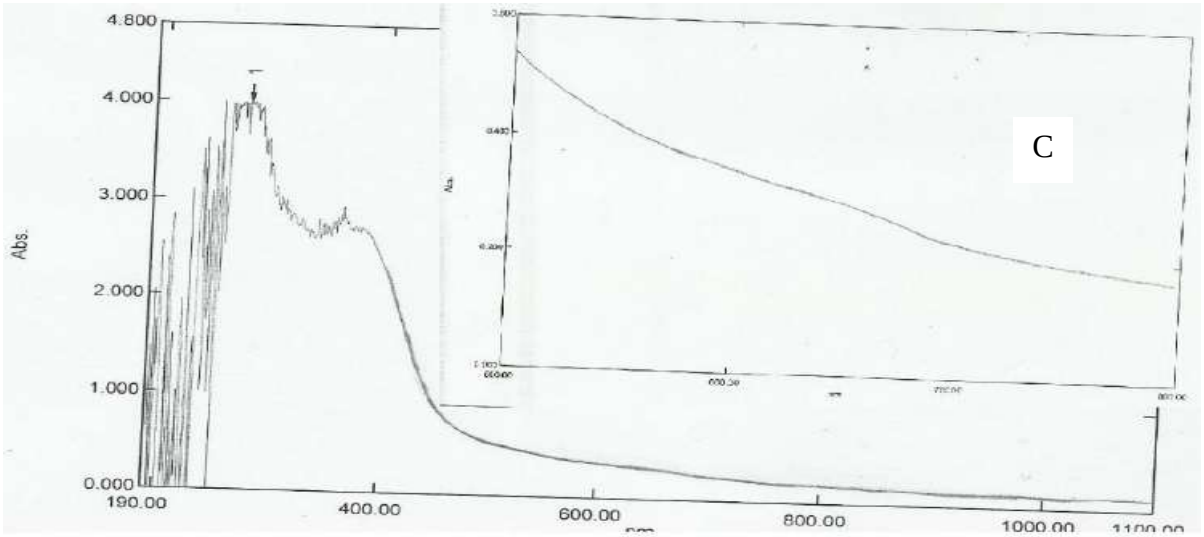



**Figure (3) TGA/DTA curves for :A( ligand(L)), B (CoLAla), C (Ni LAla), D (Ni LGly), E (CuLAla**

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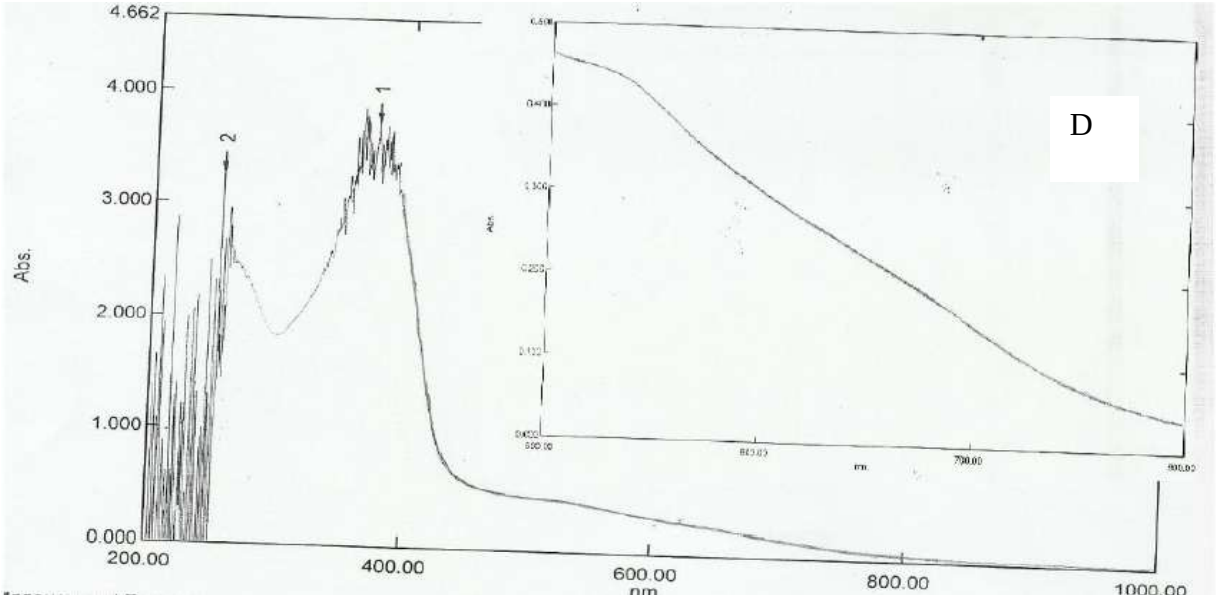
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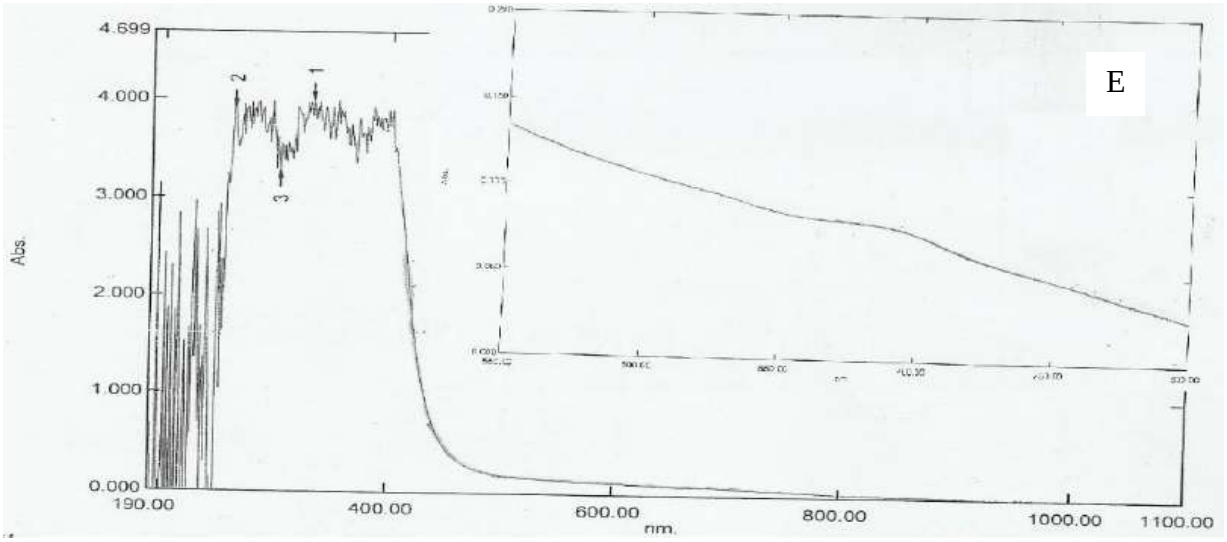
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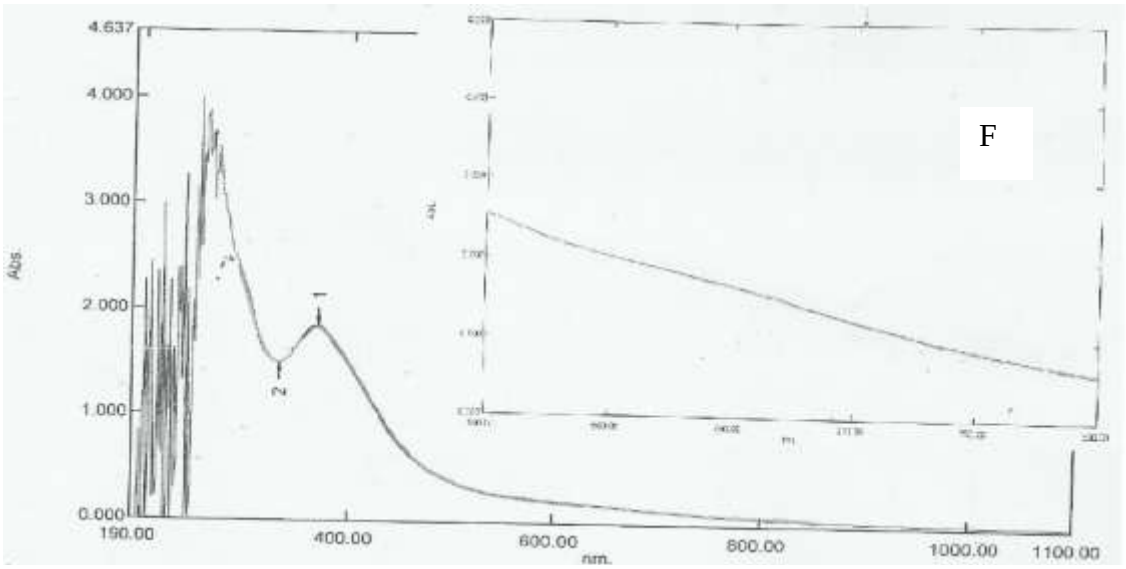


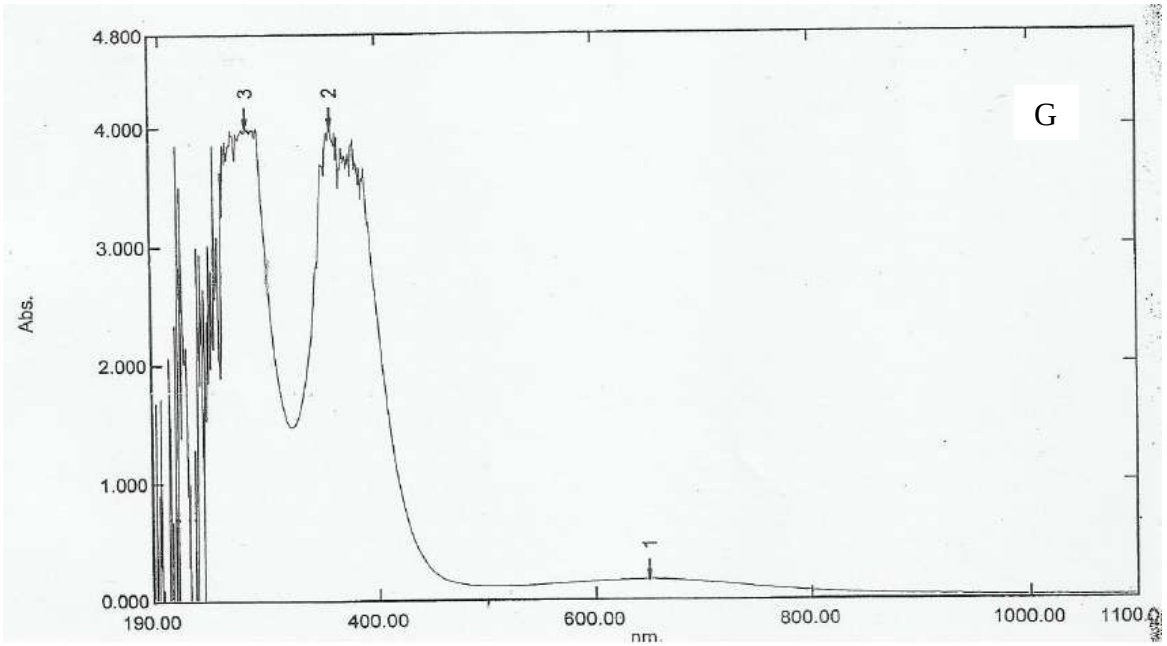


**Figure (4) UV-Vis spectra for :A( ligand(L)), B (CoLAla), C (Ni LAla), D (Cu LAla),E (Co LGly), F (Ni LGly), G (Cu LGly)**

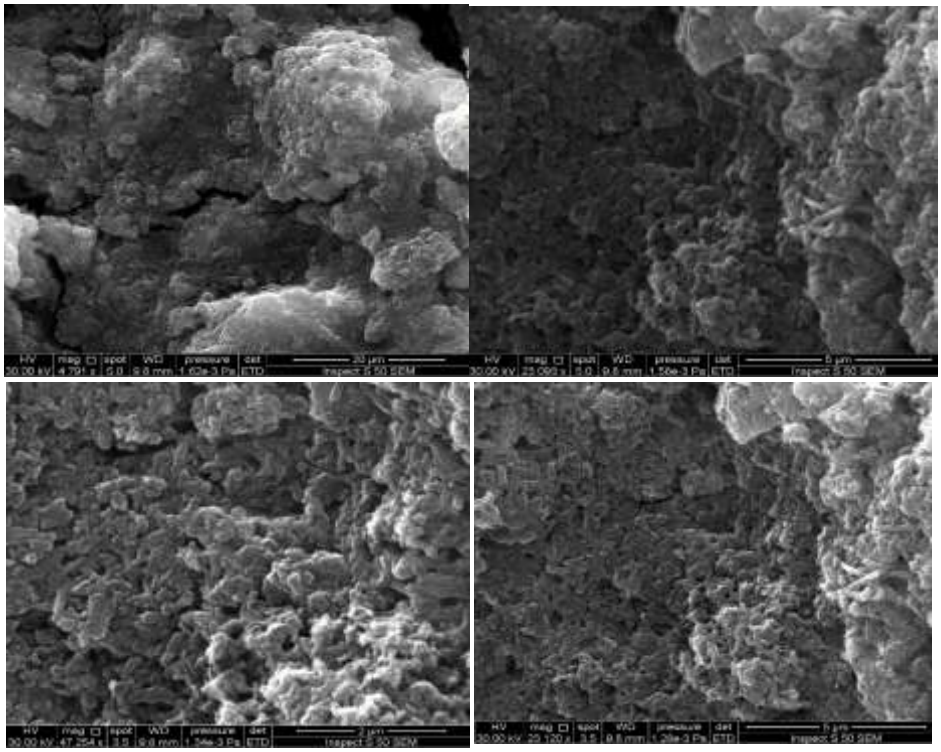

**Figure (5) SEM images for complex CuLAla at different**

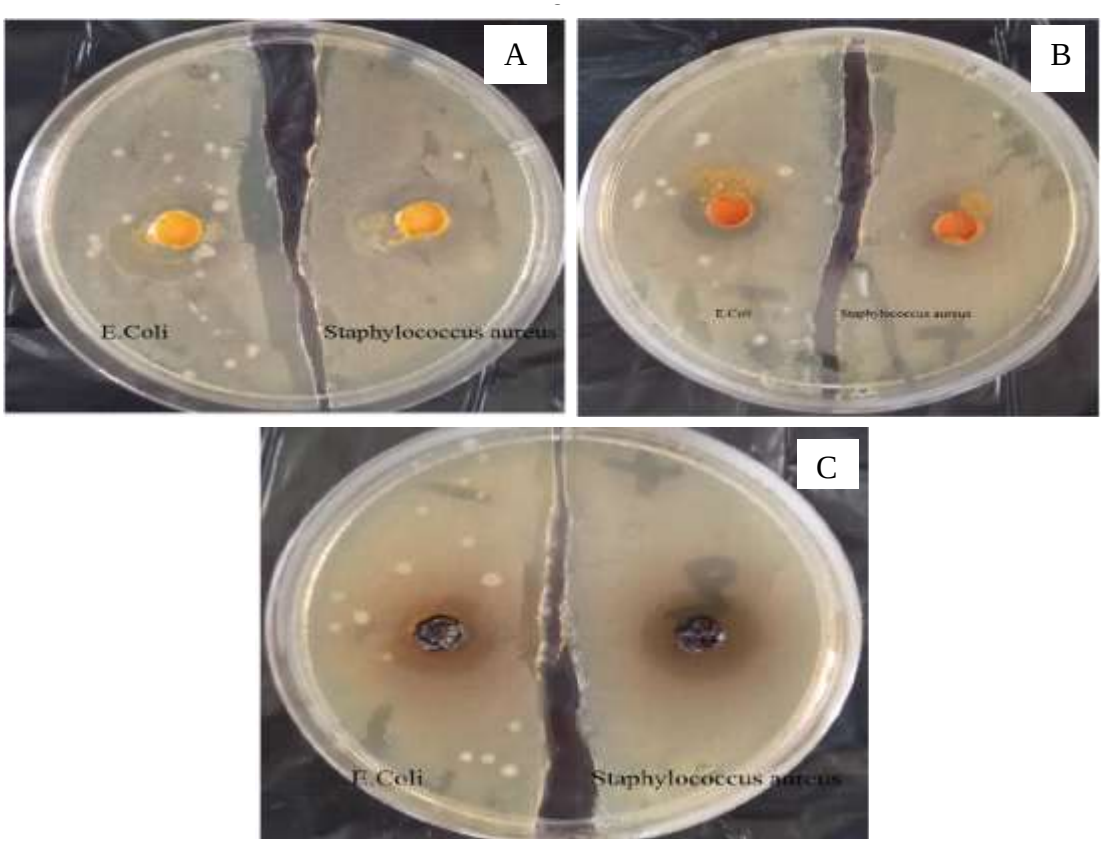


**Figure (6) Biological activity results for: A( ligand(L)), B (CoLAla), C (Co LGly)**