

Structural, Characterization, Biological Activity And Thermal Study Of New Complexes [Ni II, Hg II And La III] From Mixed Ligands (Curcumin And Azo Compounds) Type N₃O₂

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Abstract:

4-methoxyaniline reaction with 2-aminobenzothiazol in cold concentrated hydrochloric acid with 10% NaNO₂ to produce compound [L]. Compound [L] was reacted with curcumin and some transition metals complexes [Ni(II), Hg(II) and La(III)] in EtOH to yield new complexes. The structure of synthesized complexes described via FTIR , UV-Visible Spectroscopy, TGA-DSC ,A.A., chloride content, conductivity and the elemental analysis (CHNS). Complexes showed biological activity towards the E-coli (G-), Pseudomonas (G-), S. aureus (G+), Proteus (G-) and fungi. Based on the results that have been obtained from the above approaches, the proposed geometrical structures for every prepared complex have been of the octahedral formula.

Key-words: Azo compound , curcumin , Aniline.

Introduction

Aniline (nitrogen compound) is an organic compound with the formula C₆H₅NH₂. Consisting of a phenyl group devoted to an amino group, aniline is the humblest aromatic amine. It is an industrially significant commodity chemical, as well as a versatile starting material for fine chemical synthesis^[1-5]. Many chemicals including π-electron either in (≡) bond or conjugated (=) bond and hetero-atoms: oxygen, nitrogen, sulphur and phosphorous were studied as metal erosion inhibitors^[6,7]. Among these, several 2-aminobenzothiazol, methoxyaniline and curcumin were reported as inhibitors of erosion and found to have good erosion inhibition effect^[8-10]. Nitrogen compounds have their uses in the rubber industry to process the rubber chemicals and products like car tyres, gloves, balloons, etc. It is also used as a dyeing agent for the manufacturing of clothes like jeans, etc. It is used for the production of drugs, for example, paracetamol, acetaminophen, and Tylenol^[11-14]. Aniline is prepared commercially by the catalytic

hydrogenation of nitrobenzene or by the action of ammonia on chlorobenzene. The reduction of nitrobenzene can also be carried out with iron borings in aqueous acid. A primary aromatic amine, aniline is a weak base and ^[15-17].

2. Experimental

2.1.chemicals: chemicals have been supplied from flucka and Merck.

2.2.Instrumentation : Thermal analysis TGA-DSC were analyzed and characterized. FTIR-Spectra that has been recorded on a Shimadzu – 8400s using potassium bromide disk . The C.H.N.S. were performed on an European Elemental .

2.3. Preparation of facilities:

2.3.1.preparation of 2-(benzo [d]thiazol-2-yl diazenyl)-4-methoxyaniline

In a round bottom flask , 2-aminobenzothiazol (0.002mol) in mixture contained [(10 ml) ethanol, (10 ml) distilled water and (2 ml) HCl and cooled the mixture to (0-5)°C for 30 minutes then add mixture slowly and constant stirring for 4-methoxyaniline (0.002mol) in sodium hydroxide solution pH(5-6), then filtration and recrystallized and dried by anhydrous CaCl₂ to yield green precipitate , yield (30%) , M.P.=196-199 °C.

2.3.2.preparation of Metal Complexes [Ni(II),Hg(II) and La(III)]with ligand (L) and curcumin

The (1:1:1) chelate complexes metal, ligand(L) and curcumin have been synthesized via dissolving (8mmole) (L) in (10ml) of the absolute solvent, then mixed with solution containing metal chloride salts of (NiCl₂.6H₂O, HgCl₂ and LaCl₃) dissolved in the absolute ethanol (10 mL) and (8mmole)Curcumin dissolved in (20 ml) ethanol. The mixture refluxed a period (3hrs.) on water bath, on the cooling of contents, complexes have been separated out. The creation has been filtered, lapped by the ethanol and dried under the vacuum.

3. Results and Discussion

3.1. FT-IR of complexes

All complexes have been synthesized by reaction of [L] with curcumin and metal salt in ethanol. FT-IR of complexes [Ni(cur)(L)(H₂O)]Cl, [Hg(cur)(L)(H₂O)]Cl, [La(cur)(L)(Cl)]Cl^[18-21].

Table (1): The characteristic infrared bands compounds

Comp.	Color++99	M.P.	$\nu(\text{O-H})$	$\nu(\text{NH}_2)$	$\nu(\text{C=O})$	$\nu(\text{N=N})$	$\nu(\text{M-O})$	$\nu(\text{M-N})$
L	Brown		-----	3387 3441	---	1504	--	---

Cur.	orange	183-185	3502-3200	----	1627	----	--	--
L+ Cur.+Ni	Dark brown	>300	3392	3039 2937	1595	1508	472	550
L+cur.+ Hg	Red-brown	>300	3365	2835 2358	1591	1508	474	542
L+Cur.+ La	Dark yellow	>300	3379	2929 2839	1500	1400	462	520

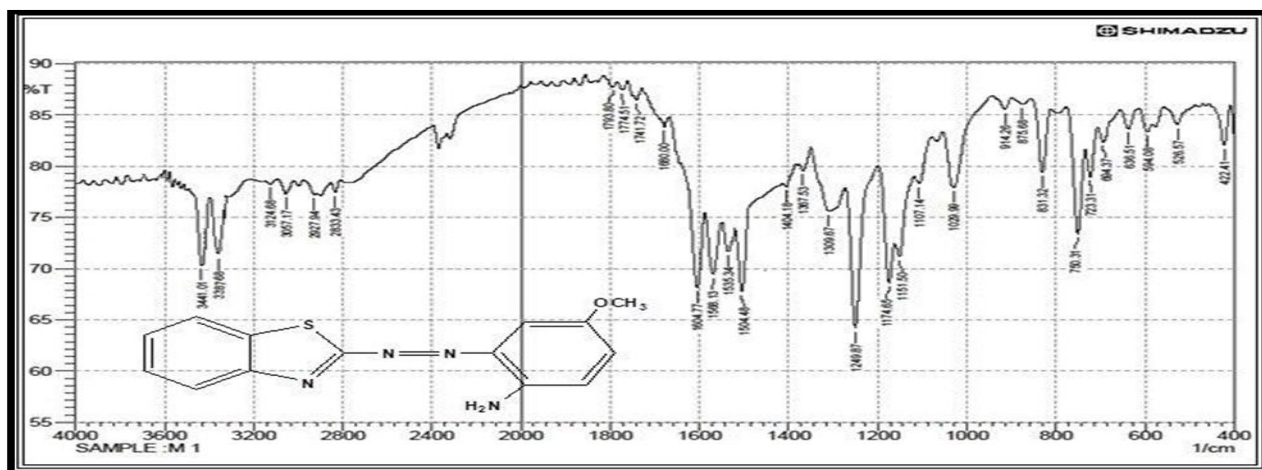


Fig1. FTIR of ligand (L)

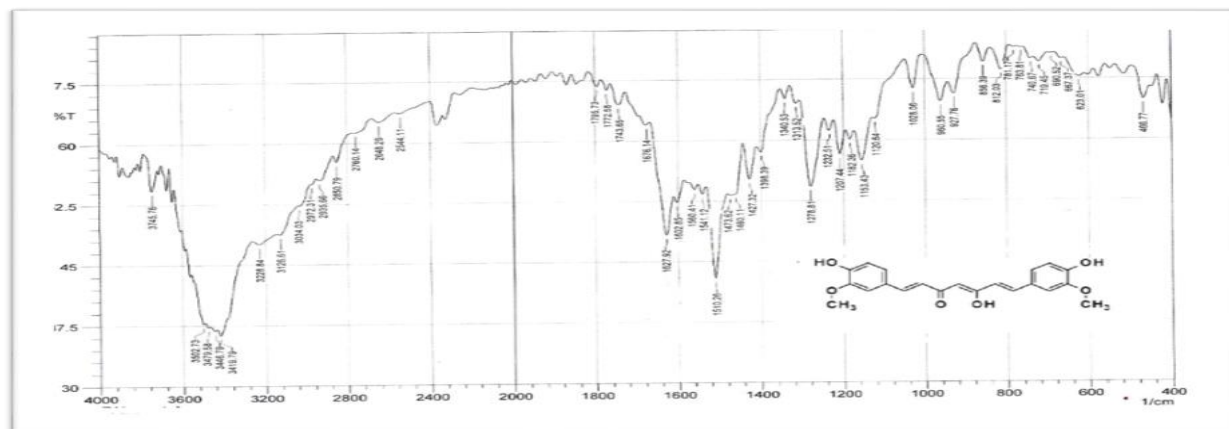


Fig2. FTIR of curcumin

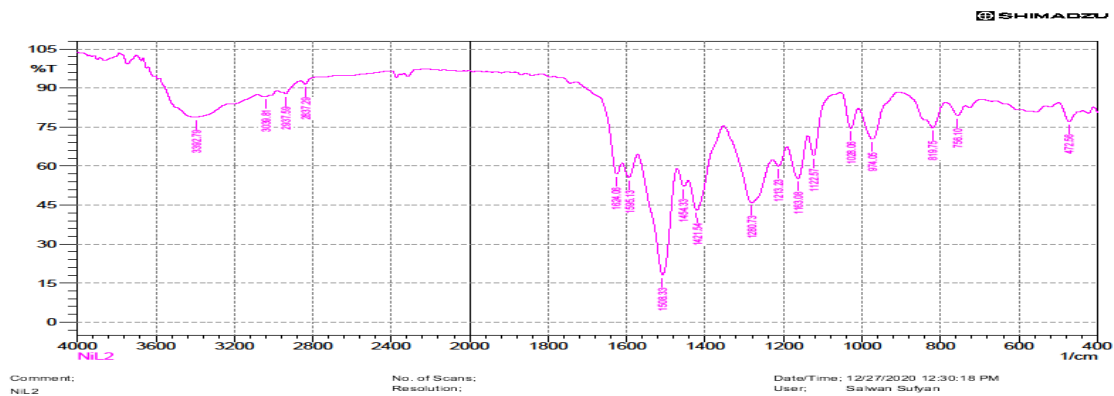


Fig3. FTIR of $[\text{Ni}(\text{cur})(\text{L})(\text{H}_2\text{O})_2]\text{Cl}$

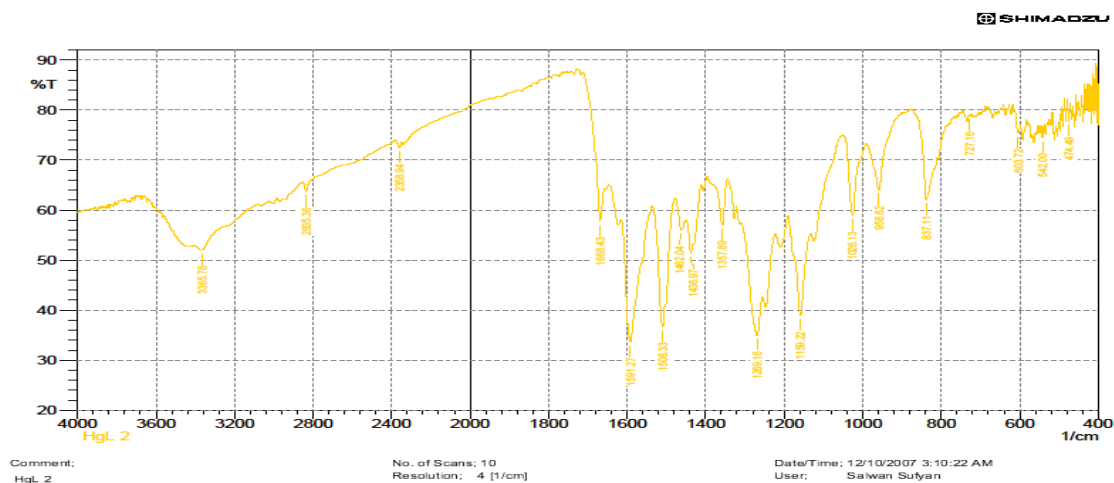


Fig.(4) FT-IR of $[\text{Hg}(\text{cur})(\text{L})(\text{H}_2\text{O})_2]\text{Cl}$

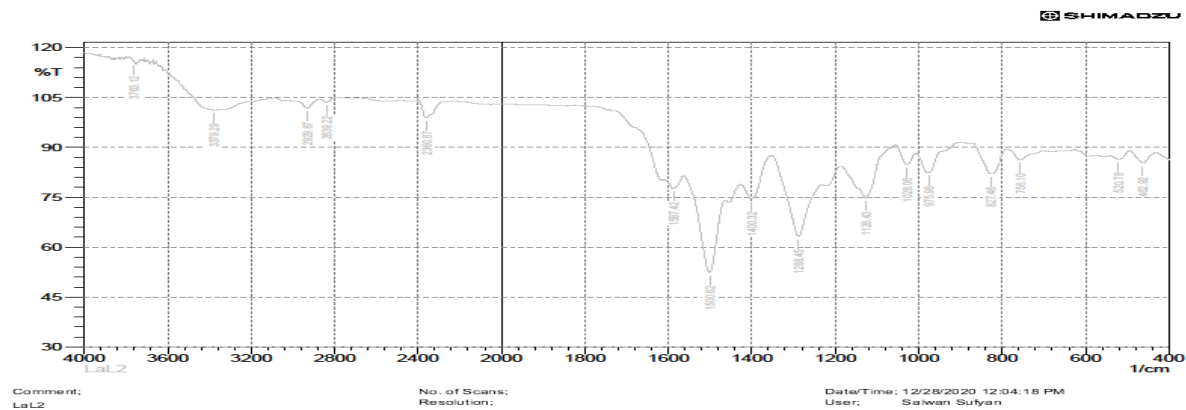


Fig.(5) FT-IR of [La(cur)(L)Cl]Cl

3.2.UV-Visible data for the complexes :

The UV-Visible of ligand [L] and curcumin fig.(6,7)spectra characterised mainly by two peaks of absorption at (233nm, 261nm)and (268 nm , 334nm) assigned to ($\pi \rightarrow \pi^*$) and ($n \rightarrow \pi^*$)respectively. Those electronic transitions have been lifted near higher or lower frequencies in the electronic spectra of every primed complex, confirm the ligand's coordination with ions of the metal^[22-25]

Table (2) the electronic results of compounds and conductivity

Compounds	conductivity	λ (nm)	ν - (cm^{-1})	ϵ_{max} L/mol.cm	Transition
Curcumin	-----	268	37313	530	$\pi \rightarrow \pi^*$
		334	29940	538	$\pi \rightarrow \pi^*$
		434	23041	2065	$n \rightarrow \pi^*$
L	-----	240	41700	1038	$\pi \rightarrow \pi^*$
		304	32900	661	$n \rightarrow \pi^*$
		383	26100	2306	$n \rightarrow \pi^*$
[Ni(cur.)(L)(H ₂ O)]·Cl	1:1	320	31300	798	$\pi \rightarrow \pi^*$
		420	23800	879	$^3A_{2g} \rightarrow ^3T_{1g}(P)$
		444	22500	758	$^3A_{2g} \rightarrow ^3T_{1g}$
		686	14600	2	$^3A_{2g} \rightarrow ^3T_{2g}$
[Hg(cur.)(L)(H ₂ O)]·Cl	1:1	264	37900	769	$\pi \rightarrow \pi^*$
		422	23700	1448	Charge transfer
[La(cur.)(L)(Cl)]·Cl	1:1	268	37300	1726	$\pi \rightarrow \pi^*$
		417	24000	1192	Charge transfer

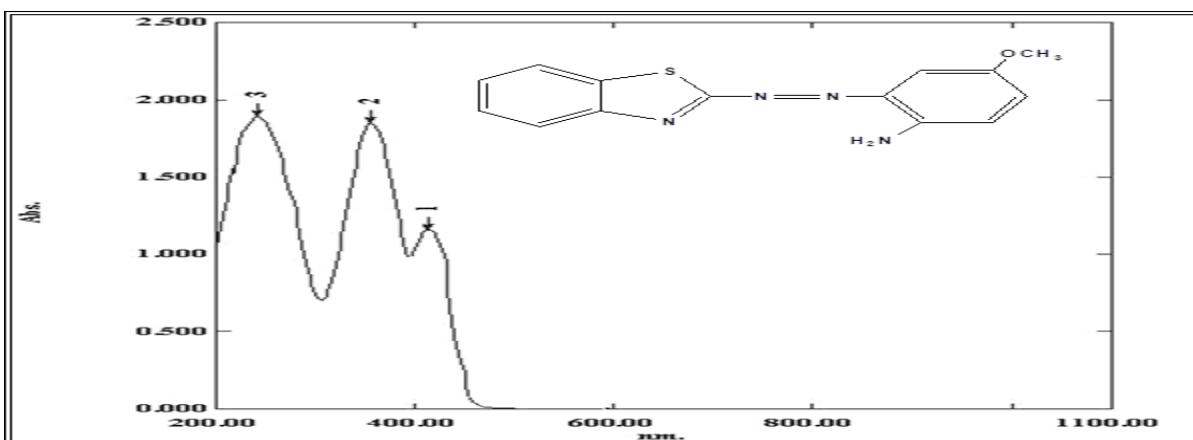


Fig6. The electronic spectrum of ligand L

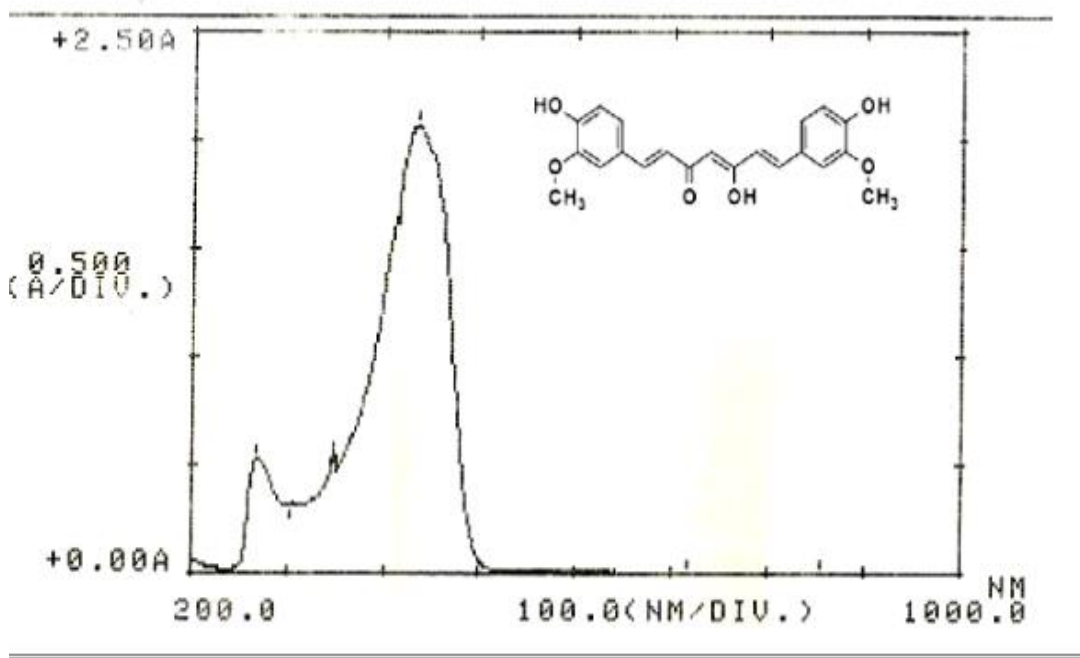


Fig7. the electronic spectrum of Curcumin

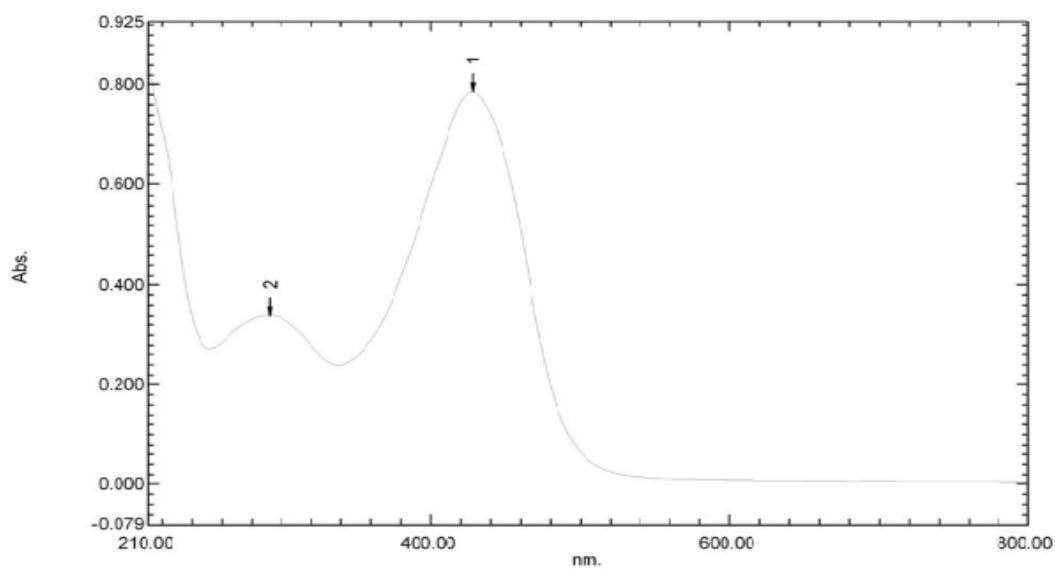


Fig8. Electronic spectrum of [Ni(cur)(L)(H₂O)]Cl

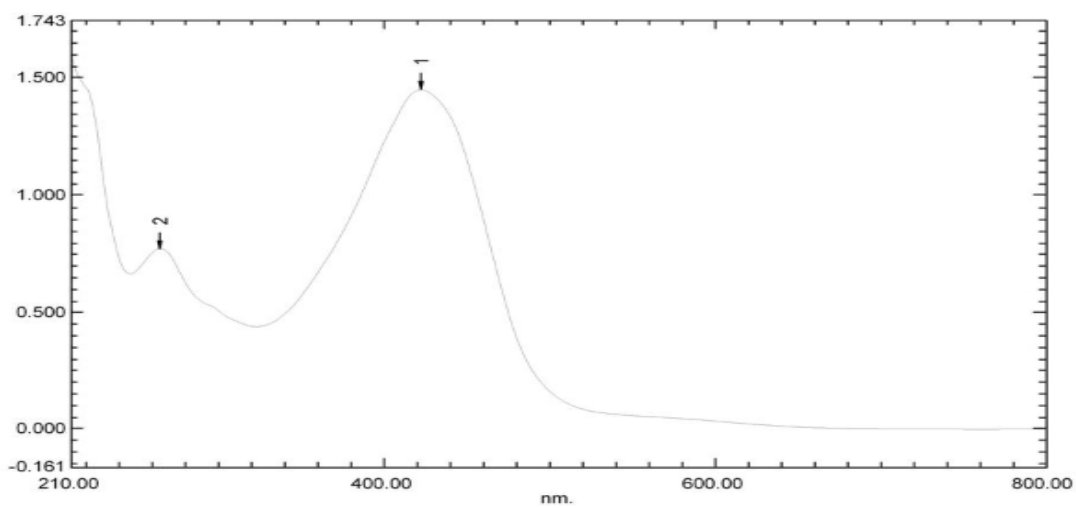


Fig.(9) the electronic spectrum of [Hg(cur)(L)(H₂O)]Cl

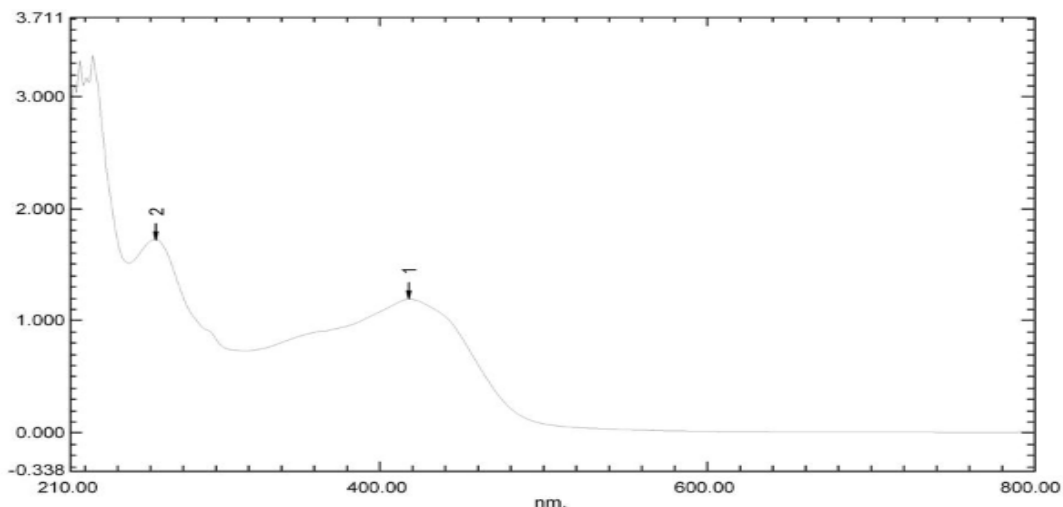


Fig10. Electronic spectrum of [La(cur)(L)(Cl)]Cl

3.3. Thermal Decomposition of the ligand (L) and [Ni(cur)(L)(H₂O)]Cl

Table (3) thermal analyses of ligand (L) and [Ni(cur)(L)(H₂O)]Cl

Complexes	Stage	Decomposition Temperatures Initial-Final (°C)	Estimated (i.e. Computed)		Assignments
			Mass Loss	Total mass Loss	
L	1	12.8-287.0	4.200 (4.214)	18.07 (18.12)	-(C ₃ H ₅ O)
	2	287.0-313.7	5.850 (5.879)		-(C ₄ H ₃ N ₂)
	3	313.7-524.4	8.020 (8.032)		-(C ₅ H ₂ NS)
[Ni(cur.) (L)(H ₂ O)]Cl	1	75-323.99	4.67 (4.68)	7.205 (7.209)	-(C ₉ H ₁₆ O ₄ Cl)
	2	323.99-489.24	2.752		-(C ₅ H ₁₁ N ₂ O ₂)

			(2.757)	
	3	489.24-595.38	1.366 (1.369)	-(C ₃ HN ₂)

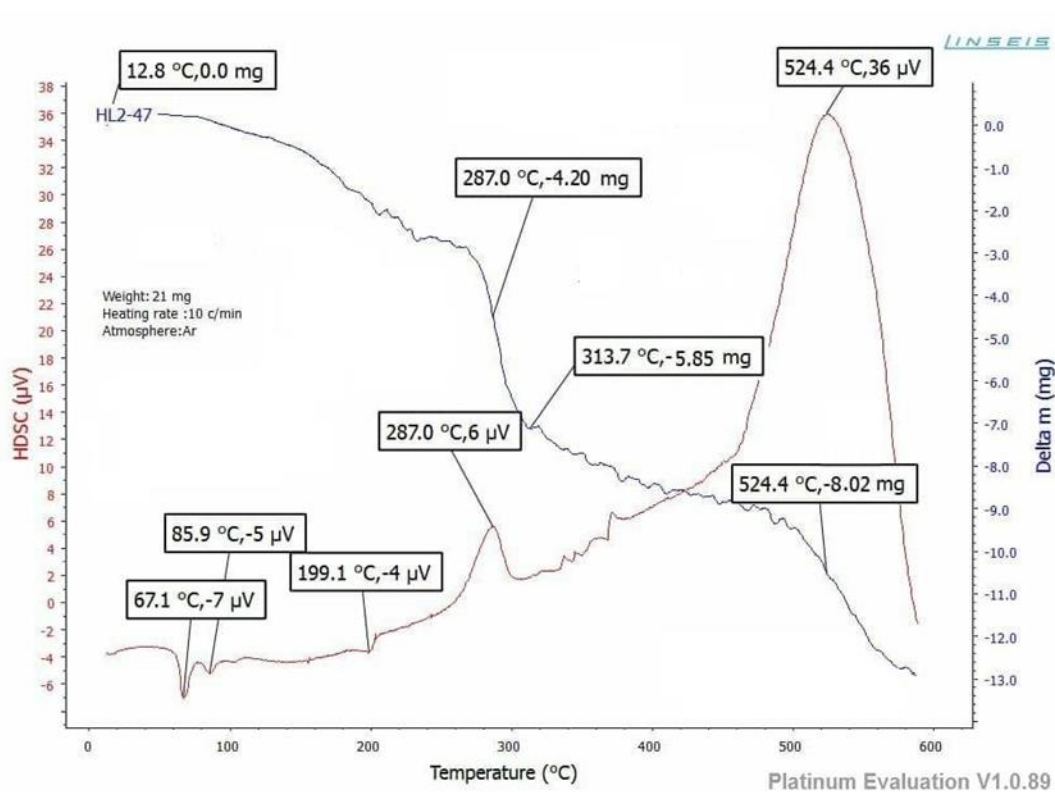


Fig11. thermal analysis of the ligand (L)

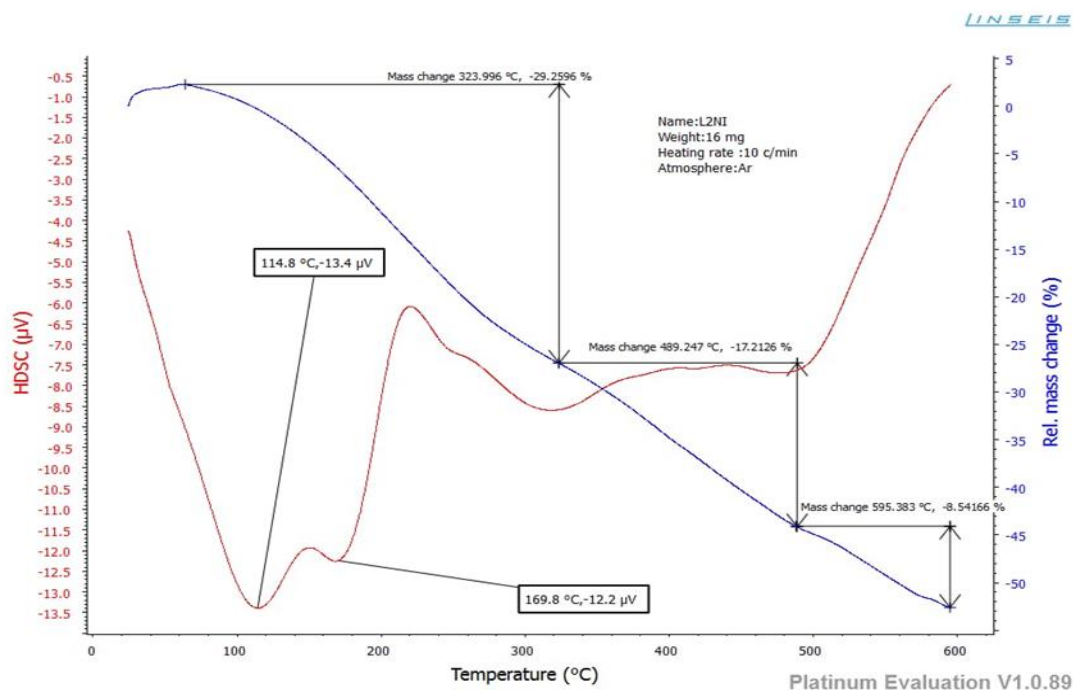


Fig12. thermal analysis of the $[\text{Ni}(\text{cur})(\text{L})(\text{H}_2\text{O})]\text{Cl}$

3.4. Biological screening: The test of the anti-bacterial activity

In this work, the synthesized compounds were checked for the anti-bacterial activity against the strains of E-coli(G-), S. aureus(G+), Proteus(G-) and Pseudomonase(G-) by the approach of the agar diffusion [26-28]. Every compound has been dissolved in the ethanol for the purpose of giving a final 0.001mg/ml concentration, and from data listed in Table4, every compound has shown a biological activity against the four types of the bacteria, except Ni-complex with Pseudomonase has no biological activity [inhibition zone=0].

Table4. Biological activity of synthesized compounds

Compounds	S.aureus (G+)	E-coli (G-)	Pseudomonas (G-)	Proteus (G-)
L	3	2	2	3
Ni-complex	5	4	0	3
Hg-complex	17	7	15	9
La-complex	8	4	9	7
Control	2	7	11	5

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