

Synthesis, Characterization and Antibacterial Activity of Mono- and Binuclear Co(II) and Cr(III) Mixed-Ligand Complexes Derived from α -Amino Acid and 1,10-Phenanthroline with 1,3-Diaminopropan as a Spacer

Esraa S. Mahdi^{1,*}, Ammar J. Alabdali²

Abstract

Mixed-ligand complexes of Co(II) and Cr(III) ions were synthesized from the reaction of metals chloride salts with α -amino acid (H_2L1) as a primary ligand and 1,10-phenanthroline ($L2$) as secondary ligand. The ligand 2-(benzo[d]thiazol-2-ylamino)-2-(2-hydroxynaphthalen-1-yl) acetic acid (H_2L1) has been synthesized from the condensation reaction of aminobenzothiazol with 2-hydroxy-1-naphthaldehyde in the presence of HCN as three components–one pot reaction. H_2L1 considered as a tridentate chelater of ONO-coordination binding type toward the metals, while $L2$ considered as bidentate chelater of NN-coordination binding type toward the metals. The suggested formula for the mononuclear complexes were $[Co(L1)(L2)H_2O]$ and $[Cr(L1)(L2)Cl]$, and the formula of the binuclear complexes are: $Na_2[Co_2(L1)_2(L2)_2(L3)]$, and $[Cr_2(L1)_2(L2)_2(L3)]$ where $H_2L3 = 1,3$ -diaminopropan which is considered as a bridging ligand or spacer linkage between metals of mononuclear complexes. The obtained compounds were characterized by different spectroscopic techniques like FTIR, 1H -NMR, ^{13}C -NMR, UV-Vis, TGA, melting point and conductance measurement, and the geometry was found to be a distorted octahedral. In vitro antibacterial activity of the ligands, salts, and metal complexes was tested against two pathogenic bacteria: *Pseudomonas* (gram negative) and *Escherichia coli* (gram positive), using the well diffusion method.

Keywords: α -Amino nitrile, α -amino acid, mononuclear, binuclear, mixed-ligand complex, spacer

INTRODUCTION

One of the most interesting features of transition metals is their ability to synthesize coordination molecules [1]. The presence of chelators provides the complexes an extra degree of stability, which is especially helpful in cases when the ring has five or six members. Molecular structures known as coordination complexes are distinguished by the presence of covalent bonds that are coordinated to metal ions [2, 3]. During the past several decades, transition metal complexes have played an essential part in the field of chemotherapeutics, namely in the diagnosis and treatment of cancer [4], and to understand the chemistry of metal-drug complexes, especially in the process of developing more biologically active treatments [5]. 1,10-Phenanthroline is a bidentate, heterocyclic diamine which is widely used as a chelating agent for transition metal ion, and the resultant complexes have played an important role in the development of complexes [6]. 1,10-phenanthroline is a rigid planar, hydrophobic, electron-poor heteroaromatic

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compound. It has two nitrogen donor atoms at 1- and 10-position [7], therefore it is an excellent chelating ligand for the formation of metal complexes due to the presence of lone pairs of electrons on the nitrogen atoms, which, when paired with the rigidity of the aromatic ring structure, results in the phen molecule. The propensity of heteroaromatic nitrogen atoms to function as poor donors is compensated for by the ability of phenanthroline to behave as outstanding acceptors [8]. The compound 1,10-phenanthroline and many of its derivative's complexes do have antibacterial effects [9, 10]. Cobalt +2 and +3 oxidation states compounds are categorized as coordination complexes, which are molecules or ions with cobalt attached to a number of ligands, As "oxygen carriers," complexes made of metals like cobalt and Schiff bases have been studied [11].

For more than 40 years, Cr(III) has been studied for its biochemical and nutritional properties, but no consensus has been reached on its necessity, application, or mechanism of action. Significant results and theories have been established in more recent studies of the function of Cr(III) [12].

Mixed-ligand complexes were put through a series of tests to determine the antibacterial activity against a variety of pathogens. The antibacterial activity of binary complexes was shown to be lower when compared to that of their equivalent tertiary complexes; this is because the complexes have a larger planar area, as well as technologies that make stacking stronger than it would be. Otherwise, changes in the antimicrobial activity of various complexes against harmful bacteria and fungus may be attributed to either differences in the ribosomes found in microbial cells or the impermeability of the cells that make up the microorganisms [13].

Antibiotic overuse has resulted in a rise of bacteria that are resistant to commonly used antibiotics. Researchers are discovering novel active compounds to overcome this problem of microbial resistance to antibiotics. Two mononuclear and binuclear mixed ligand complexes of Co(II) with 1,10-phenanthroline, acetamide and ethylenediamine, were synthesized to have octahedral geometry. In vitro antibacterial activity was tested on two Gram positive (*Staphylococcus aureus*, *Streptococcus pyogenes*) and two Gram negative (*Escherichia coli* and *Klebsiella pneumoniae*) bacteria using disc diffusion method. The complexes performed good against the Gram-negative bacteria. Even at lower concentrations of complexes, the antibacterial effect was found to be identical to the commercial drug gentamicin. Thus, these complexes could be used as an alternative to metal-based antibiotics after in vivo cytotoxicity testing [14].

MATERIALS AND METHODS

Chemicals

The chemicals used in this work obtained from different companies and without further purification. Acetic acid glacial 99% (Analar), amino benzothiazol 98% (Alfa Aesar), chromium(III) chloride hexahydrate, cobalt (II)chloride hexahydrate 98% (BDH), 1,3-diaminopropane 98% (Merck), 1,10-phenanthroline 99% (Riedel-de Haenagseeleze-Hannoverand) and 2-hydroxy 1-naphthaldehyde 98% (Sigma Aldrich).

Instruments

The infrared (IR) spectra were recorded using Fourier transform infrared (FTIR), Bruker Alpha, in the wave number range 400 to 4000 cm^{-1} . Thermogravimetric analysis (TGA) was performed with Shimadzu Analyzer maintained at 6000°C. Proton nuclear magnetic resonance ($^1\text{H-NMR}$) and $^{13}\text{C-NMR}$ spectra were recorded on a Bruker NMR spectrophotometer model ultra-shield at 400 MHz, using DMSO-d₆ as solvent. UV-Vis spectra were recorded with Shimadzu UV-Vis 1600 in a 10 mm quartz cell in the range 200 to 1000 nm and molar conductivity of 10^3 M of the prepared complexes in DMSO as a solvent, using Corning Conductivity Meter 220 Inluba WTW balance.

Methods

Synthesis of primary ligand ($\text{H}_2\text{L1}$) (Figure 1):

A total of 1 mmol (0.17 g) of 2-amino benzothiazole was added to 1 mmol (0.15 g) of 2-hydroxy-1-naphthaldehyde which was dissolved in 3.5 mL of glacial acetic acid and the reaction mixture was refluxed for 2 hours with stirring, followed by addition of 2 mmol (0.06 g) of KCN, then an excess of 1.5 mL of glacial acetic acid (HAc) was added. The reaction mixture was stirred for 1 hour, α -amino nitrile was produced, which hydrolyzed by adding 10 mL of 10% H_2SO_4 with reflux and stirring (one-pot reaction). The solution was neutralized using ammonia solution under cooling by ice then left overnight. The precipitate was formed, collected by filtration, and washed with distilled water followed by overnight oven drying. The light brown α -amino acid (H_2L1) was collected. The melting point of H_2L1 was $127^\circ C$ to $130^\circ C$ and yield percentage was 73.5%. The scheme of the reaction path is presented in Figure 1.

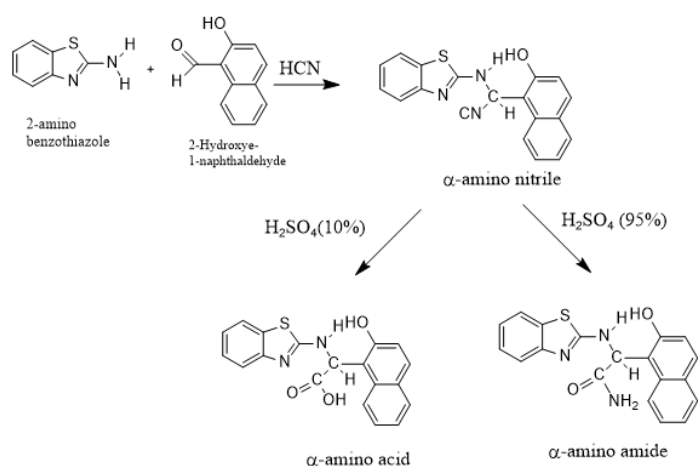


Figure 1. Synthesis of mononuclear mixed ligand complexes.

Two equivalents of sodium metal dissolved in 10 mL alcoholic basic medium (C_2H_5OH/C_2H_5ONa) to be ready to interact with one equivalent of α -amino acid H_2L1 . The primary ligand Na_2L1 was added into metal salt $CoCl_2 \cdot 6H_2O$ or $CrCl_3 \cdot 6H_2O$ with stirring. The secondary ligand 1,10-phenanthroline (L2) was added directly to the mixture with reflux for 4 hours to synthesize mononuclear complexes $[Co(L1)(L2)H_2O]$ and $[Cr(L1)(L2)Cl]$, respectively. The mixture was cooled to room temperature, and precipitated solids were filtered and washed with absolute ethanol to remove any unreacted ligands. Melting points were $97^\circ C$ to $100^\circ C$ and $<36^\circ C$, respectively, and yield percentages were 72.1% and 69.5%, respectively. The reaction scheme is shown in Figure 2.

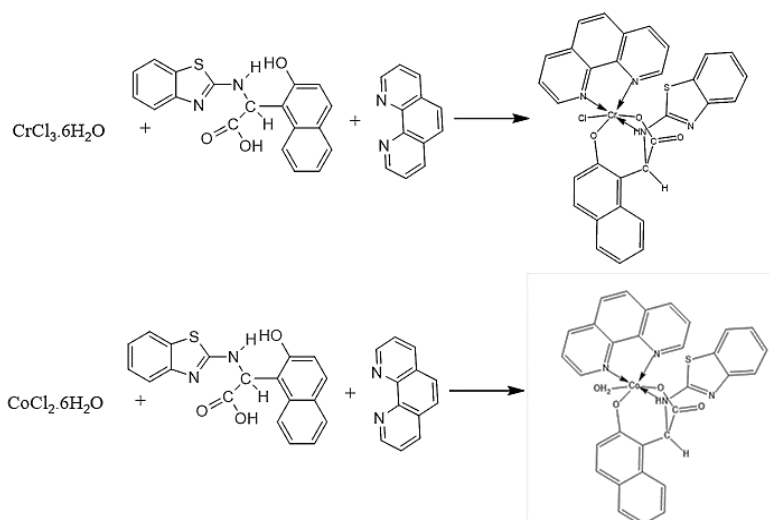


Figure 2. Synthesis of binuclear mixed ligand complexes.

Two mmoles of mononuclear complexes $[\text{Co}(\text{L1})(\text{L2})\text{H}_2\text{O}]$ or $[\text{Cr}(\text{L1})(\text{L2})\text{Cl}]$ was dissolved in 10 mL of hot absolute ethanol, 1 mmol of propylene di amine ($\text{H}_2\text{L3}$) was added directly to the complexes with reflux in water bath for 2 hours to synthesize binuclear complexes $\text{Na}_2[\text{Co}_2(\text{L1})_2(\text{L2})_2(\text{L3})]$ and $[\text{Cr}_2(\text{L1})_2(\text{L2})_2(\text{L3})]$, respectively. The complexes were allowed to cool at room temperature and kept aside for slow evaporation, crystals were collected washing by cold ethanol and filtration then dried in a desiccator. Melting points were 102°C to 105°C and 97°C to 100°C , respectively and yield percentages were 66.4% and 60.7%, respectively. The reaction scheme is shown in Figure 3.

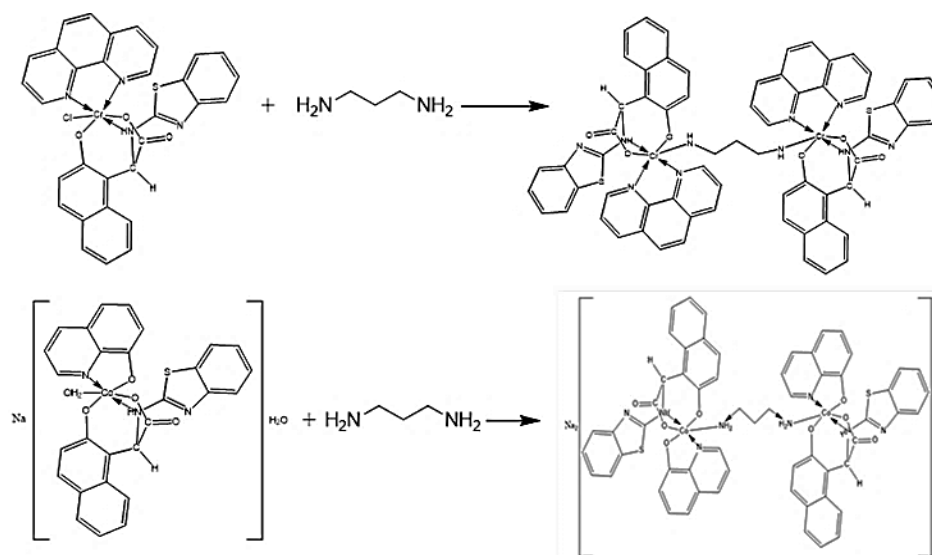


Figure 3. Synthesis of binuclear complexes.

RESULTS AND DISCUSSION

Chemical Study

Chemical studies helped to characterize the prepared ligands and complexes. Several methods are employed. These techniques are FT-IR, TGA, $^1\text{H-NMR}$, $^{13}\text{C-NMR}$, UV-Vis and conductivity measurements. These techniques were used to identify the various natures of chemical bonds that are present in the chemical structure of ligands and complexes.

Infrared Spectroscopy

FTIR spectra help in getting an idea about how the ligand is coordinated with the metal ion. The characteristic frequencies of the expected functional groups are presented in Table 1.

Table 1. The characteristic absorption bands of the ligand and complexes

Comp.	$\nu\text{O-H}$			$\nu\text{N-H}$		$\nu\text{C-H}$ Arom.	$\nu\text{C-H}$ Aliph.	$\nu\text{C=O}$ Carboxyl	νCO_2^- Carboxylate		$\nu\text{C=C}$ Aromatic
	Acid	Alcohol	Water	Amino acid	Diamine				Assym.	Sym.	
L1	3388	3340	–	3269	–	3054	2930	1692	–	–	1524 1444
$[\text{Co}(\text{L1})(\text{L2})\text{H}_2\text{O}]$.	–	–	3387	3200	–	3057	2970	–	1615	1374	1586 1508
$\text{Na}_2[\text{Co}_2(\text{L1})_2(\text{L2})_2(\text{L3})]$	–	–	–	3210	3327	3048	2956	–	1639	1389	1559 1476
$[\text{Cr}(\text{L1})(\text{L2})\text{Cl}]$	–	–	–	3200	–	3053	2971	–	1597	1376	1577 1516 1461
$[\text{Cr}_2(\text{L1})_2(\text{L2})_2(\text{L3})]$	–	–	–	3211	3329	3088	2921	–	1620	1384	1556 1471

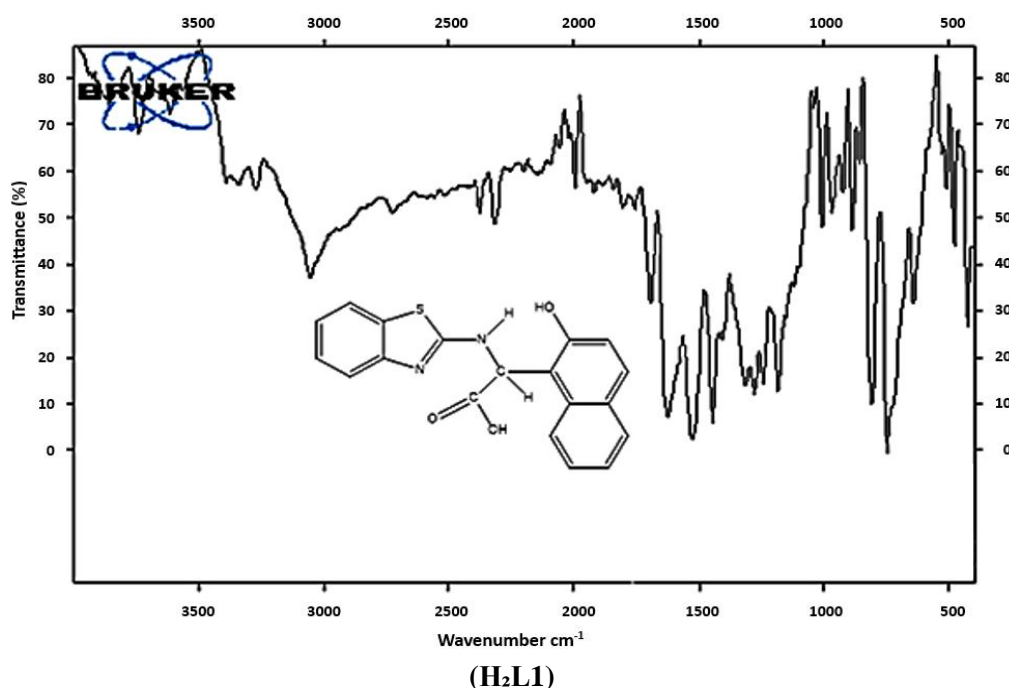
FTIR spectra of H₂L1 compound illustrated sharp peak of carbonyl group ν (C=O) at 1692 cm⁻¹ and appearance of weak band at 3388 cm⁻¹ of ν (-OH) group of carboxylic acid [15]. Bands at 3340 and 3269 cm⁻¹ were for naphthalenic (-OH) and amino (-NH) groups, respectively. The peaks at 1524 cm⁻¹ and 1444 cm⁻¹ represented stretching bands of ν (C=C) aromatic group [16]. The bands appearing at 3054 cm⁻¹ and 2930 cm⁻¹ were of ν (C-H) aromatic and aliphatic stretching, respectively.

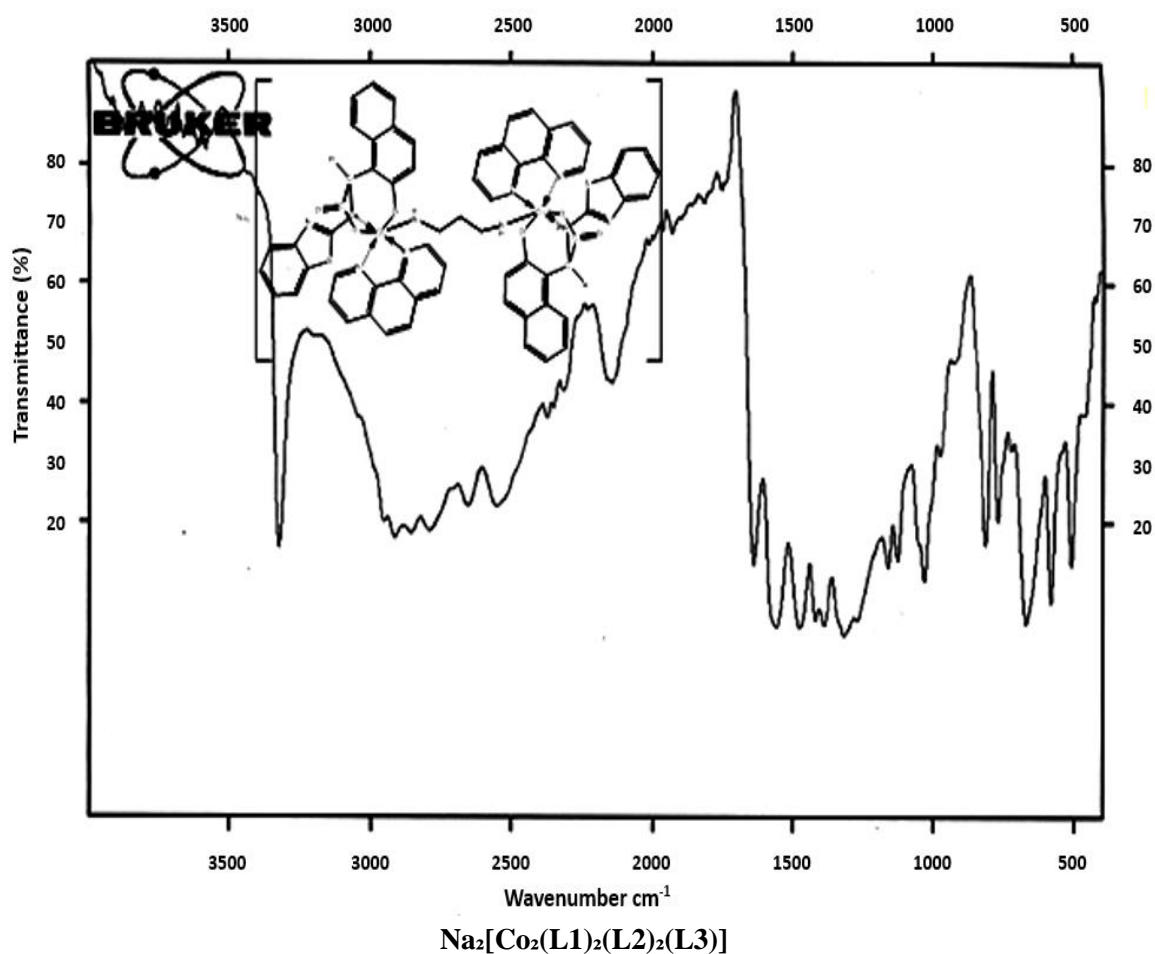
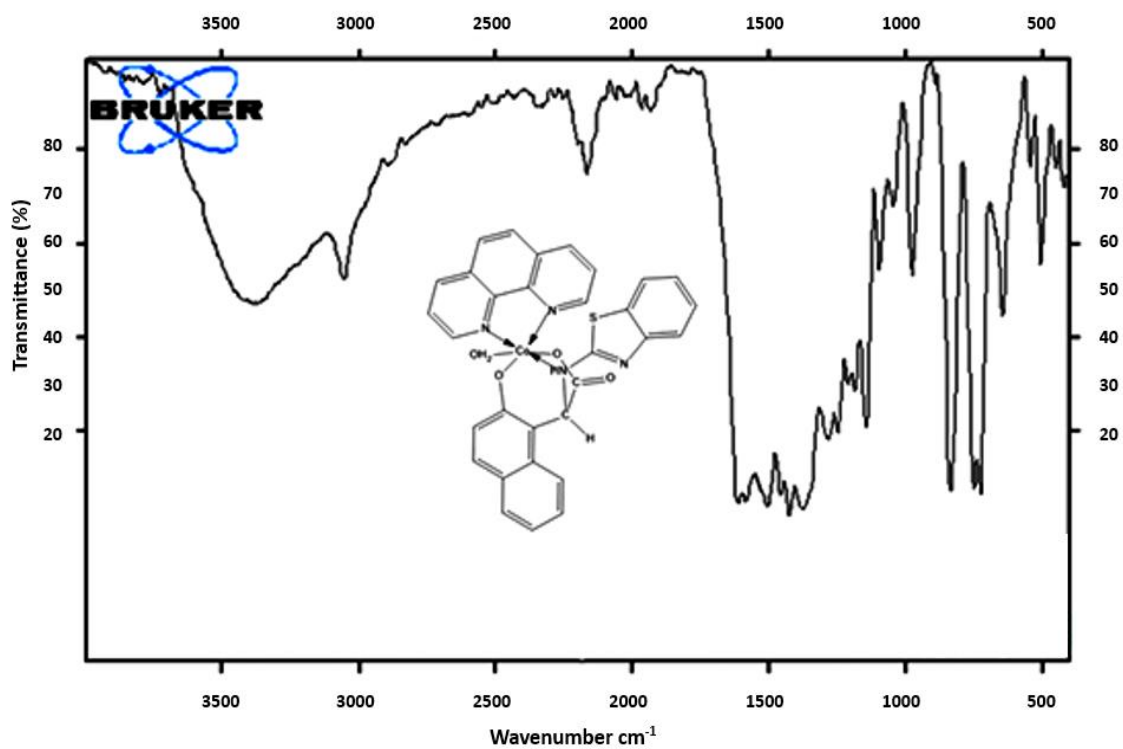
The infrared spectrum of [Co (L1) (L2) H₂O] showed the ν (N-H) bending band of amino acid shifting to low vibrational frequency at 3200 cm⁻¹, and there is a broad band at 3387 cm⁻¹ for lattice water molecule. The bands appearing at 1615 cm⁻¹ and 1374 cm⁻¹ belonged to asymmetric and symmetric ν (CO₂⁻), respectively. The aromatic and aliphatic of ν (C-H) showed at 3057 cm⁻¹ and 2970 cm⁻¹, respectively. The bands at 1586 cm⁻¹ and 1508 cm⁻¹ represented ν (C=C) aromatic groups.

The FTIR spectrum of Na₂ [Co₂ (L1)₂ (L2)₂ (L3)] shows a peak around 3210 cm⁻¹ which is due to ν (N-H) for amino acid and the band at 3327 cm⁻¹ belong to bridging ligand (L3), the aromatic and aliphatic bands of ν (C-H) appeared at 3048 cm⁻¹ and 2956 cm⁻¹, respectively. Furthermore, there are peaks observed around 1559 cm⁻¹ and 1476 cm⁻¹ which are due to ν (C=C) aromatic group. There is peak at 1639 cm⁻¹ that belongs to asymmetric carboxylate group ν (CO₂⁻) while the peak at 1389 cm⁻¹ is attributed to symmetric carboxylate group ν (CO₂⁻).

The FTIR spectrum of [Cr (L1) (L2) Cl] complex shows a peak at 3200 cm⁻¹ that corresponds to band of ν (N-H) group of amino acid. The bands of ν (C=C) aromatic group were at 1577 cm⁻¹, 1516 cm⁻¹ and 1461 cm⁻¹. The bands of ν (C-H) aromatic and aliphatic were found at 3053 cm⁻¹ and 2971 cm⁻¹, respectively. There are peaks at 1597 cm⁻¹ and 1376 cm⁻¹ that belong to asymmetric and symmetric carboxylate group ν (CO₂⁻), respectively.

The FTIR spectrum of [Cr₂ (L1)₂ (L2)₂ (L3)] showing a peak at 3211 cm⁻¹ illustrates shifting bands of ν (N-H) group of amino acid and the band at 3329 cm⁻¹ belongs to ν (-NH₂) group of bridging ligand. There are peaks at 1620 cm⁻¹ and 1384 cm⁻¹ that belong to asymmetric and symmetric carboxylate group ν (CO₂⁻), respectively. There are peaks observed around 1556 cm⁻¹, 1471 cm⁻¹ and 1421 cm⁻¹, which are due to ν (C=C) aromatic group. The aromatic and aliphatic bands of ν (C-H) appeared at 3088 cm⁻¹ and 2921 cm⁻¹, respectively. Infrared spectra for mono- and binuclear Co(II) and Cr(III) complexes are shown in Figure 4.





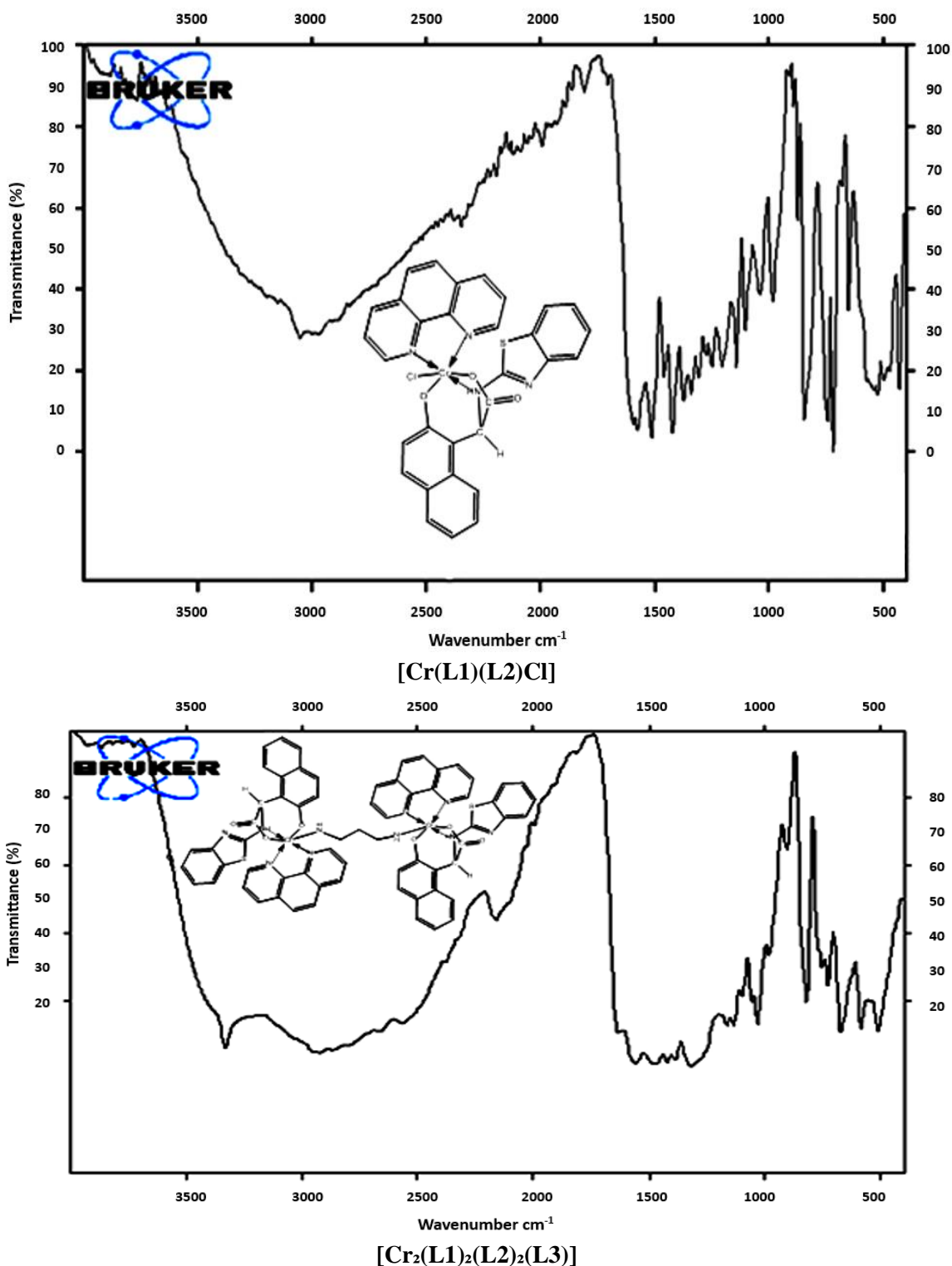


Figure 4. Infrared spectrum for mono and binuclear Co (II) and Cr (III) complexes

$^1\text{H-NMR}$ spectrum of $\text{H}_2\text{L1}$

The $^1\text{H-NMR}$ spectrum of prepared α -amino acid ($\text{H}_2\text{L1}$), Figure 5, shows multiplet signals at 7.00 to 8.00 ppm indicating C-H presence of phenyl groups, singlets signal at 3.37 ppm belongs to C-OH group of phenol, while doublet signal at 6.98 ppm is for N-H group and 13.08 ppm of O-H group of carboxylic acid of amino acid compound [17, 18].

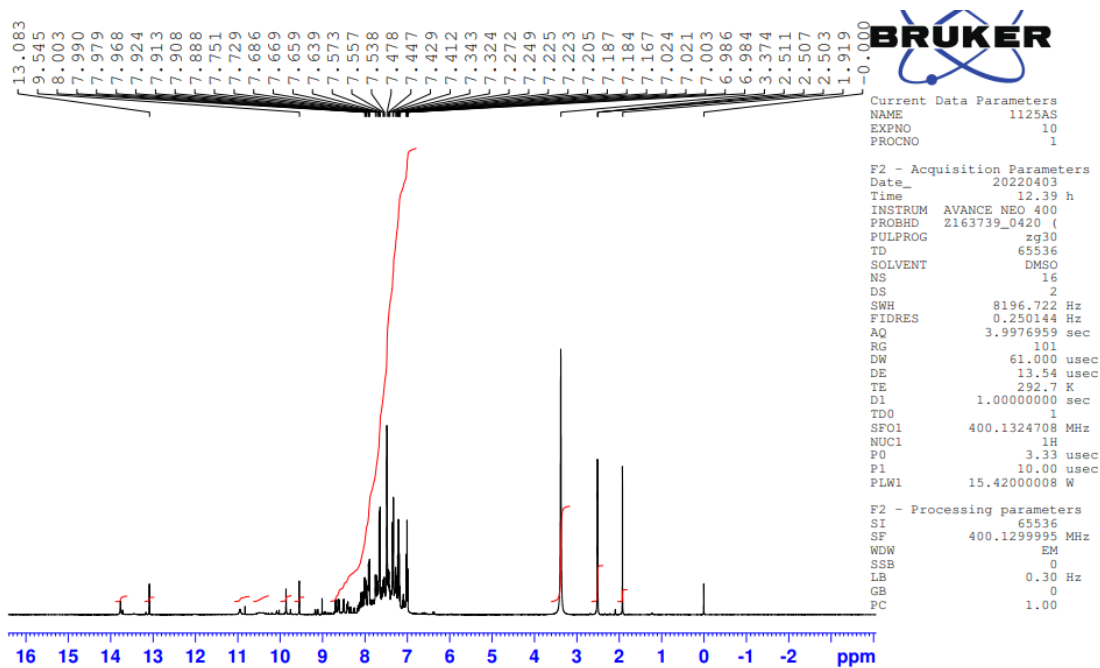


Figure 5. ^1H -NMR spectrum of α -amino acid ($\text{H}_2\text{L1}$)

^{13}C -NMR Spectrum of $\text{H}_2\text{L1}$

The ^{13}C -NMR spectrum of α -amino acid ($\text{H}_2\text{L1}$), Figure 6, shows the carboxylic group appearing as expected at 166.88 ppm. The aromatic carbon signals of phenyl ring of benzothiazol appeared at 153.29 ppm, while the aliphatic carbon signals at 21.52. Signals related to aromatic carbon were detected at range 111.07 to 135.50 ppm and N-C signals at 40.59 ppm [19–21].

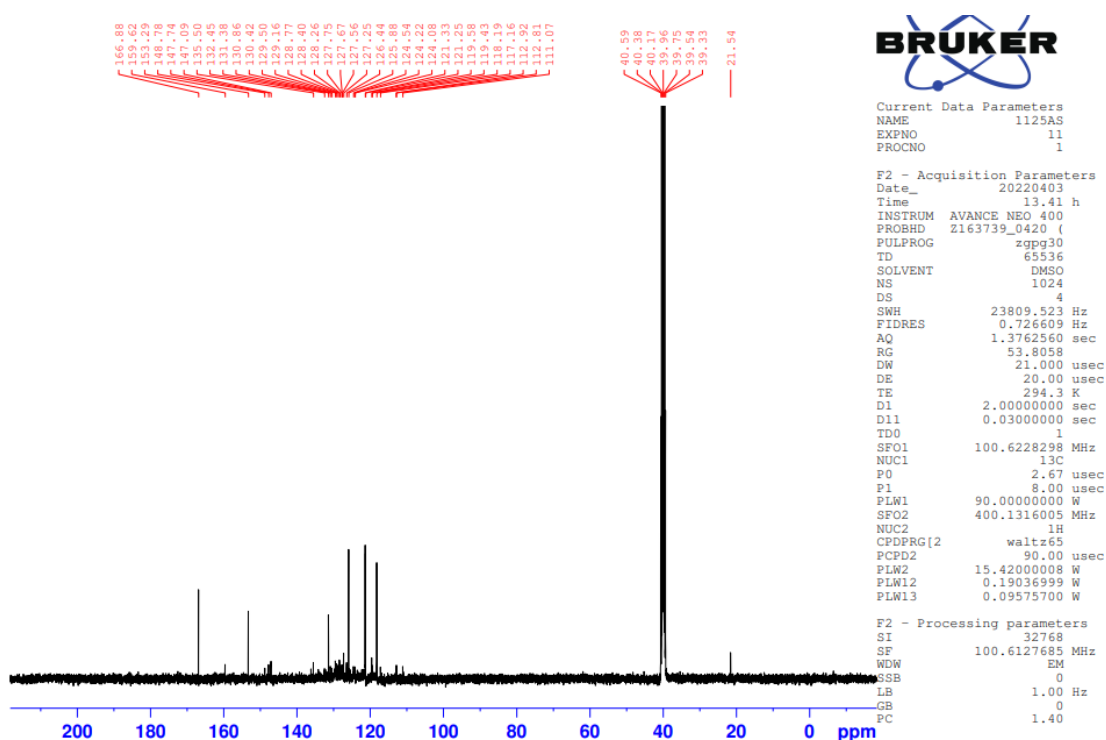


Figure 6. ^{13}C -NMR spectrum of α -amino acid ($\text{H}_2\text{L1}$)

Ultraviolet-Visible spectroscopy

Mono- and binuclear Co(II) complexes spectra showed in general three d-d electronic transition band of ${}^4T_{1g} \rightarrow {}^4T_{2g}$, ${}^4T_{1g} \rightarrow {}^4A_{2g}$ and ${}^4T_{1g} \rightarrow {}^4T_{2g}(P)$ that represented ν_1 , ν_2 and ν_3 respectively. and mono- and binuclear Cr(III) complexes spectra showed in general two d-d electronic transition ${}^4A_{2g} \rightarrow {}^4T_{2g}(F)$ and ${}^4A_{2g} \rightarrow {}^4T_{1g}(F)$ that represented ν_1 and ν_2 , respectively. ν_3 ${}^4A_{2g} \rightarrow {}^4T_{1g}(P)$ band could be shifted to higher frequency and interfered with ligand field transition. Charge transfer transitions accord around 400 nm which represents ligand to metal charge transfer due to the effect of ligands (1,10-phen, α -amino acid and bridging ligand (1,3-diaminopropan)) coordination with the metal ion. Mixed ligand complexes of Co(II) and Cr(III) included variable types of ligands, which consisted of $\pi \rightarrow \pi^*$ and $n \rightarrow \pi^*$ electronic transitions at the range 200 to 350 nm as shown in Figure 7. Table 2 presents the electronic transition spectral data of Co(II) and Cr(III).

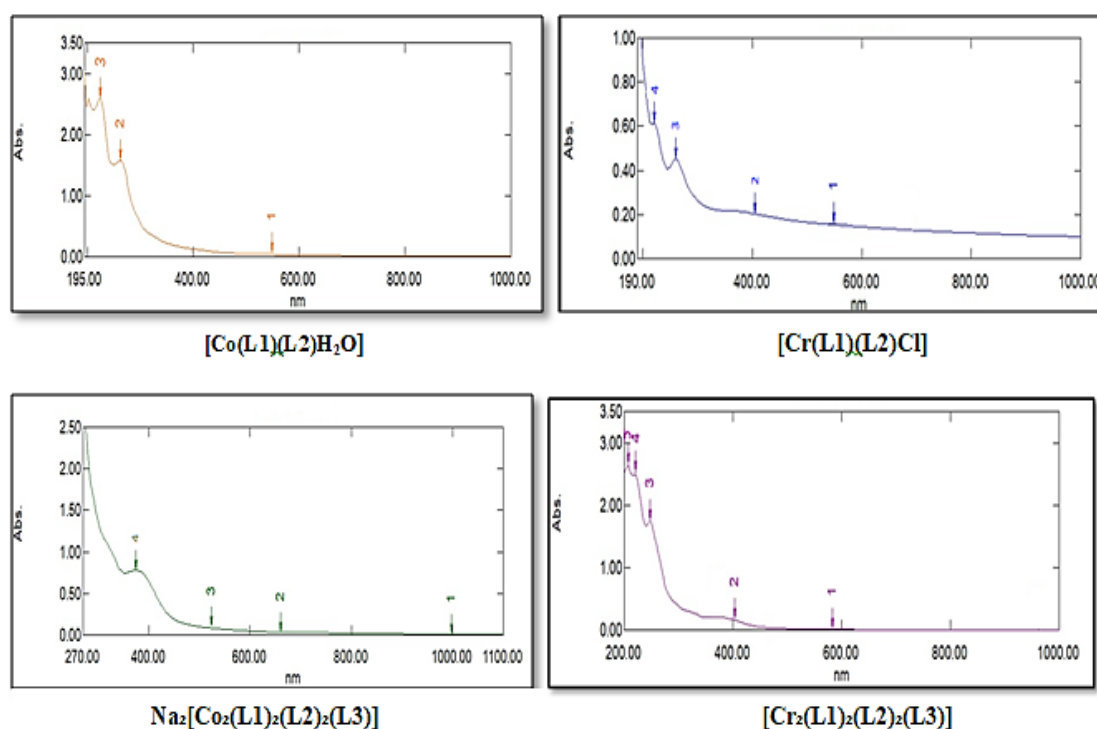


Figure 7. Ultraviolet-visible spectrum for Co(II) and Cr(III) complexes

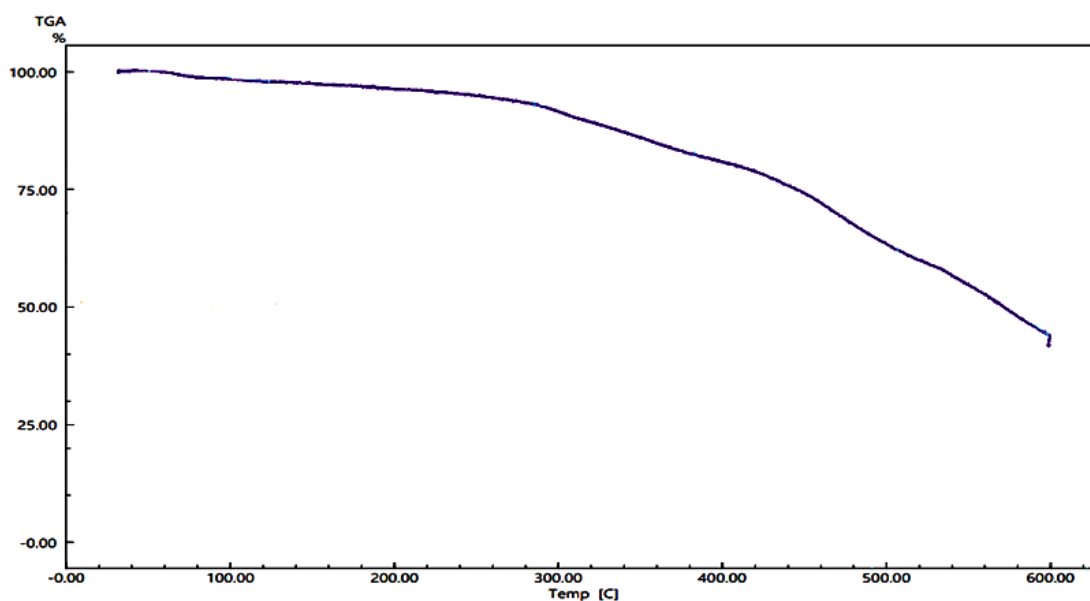
Table 2. Electronic transition spectral data of Co (II) and Cr (III) complexes of mixed ligand complexes

Complex	λ nm	ν cm^{-1}	Assignments	Suggested geometry
[Co(L1)(L2)H ₂ O]	966	10,351	${}^4T_{1g} \rightarrow {}^4T_{2g}(F)$	distorted octahedral
	669	14,947	${}^4T_{1g} \rightarrow {}^4A_{2g}(F)$	
	574	17,421	${}^4T_{1g} \rightarrow {}^4T_{2g}(P)$	
	262	38,167	$\pi \rightarrow \pi^*$	
	224	44,624	$n \rightarrow \pi^*$	
Na ₂ [Co ₂ (L1) ₂ (L2) ₂ (L3)]	998	10,020	${}^4T_{1g} \rightarrow {}^4T_{2g}$	distorted octahedral
	660	15,151	${}^4T_{1g} \rightarrow {}^4A_{2g}$	
	525	19,047	${}^4T_{1g} \rightarrow {}^4T_{2g}(P)$	
	374	26,737	C.T	
[Cr(L1)(L2)Cl]	570	17,543	${}^4A_{2g} \rightarrow {}^4T_{2g}(F)$	distorted octahedral
	401	24,937	${}^4A_{2g} \rightarrow {}^4T_{1g}(F)$	
	300	33,333	$n \rightarrow \pi^*$	
	237	42,194	$\pi \rightarrow \pi^*$	
[Cr ₂ (L1) ₂ (L2) ₂ (L3)]	592	16,891	${}^4A_{2g} \rightarrow {}^4T_{2g}(F)$	distorted octahedral
	402	24,875	${}^4A_{2g} \rightarrow {}^4T_{1g}(F)$	
	248	40,322	$n \rightarrow \pi^*$	
	225	44,444	$\pi \rightarrow \pi^*$	
	208	48,076	$\pi \rightarrow \pi^*$	

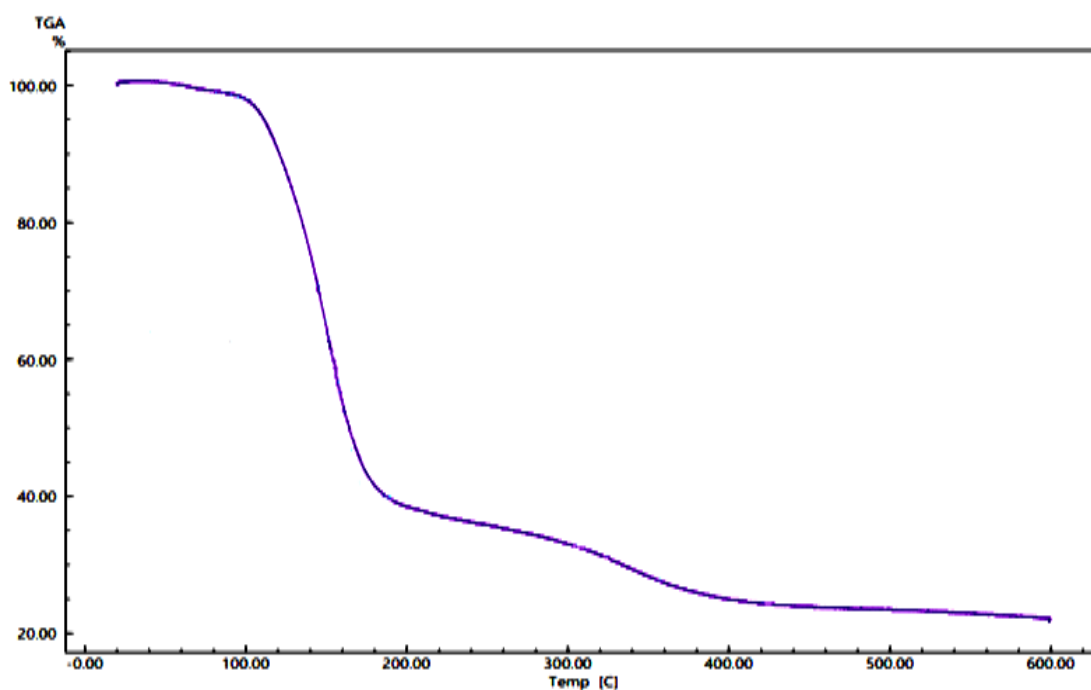
Thermal Analysis

The results obtained from the TGA of the mixed ligand complexes within the range of 20°C to 600°C under nitrogen atmosphere are described in Table 3 based on the thermograms in Figure 8.

At low temperatures, the initial weight loss is detected from the curve of the thermograph of Co(II) and Cr(III) mixed ligand complexes, where the lattice water molecules are released first, followed by coordinated water molecules, as all compounds disintegrate gradually. The following stages include the degradation of ligands and departure of organic compounds noticed by the braking energies of coordinated bonding. The final stage of the thermal analysis produced some metal oxides, so TGA results illustrated the conformation of the proposed structure of the prepared complexes.



[Co(L1)(L2)H₂O]



Na₂[Co₂(L1)₂(L2)₂(L3)]

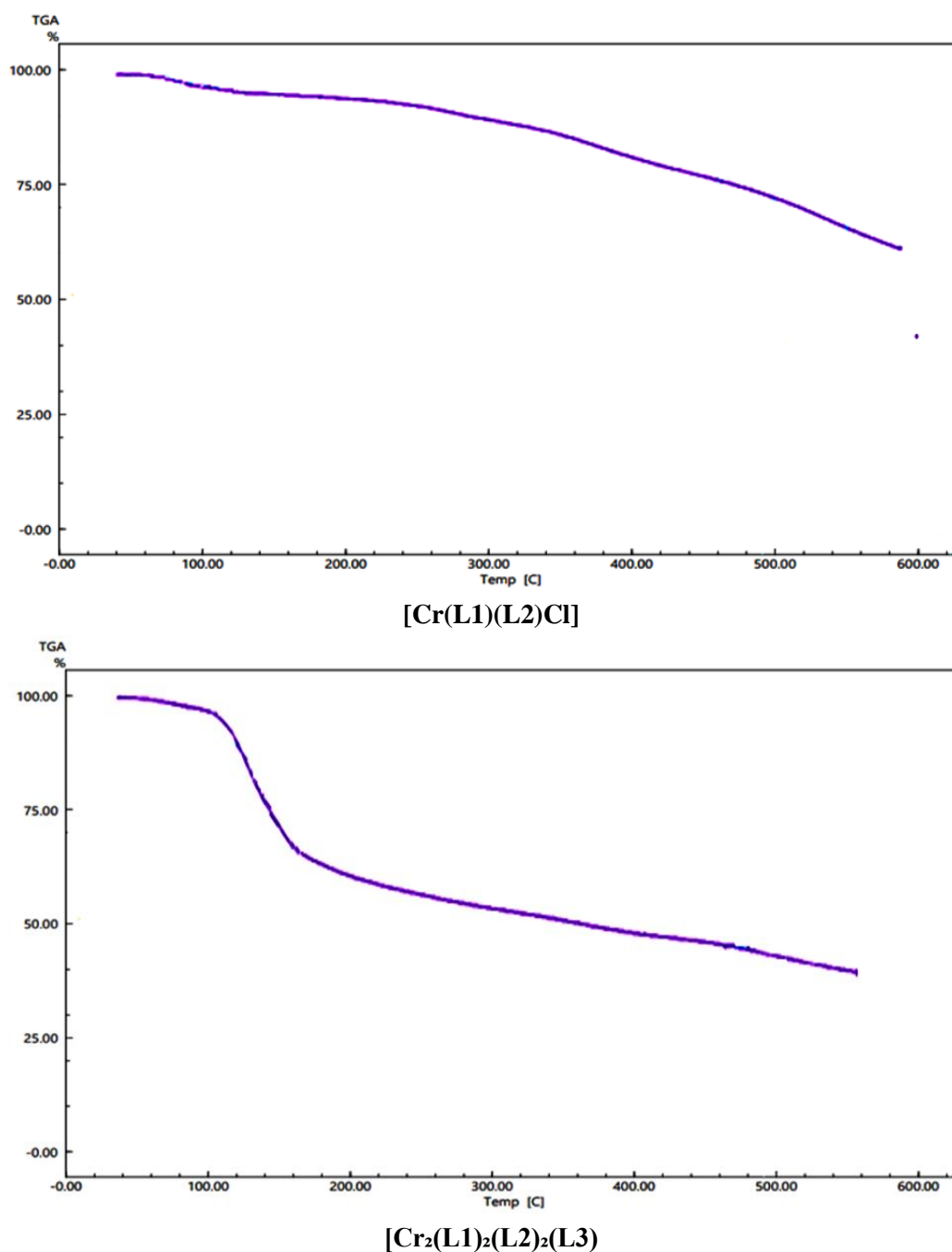


Figure 8. TGA for Co (II) and Cr (III) complexes

Table 3. Data of thermal decomposition of mixed ligand complexes

Complexes	M.Wt	Stage	TG range/°C	Assignments loss	Mass loss%	
					Cal.	Found
[Co(L1)(L2)H ₂ O]	623	1	20-140	-H ₂ O (coordinated)	2.8 %	2.5 %
		2	140-600	-CO ₂ benzen + naphthoxide	44 %	43 %
Na ₂ [Co ₂ (L1) ₂ (L2) ₂ (L3)]	1328	1	20-300	-2(L1)	52 %	67 %
		2	300-600	-(NH ₂) ₂ (CH ₂) ₂	5.5 %	6 %
		3	>600	-CO ₂ (L3) ₂	37 %	27 %
[Cr(L1)(L2)Cl]	634	1	20-600	-naphthoxide	22 %	20 %
[Cr ₂ (L1) ₂ (L2) ₂ (L3)]	1268	1	20-600	2(L1) + (L4)	60.7 %	62 %

Molar Conductivity Measurement

The molar conductance of Co(II) and Cr(III) complexes were measured by using dimethyl sulfoxide (DMSO) as solvent at 10^{-3} M. It was noticed that some of the complexes are electrolytic and others are not. This occurred due to the charged ligand and metals. L1 ligand are charged of -2 , while the bridging ligand when coordinating and cooperating with L2 ligand lost two protons ($L3^{2-}$) from both terminal side, therefore considered as H_2L3 in those complexes $Na_2 [Co_2 (L1)_2 (L2)_2 (L3)]$ and $[Cr_2 (L1)_2 (L2)_2 (L3)]$. Table 4 illustrates the electrolytic behavior of the complexes.

Table 4. Conductivity of the prepared complexes in dimethyl sulfoxide (DMSO).

Compound	Cond. Ms/cm	Electrolytic properties
$[Co(L1)(L2)H_2O]$	10	Non-electrolytic
$Na_2 [Co_2 (L1)_2 (L2)_2 (L3)]$	68	Electrolytic
$[Cr(L1)(L2)Cl]$	12	Non electrolytic
$[Cr_2 (L1)_2 (L2)_2 (L3)]$	6	Non electrolytic

Biological Activity

In vitro bacterial activities of the ligand and the complexes have been carried out against the pathogenic bacteria like Gram-negative bacteria *Pseudomonas* and Gram-positive *Escherichia coli* using nutrient agar medium by well diffusion method. The test solutions were prepared with DMSO as a solvent which having a concentration 10^{-3} mol/L and soaked in filter paper of 5 mm diameter and 1 mm thickness. These discs were placed on the already seeded plates and incubated at $37^\circ C$ for 24 hours. The diameters of the inhibition zone around each disc were measured after 24 hours. From the results presented in Figure 9, it can be noted that all the compounds have no effects of the biological activity of *Pseudomonas* except L2 have the most effect on *E. coli* compared with the other prepared complexes. Table 5 illustrates the results showing the inhibition of growth of bacteria due to the compounds.

Table 5. Antibacterial activity of the prepared compounds

No.	Compound	<i>Pseudomonas</i>	<i>Escherichia coli</i>
3	$[Co(L1)(L2)H_2O]$	--	+
4	$Na_2 [Co_2 (L1)_2 (L2)_2 (L3)]$	--	+
7	$[Cr(L1)(L2)Cl]$	--	+
8	$[Cr_2 (L1)_2 (L2)_2 (L3)]$	--	--
10	L2	--	++
11	L1	--	--
12	$CoCl_2 \cdot 6 H_2O$	--	--
13	$CrCl_2 \cdot 6 H_2O$	--	--
14	Tetracycline	+	+
15	DMSO	--	--

Note: -- = No inhibition + = 5–10 mm; ++ = 11–20 mm.

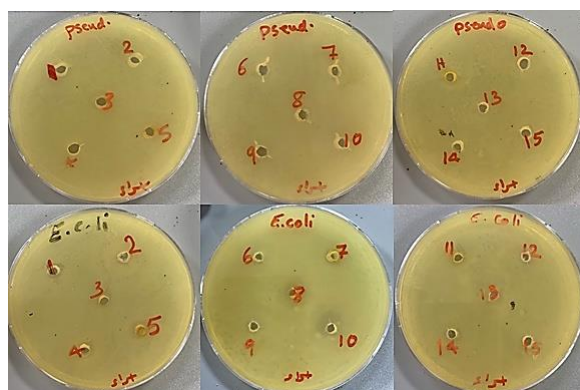


Figure 9. Zone effect of the compounds on Gram-negative *Pseudomonas* and Gram-positive *Escherichia coli*.

CONCLUSION

The L1 ligand is considered as a primary tridentate chelating ligand of N₂O-type coordination, hence it was charged due to the basic reaction medium. The L2 ligand is considered as bidentate secondary ligand and it is generally strong. The six coordination numbers of Co(II) complexes are satisfied by aqua molecule, while Cr(III) complexes are satisfied by chloride molecule in mononuclear complexes, respectively. The bridging ligand behavior when coordinated and cooperated with L2 in Na₂ [Co₂ (L1)₂ (L2)₂ (L3)] and [Cr₂ (L1)₂ (L2)₂ (L3)] lose two protons and are considered as H₂L₄. As regards antibacterial activity, *Pseudomonas* was very aggressive and resistant against the complexes with no promising results, while their effect on *E. coli* showed promising results.

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