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Synthesis and characterization of Mn(II),Co(II), Ni(II),Cu(II) ,Ca(II) complexes with the ligand derived from indomethacin

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Article Informations

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A B S T R A C T

The 2-(1-(4-chlorobenzoyl)-5-methoxy-2-methyl-1H-indol-3-yl)-N'-(4-methylbenzylidene)acetohydrazide ((IB) was synthesized through the condensation of hydrazide derived **from indomethacin** with 4-methyl benzaldehyde. Additionally, the complexes obtained from the ligand were characterized by infrared, UV-visible, 1H, 13C NMR spectroscopy, elemental analysis, atomic absorption, molar conductance, and magnetic susceptibility. Measurements have shown that the ligand behaves as a bidentated ligand and is coordinated to the central metal ion through the oxygen atom for the carbonyl group and the nitrogen atom for the azomethine group. The structure of the ligand and complexes was confirmed. With the exception of the copper complex, which has an octahedral geometry, all of the complexes have adopted a tetrahedral geometry.

Introduction

A nonsteroidal anti-inflammatory medicine (NSAID) is used most often in diseases that are inflammatory is indomethacin[1,2]. Hydrazones derived from NSAIDs exhibit unique biological characteristics, including anti-inflammatory, analgesic, and anticonvulsant effects [3]. Schiff bases are compounds with the functional group azomethine -C=N-, which is an imine. These compounds function as important chelating agents because they contain oxygen or nitrogen, and they also function as multidentate ligands, coordinating with different metal ions in a variety of oxidation states and geometrical shapes. Because of their wide variety of biological activities, these compounds are highly significant in the pharmaceutical and medicinal fields.It is widely acknowledged that arylhydrazone complexes of metal ions provide helpful models for ellucidating the processes leading to hydrazine derivatives' inhibition of enzyme activity, as well as that NSAIDS ligands and complexes are of great importance as a result of their many applications in various fields, starting with the medicinal and veterinary fields, in addition to industrial applications and organic synthesis[4-6]. In the biological field, many studies have proven that the biological activity of complexes prepared from NSAIDS ligands with the metal atom is higher than in the ligand as antibacterial, antifungal, antimicrobial, and anticancer[7-9]).This kind of ligand is distinguished by the development of several stereotypes and the presence of various donor atoms, such as nitrogen and oxygen,which allow it to coordinate with the center metal atom[10],11]. The aim of the work is the preparation of new complexes derived from indomethacin and the characterization of these complexes by different meaurements.

Materials and Methods

High-purity chemicals and solvents from Merch and Fluka were used to produce the compounds. The melting point temperature was determined using the Stuart-Smpio melting point apparatus.

The carbon, hydrogen, and nitrogen elements of the ligands and complexes created with an EA 3000 V.3.0 single Euro equipment were also analyzed, in addition to the elements of the prepared complexes being determined using Shimadzu atomic absorption. Complexes' molar conductance was measured in DMSO at room temperature for 10-3 M using the BC 3020 professional Bench top conductivity. The complexes' magnetic susceptibility was measured at room temperature using the Faradys method by the Johnson Mattey catalytic system division in England . The electronic spectra (UV-Vis) of the prepared ligand and complexes were recorded using a UV-visible Spectorphotometer (Shimadzu-1650PC), and the solvent (DMF) was used. Using Shimsdzu (FTIR-8400) equipment, infrared spectra measurements of the synthesized ligand and complexes were performed in the 400–4000 cm-1 range. (13C, 1H.NMR) spectra of the prepared ligand were recorded using a Bruker device (300 MHz) and using DMSO-d6 as a solvent.

Synthesis of the ligand

Synthesis of ethyl 2-(1-(4-chlorobenzoyl)-5-methoxy-2-methyl-1H-indol-3-yl)acetate

Indomethacin ester is prepared according to Fisher's esterification [12, 13] by dissolving (0.03 mol, 10.73 gm) of carboxylic acid (indomethacin) in 50 ml of absolute ethanol and adding 3 ml of concentrated sulfuric acid. The mixture rises for about 8 hours, and after cooling, it is neutralized.With a solution of sodium bicarbonate (NaHCO3) until the ester separated. the recrystallization of ester from pure ethanol and vacuum-dried , and its m.p 115-117 oC, Anal. Calc. C21H20 NO4Cl : C, 65.36 ; H , 5.18 ; N,3.63 ,Found; C,64.65; H, 5.03,N,3.28 %.

Synthesis of 2-(1-(4-chlorobenzoyl)-5-methoxy-2-methyl-1H-indol-3-yl)acetohydrazide

This hydrazide is prepared [14] by dissolving (0.02 mol, 7.71 g) of the ester prepared in the first step in 35 ml of hot absolute ethanol, then reacting it with aqueous hydrazine (85%), then refluxing the mixture for 24 hours, after which the precipitate is obtained, which is recrystallized with ethanol. m.p 137-139 oC, Anal. Calc. C19H18 N3O3Cl : C, 61.37 ; H , 4.84 ; N , 11.30,Found; C,61.19; H, 4.33; N, 11.14 %.

Synthesis of 2-(1-(4-chlorobenzoyl)-5-methoxy-2-methyl-1H-indol-3-yl)-N'-(4-methylbenzylidene)acetohydrazide (IB)

Dissolve (0.01mol, 3.71 gm) of the prepared hydrazide in 25ml of hot absolute ethanol, then add (0.01mol, 1.2gm) of 4- methyl benzaldehyde with a few drops of glacial acetic acid. Then the mixture refluxing for five hours, and the precipitate is obtained, which is recrystallized with ethanol [15], m.p 168-169oC . Anal. Calc. C27H24N3O3Cl: C,68.42 ; H , 5.06; N, 8.87; Found : C, 68.29 ; H, 4.83 ; N, 8.57 %.

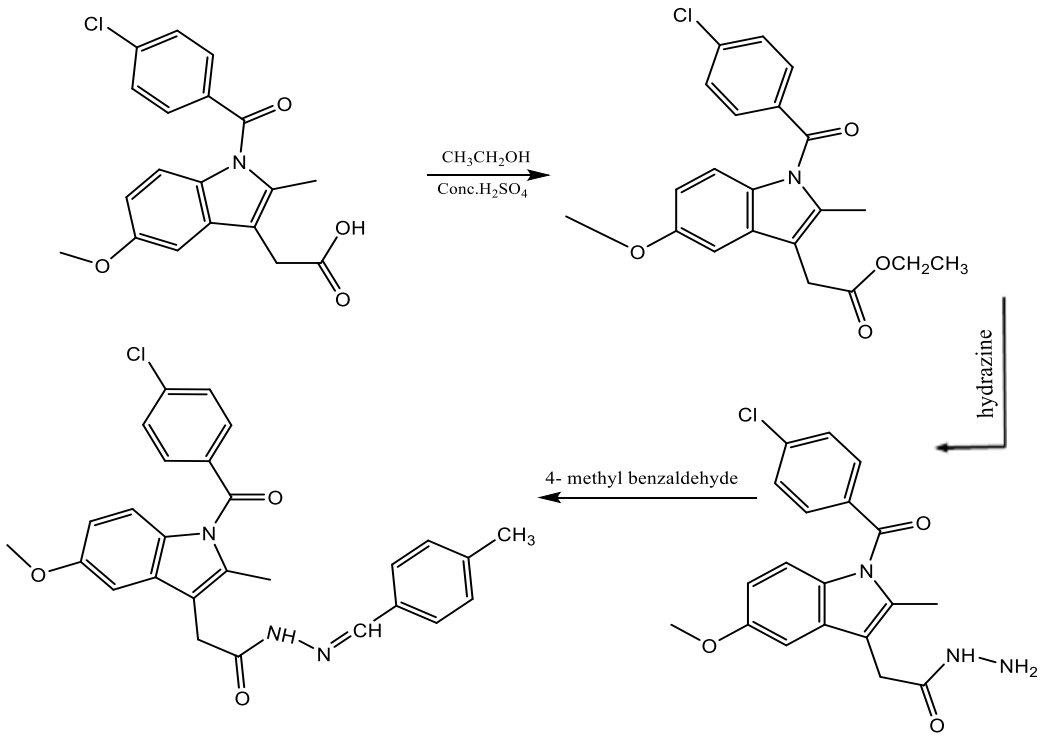

Figure 1. The structure of the ligand(IB)

Synthesis of the complexes

The complexes were prepared in molar ratios of (1:1) (metal:ligand) by adding (0.004 mol) of the metal (II) chloride MnCl2.4H2O 0.79 gm, CoCl2.6H2O 0.95 gm, NiCl2.6H2O 0.95 gm, CuCl2.2H2O 0.68 gm, and CaCl2 0.44 gm dissolved in 10 ml of absolute ethanol to the ligand solution (0.004 mol, 1.89gm) dissolved in 15 ml of absolute ethanol, and the mixture refluxed with continuous stirring for 3 hours. The solution was then cooled at room temperature as a precipitate, which was separated by filtration, washed with small amounts of cold ethanol, and dried [16].

Results and discussion

Most of the produced ligand and complexes are colorful and insoluble in water, but they dissolve at 10-3 M in organic compounds like dimethyl sulfoxide. The results of the measurements showed that all of the prepared complexes are non-electrolytic, as it is possible for the chloride ion to escape inside the coordination sphere [17]. The elemental values and physical characteristics of the ligands and complexes shown in Table 1.

Table 1. The ligands and complexes' elemental values and physical properties

No.	Formula	Colour	m.p ºC	Ω $Ohm^{-1}.cm^2.mol^{-1}$	calculate (Found)%			
					%C	**%H**	**%N**	**%M**
1	$[Mn(IB)Cl_2]$	White Yellowish	269	16	54.05 (53.85)	4.00 (3.76)	7.00 (6.89)	9.16 (9.02)
2	$[Co(IB)Cl_2]$	Brown	289	13	53.69 (53.42)	3.97 (3.65)	6.96 (6.54)	9.76 (9.58)

3	$[Ni(IB)Cl_2]$	Green	250	17	53.72 (53.51)	3.97 (3.73)	6.96 (6.47)	9.71 (9.46)
4	$[Cu(IB)\ Cl_2(H_2O)_2]$	Gray	278	20	50.31 (50.11)	4.03 (3.79)	6.52 (6.32)	9.86 (9.67)
5	$[Ca(IB)Cl_2]$	White	291	14	55.43 (55.21)	4.10 (4.03)	7.18 (6.89)	6.85 (6.65)

IR spectra

The IR spectra of the liqand show bands at 1620, 1533, 1002 cm^{-1}, which belong to the groups $\boldsymbol{\nu}$(C=O-NH, $\boldsymbol{\nu}$(C=N), and $\boldsymbol{\nu}$(N−N), respectively and these bands are shifted to lower values, which indicates coordination through the carbonyl oxygen atom and azomethine nitrogen atom . The ligand spectrum showed the stretching frequency of the $\boldsymbol{\nu}$(N−H) and $\boldsymbol{\nu}$(-C=O-Ar) groups at 3265 cm^{-1} and 1645 cm^{-1}, respectively[18],[19]. These bands appear in the spectra of the metal complexes in the same region or a little shifted, indicating that they are not participate in the coordinated to the central metal ion [20]. The appearance of the band at 3356 cm^{-1} in the spectra of the copper complex shows that the H_2O group is in coordination with the copper ion [21].The IR spectrum of the complexes showed new bands at the range 435-539 cm^{-1} and 547-596cm^{-1} which represented to the $\boldsymbol{\nu}$(M−N) and $\boldsymbol{\nu}$(M−O),respectively [22]. The FTIR spectra of the ligands and complex $[Ni(IB)Cl_2]$ shown in Figure 2,3 and Table 2.show the selected bands of the ligand and thir complexes.

Table 2. The selected infrared bands cm^{-1} of the ligand and thir complexes

Compound no.	υ (N-H)	υ (-C=O-Ar)	υ (C=O-NH)	υ (C=N)	υ (N-N)	υ (M-N)	υ (M-O)
IB	3265	1645	1620	1533	1002	---	---
1	3259	1644	1571	1555	1019	439	547
2	3260	1641	1572	1549	1097	452	549
3	3260	1646	1607	1570	997	435	596
4	3260	1650	1596	1547	1073	544	494
5	3259	1641	1605	1540	1020	551	509

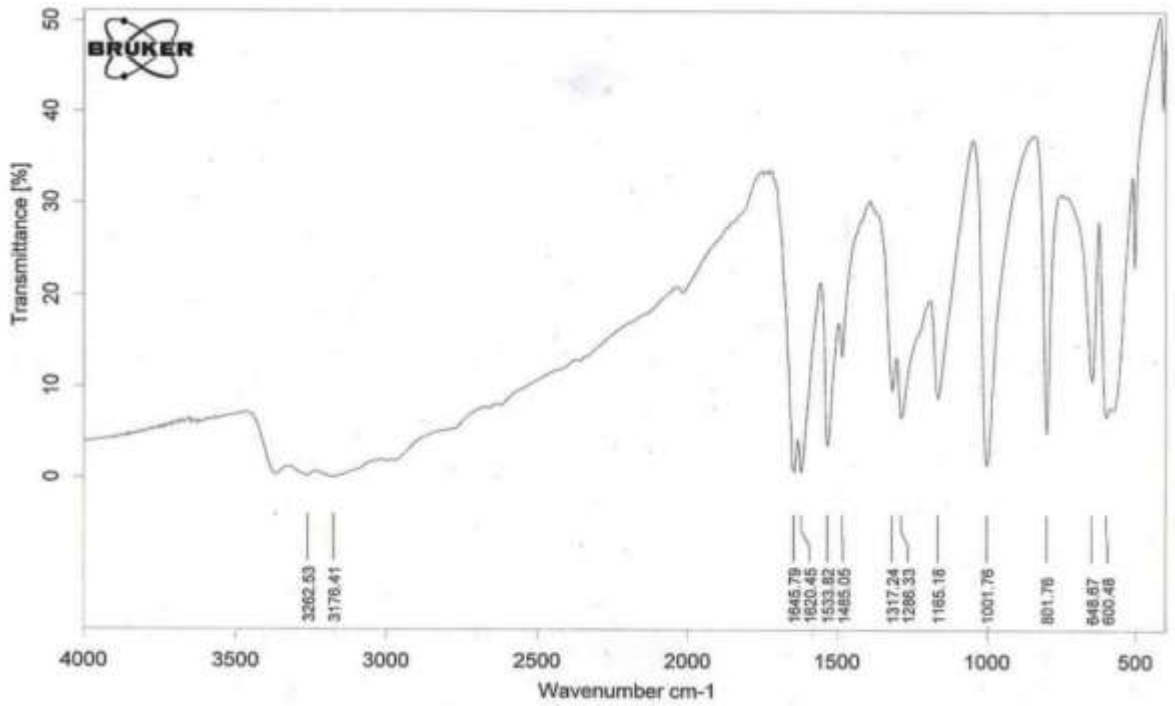

Figure 2. The ligand Infrared spectra (IB)

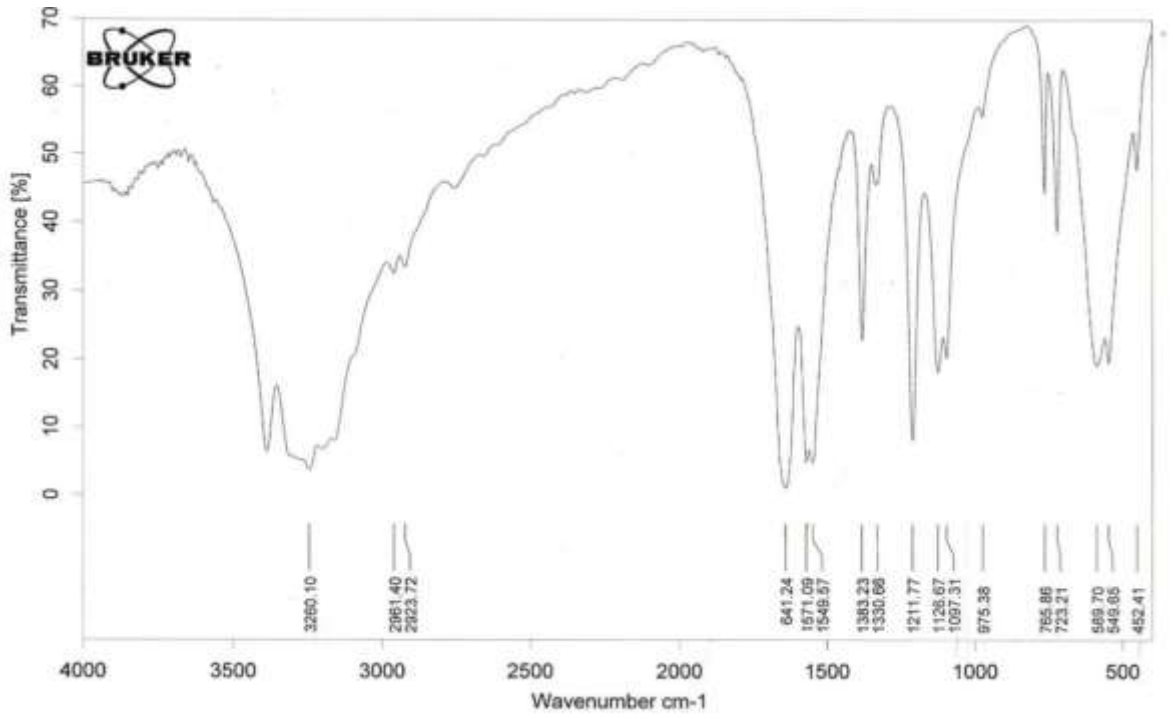

Figure3. The complex infrared spectra[$Co(IB)Cl_2$]

Magnetic Moment measurements an Electronic Spectral

The free hydrazone ligand shows tow bands at 32679 and 29411 cm-1 assinged to the $\pi \rightarrow \pi^*$ and n π^* transitions, respectively [23] Three peaks were visible in the Mn^{+2} complex's electronic spectra at 14245 cm^{-1}, 11655 cm^{-1}, and 10162 cm^{-1}, which were assigned to $^6A_1 \rightarrow {}^4E_1$, $^6A_1 \rightarrow {}^4T_2$, and $^6A_1 \rightarrow {}^4T_1$, respectively and the manganese complex's magnetic moment value is 4.96 B.M., suggesting tetrahedral geometry around the manganese ion [24],[25] .The Co^{+2} complex spectra show band at 11494cm^{-1} due to the $^4A_2(F) \rightarrow {}^4T_1(P)$ υ3 transition, and the cobalt complex's magnetic moment value is 4.41 B.M.,These results are consistent with the tetrahedral geometry of the cobalt complex [26]. The Ni(II) complex spectra display a band at 15015 cm^{-1} attributed to the transition $^3T_1(F) \rightarrow {}^3T_1(P)$. In addition, the complex's magnetic moment is 4.02 B.M.The observed results tetrahedral geometry of the nickel complex [27]. The UV spectra of the Cu(II) complex showed a band at 18450 cm^{-1} due to $^2Eg \rightarrow {}^2T_2g$,and the magnetic moment value for the Cu(II) complex is 2.13 B.M., which suggests an octahedral structure [28]. The complexe of Ca(II) is diamagnetic, and UV spectra showed band at 33557 cm^{-1} due to the charge transition, and these complex spectra showed no signs of d-d transition, which suggests an tetrahedral structure [29]. Figure 4 and 5 indicates the electron spectrum of the ligand (IB) and complex [$Ni(IB)Cl_2$], while Table 3 indicates the electronic transition and magnetic moment for metal complexes.

Table 3. Magnetic moment and electronic transition for metal complexes and Ligand

Compounds	λmax (nm)	Band (cm^{-1})	Assignments	Magnetic moment	Structures
ligand (IB)	306	32679	$\pi \rightarrow \pi^*$	--	--
	340	29411	$n \rightarrow \pi^*$		
[$Mn(IB)Cl_2$]	302	10162	$^6A_1 \rightarrow {}^4E_1$	4.96	**Tetrahedral**
		11655	$^6A_1 \rightarrow {}^4T_2$		
		14245	$^6A_1 \rightarrow {}^4T_1$		
		33112	C.T		

$[Co(IB)Cl_2]$	870 290	11494 34482	$^4A_2(F) \rightarrow {}^4T_1(P)$ **C.T**	4.41	**Tetrahedral**
$[Ni(IB)Cl_2]$	666 292	15015 34240	$^3T_1(F) \rightarrow {}^3T_1(P)$ C.T	4.02	**Tetrahedral**
$[Cu(IB)Cl_2(H_2O)_2]$	654 329	15290 30395	$^2Eg \rightarrow {}^2T_2g$ C.T	2.13	**Octahedral**
$[Ca(IB)Cl_2]$	298	33557	C.T	--	**Tetrahedral**

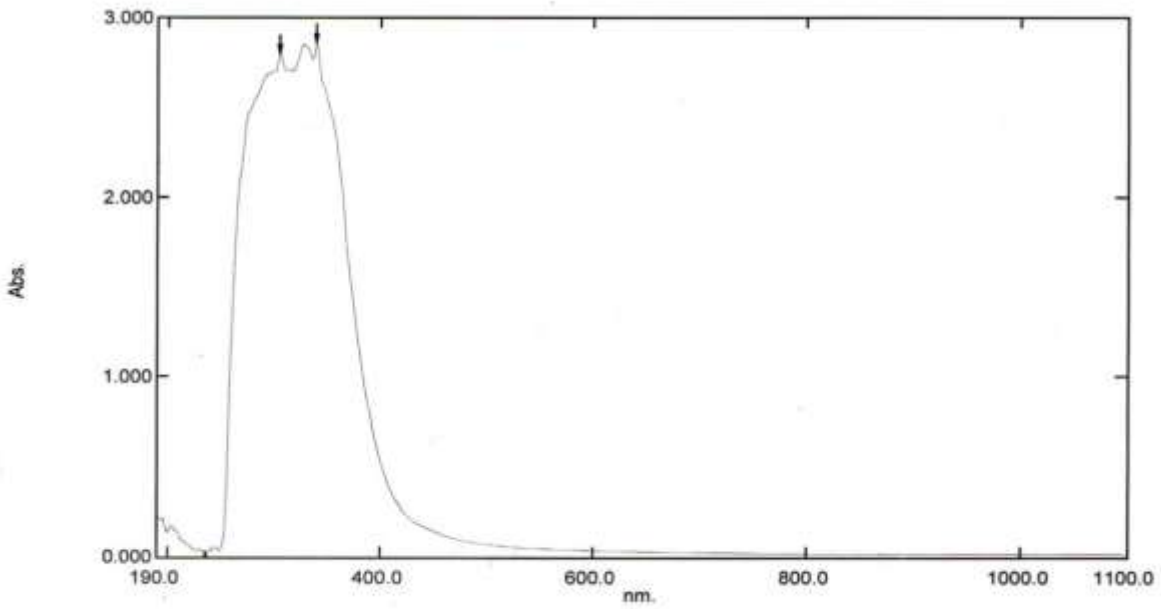

Figure 4 . Electron spectrum of the ligand(IB)

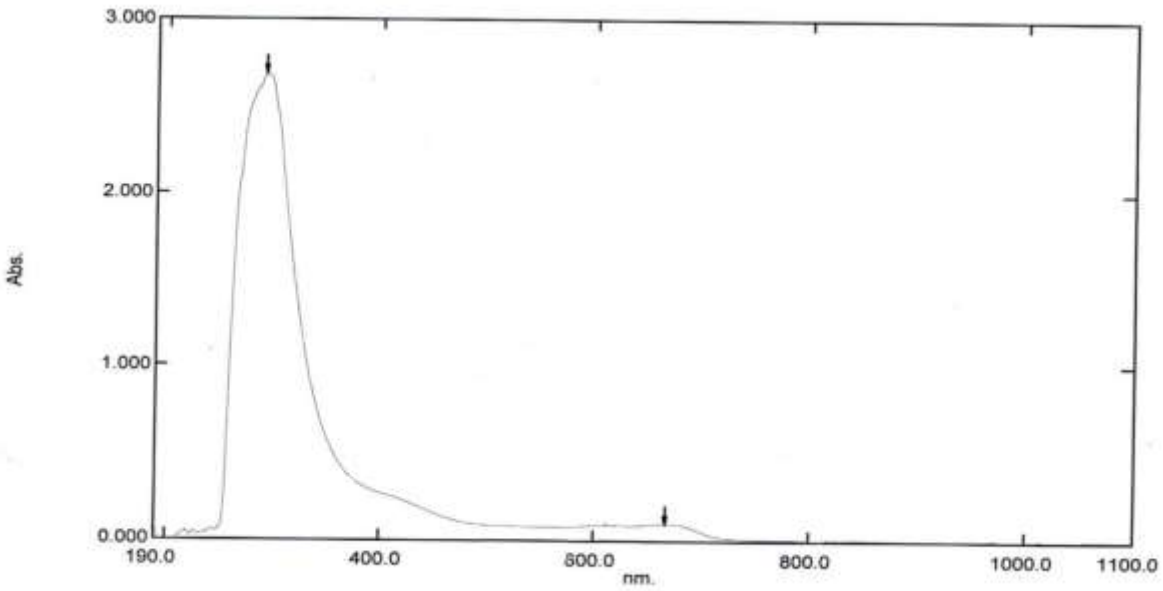

Figure 5. Electron spectrum of the complex $[Ni(IB)Cl_2]$

Nuclear Magnetic Resonance Spectra1H,13C.NMR

The ^{1}H.NMR spectra of the liqand(IB) shows signal at δ2.7 -4.2 ppm due to the aliphatic protons , and the aromatic protons showed signals at δ6.4-7.8ppm [30] .The spectra of ligand exhibit asignals at δ11.8 ppm and δ 8.3 ppm Which belong to the protons of NH and CH=N groups ,respectively [31] .
The ^{13}C.NMR spectra of the liqand exhibit two signals at 161 ppm and 173 ppm , which belongs to the carbonyl group and amide carbonyl , respectively [32][,33] . In addition to the signals at range 105-156ppm and 13-55ppm due to the carbons of aromatic rings and aliphatic, respectively, except signal at 148 ppm due to CH=N group [34]. The ^{13}C,^{1}HNMR Spectra of the ligand show in the figures 6 and 7.

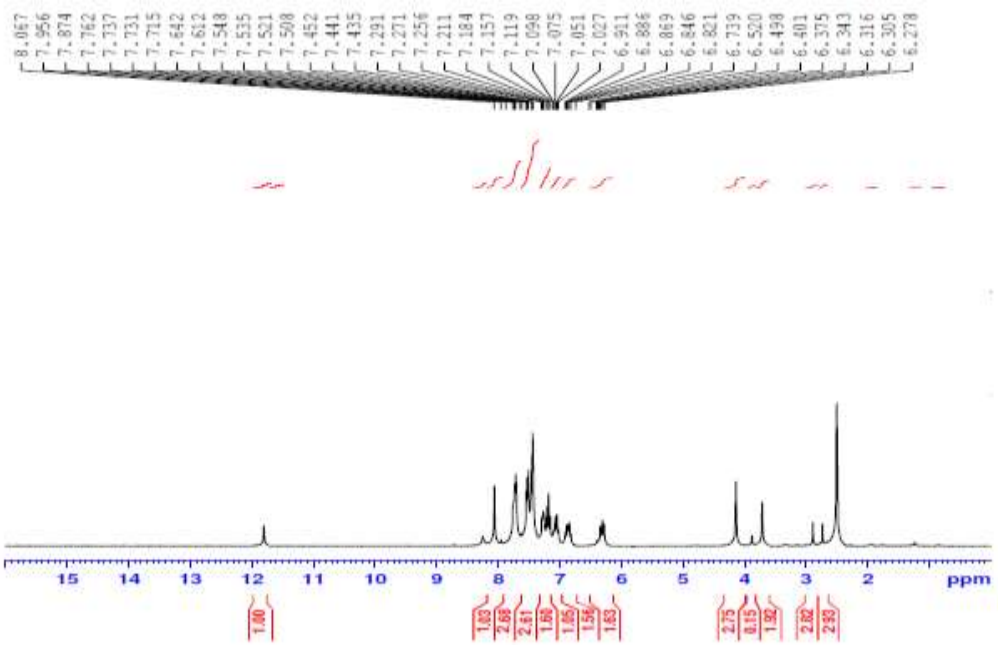

Figure 6. The ^{1}HNMR Spectra of the ligand(IB)

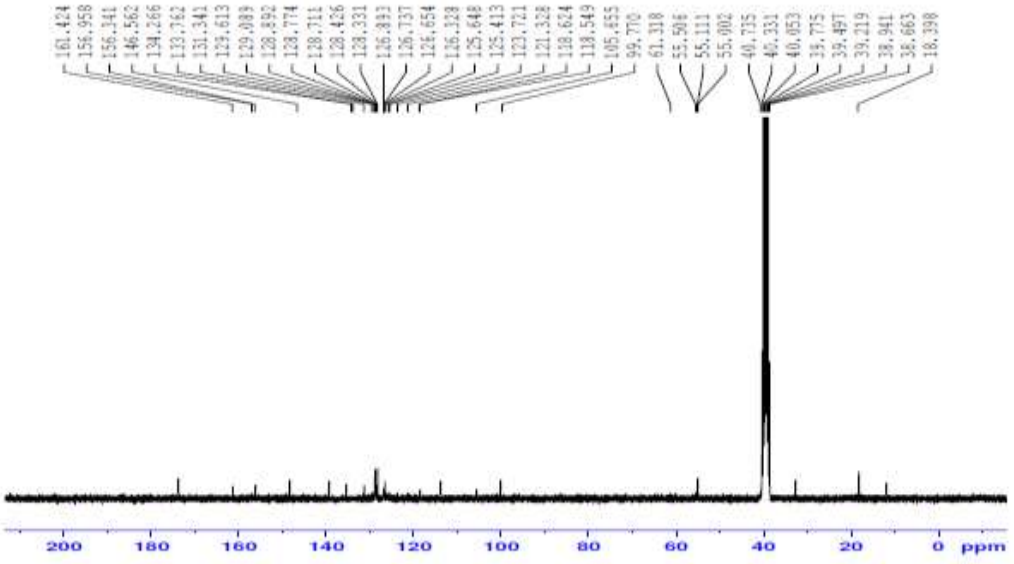

Figure 7. The ^{13}CNMR Spectra of the ligand(IB)

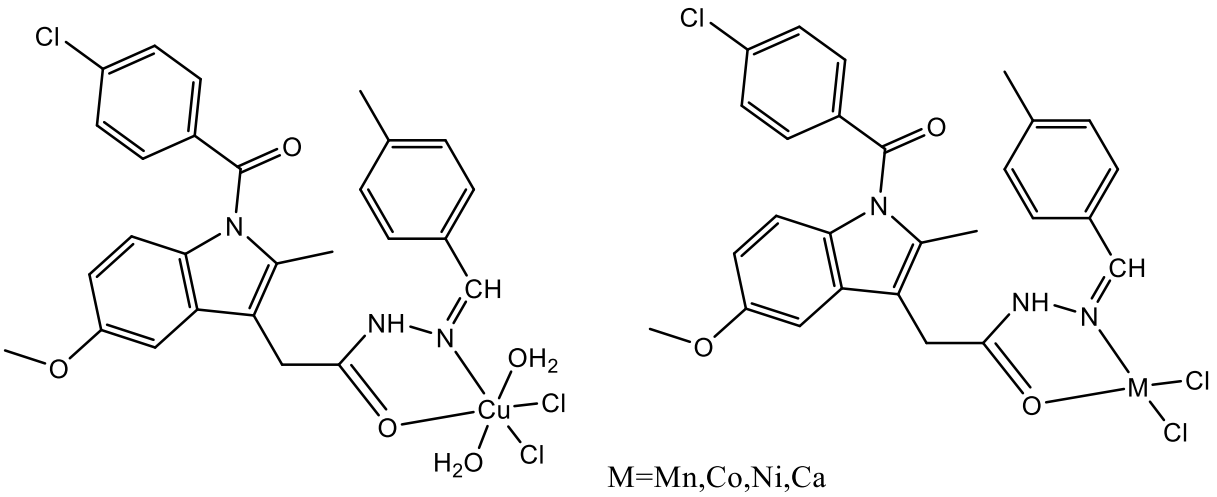

Figure 8. The structure of the prepared complexes

Conclustion

This research prepared hydrazone ligand derivative from indomethacin. It was reacted with metal salts Mn^{+2},Co^{+2},Ni^{+2},Cu^{+2},Ca^{+2}, and bsed on the measurement results explain that the ligand is bidentate and coordinated with the metal salts from the azomethine nitrogen and carbonyl oxygen atoms. These are done in addition to two chlor ions and the geometry form of the complexes which were tetrahedral expect the copper complex was octahedral.

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Competing Interests

Non.

Reference

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