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SYNTHESIS AND CHARACTERIZATION OF SOME COMPLEXES WITH IBUPROFENYL HYDRAZONE

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Abstract

Fourteen new metal complexes have been synthesized from ibuprofenyl hydrazide and vanillin or acetophenone ligands (L1 and L2) and used in preparing the corresponding Cobalt(II) , Nickel(II) , Copper(II) , Zinc(II) , Cadmium(II) complexes. The resulted complexes have been characterized by different physiochemical methods including elemental analyses, IR electronic spectra , magnetic moment measurements , molar conductance, and X-ray powder diffraction measurements, the ligands have been investigated by NMR spectra . Furthermore, the complexes have been found to have the formulas [M(L)2Cl2] where, M= Co(II) , Ni(II) , Cu(II) and Zn(II). whereas, the formula [M(L)2(H2O)2](NO3)2 , M= Co(II) , Ni(II) , [Cd(L)2]Cl2 ; L = L1 or L2 . Infrared spectral data suggest that the two ligands behave as a bidentate ligand with O,N, donor atoms towards the metal ions . Based on the above physicochemical measurements, the complexes have octahedral and tetrahedral geometries.

Keywords: X ray powder diffraction, metal complexes and ibuprofenyl complexes.

Музыка

Ibuprofen is 2-(4- isobutyl phenyl) propionic acid, is a member of non –steroidal anti-inflammatory drug (NSAIDs), known to relief a widely used non steroidal anti-inflammatory drug in treating pain and inflammatory drug (Fiori-Duarte,2019) but the long term use of this effects and nephrotoxicity.This led to the introduction of new compounds of Ibuprofen with an improved profile our aim to develop new safer drugs and improving the pharmacokinetic and pharmacotoxicological profile through complexation (Paulo-Santos et.al 2020). It has been reported that complexes of metallic salts are more potent and less toxic in many cases as compared to the parent drugs. known to relief symptoms of arthritis, primary dysmenorrhea, fever and also possess mild antiplatelet effect (Ariana,et.al,2022) is useful in sepsis-induced acute pneumonia, in retarding metastases of mammary carcinoma and in preventing oxidative lesions of lungs caused by phosgene.

High doses of ibuprofen slow down the evolution of lung disease. It also protects prostaglandin H synthase of human endothelial cells from hydrogen peroxide (Sondhi,et.al,2006). Hydrazones, a member of the Schiff base family with triatomic >C=N-N< Linkage , takes the fore front position in the development of coordination chemistry. Reports on the synthesis, characterization and structural studies on hydrazone ligands derived from (vanillin and acetophenone) show the importance of hydrazone complexes in various fields including analytical and biological field (Sheikhshoaie & Ebrahimipouret.al,2014). Hydrazone derivatives possessing anti-inflammatory, analgesic, antipyretic and antibacterial activities are also reported in the literature (Kafarska,et.al,2009,Bhandari,et.al.,2008). These complexes which plays an important role in reducing the toxicity of the parent drug and acts as a pro-drug (Abbas,et.,al. 2015, Kafarsk,et.,al. 2009).

2-Experimental:

1. Materials and Measurements :

All chemicals and solvent used for the syntheses were of analytical grade, the metal salts were commercially available pure samples and all chemicals used throughout this investigation from Merch ,B.D.H., Aldrich or Fluka and used without further purification.

2. Analytical and physical measurements

Melting point and decomposition temperature were determined using Stuart™ melting point apparatus SMP3-England. IR spectra measurements were recorded using FTIR-spectrometer (shimadzu), as ATR-unit in the range (400-4000 cm^{-1}). UV-Visible spectral measurements were recorded using Uv-vis 1900 spectrophotometer (shimadzu) for 10^{-3} M complexes in DMF solvent at room temperature, using 1cm quarts cell in range (200-900)nm . Elemental analysis were carried out on a Elementar Analasen system GmbH CHNS Germany.The NMR was recorded on Agilant Varian (USA), 500 MHz using deuterated DMSO-d^6 as a solvent . Molar conductance of complexes were measured at room temperature for 10^{-3} M in DMF using (WTW-Cond 3210, Germany).

Magnetic susceptibility of the complexes was carried out by (Sherwood Scientific MK 1 Magnetic Susceptibility Balance) at room temperature. Metal contents were estimated spectrophotometrically using atomic absorption spectrometer NOVAA 350 Scientific Equipments. X-ray powder diffraction data for complexes were measured by using XRD - Philips X'PERT powder diffractometer, Holland (University of Kashan-Iran).

Preparation of the Ligands and the Complexes:

1. Synthesis of 2(4-isobutyl phenyl) propionic acid ethyl ester (Ibuprofenyl ethyl ester) (1)

The Ibuprofen was esterified (Bhandari,et.al.,2008), by dissolving (6.18 g, 0.03 mole) of it in 20 ml of ethanol and then to that 2.0ml of sulfuric acid was added . The mixture was refluxed for 6-8 hrs. After completion of reaction solvent was removed by add 100m of cold water , followed by extracted with sodium bicarbonate and separated in the organic layer. The yield (80%) , b.p 250-252 ^{o}C, empirical formula $C_{15}H_{22}O_2$.

	%C	%H	%N
Calc.	76.92	9.40	--
Found	77.01	9.48	--

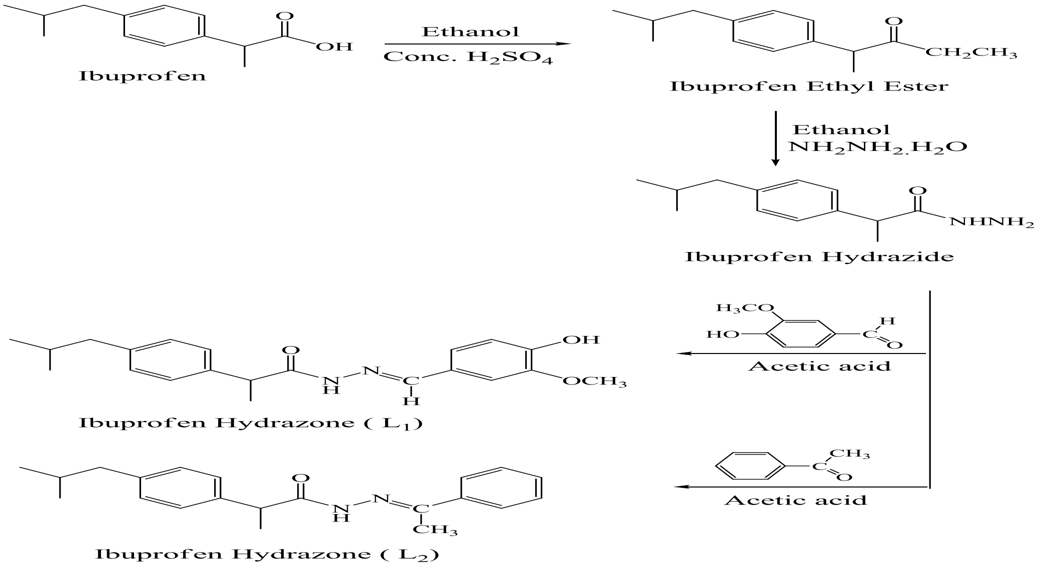
2. Synthesis of 2-(4-isobutylphenyl) propionoic acid hydazide)(2)

The hydrazide was prepared (Abbas,et.,al. 2015), by refluxing (4.68g , 0.02mol) of 2-(4-isobutyl phenyl) ethyl propionate in 15ml ethanol with an excess of hydrazine hydrate NH_2. $NH_2.H_2O$(85%) for 24h , the reaction mixture was then left to stand overnight .The compound precipitated on standing over night, filtered and washed with cold distilled water. The pure solid white hydrazide was obtained by recrystallization from ethanol, yield (81%), mp 76-78C empirical formula $C_{13}H_{20}N_2O$.

	%C	%H	%N
Calc.	70.90	9.09	12.72
Found	70.00	9.12	12.65

3. Synthesis of hydrazone ligands (L_1 & L_2)

The ligands synthesized according to the method described in the literature (Hrinath,et.,al. 2011)by reacting equimolar amount of Ibuprofenyl hydrazide and vanillin or acetophenone . A hot ethanolic solution of the ligand made by dissolving (2.2g, 0.01 mole) of Ibuprofenyl hydazide in 15 ml of ethanol has been slowly mixed with a hot ethanolic solution containing (1.52g , 0.01 mole) of vanillin or acetophenone (1.20g, 0.01 mole). The resulting mixture has been refluxed for about 6 hrs. The mixture has been left to stand for nearly two hours. The precipitated compound was filtered recrystallized from ethanol, washed with ether and dried under vacuum. The structures of the ligands are shown in Scheme 1



Scheme 1: The structures of the ligands

Table 1 : physical properties and analytical data of the ligands

ligands	Empirical formula , M.wt	Colour	m.p (C)	Yield %	Elemental analysis calcu. (found)		
					% C	% H	% N
L_1	$C_{21}H_{26}N_2O_3$ 354	white	103-104	71%	71.16 (71.21)	7.39 (7.41)	7.90 (8.00)
L_2	$C_{21}H_{26}N_2O$ 322	white	108-110	79%	78.20 (78.85)	8.07 (8.13)	8.69 (8.72)

4. Synthesis of the complexes

According to the following procedure, the complexes were obtained by adding an ethanolic solution of (Baligarand Revankar,2006) metal(II) chloride or nitrate (0.005mol) with the ligand L_1 (7.08 gm. , 0.01mol) or L_2 (6.44gm , 0.01mol) in the mole ratio 2:1 (L:M) after Mixing, an ethanolic solution has been refluxed for 5hrs . The volume of the solution was reduced to its half and the precipitated complexes washed with ethanol, followed by diethyl ether and dried in an oven at (70-80) C°.

Table 2: Weight of metal salts used to prepare complexes.

Metal salt	Wt(g) metal salt	metal salt	Wt(g) metal salt
$CoCl_2.6H_2O$	1.19	$CuCl_2.2H_2O$	0.85
$Co(NO_3)_2.6H_2O$	1.45	$Zn Cl_2$	0.68
$NiCl_2.6H_2O$	1.18	$Cd Cl_2$	0.91
$Ni(NO_3)_2.6H_2O$	1.45		

Table 3 : Characterization , analytical , molar conductance and magnetic susceptibility data of the complexes .

NO.	Formula	Mol. Weight	Color	Yield	m.p (C)°	Meff B.M	Λ_M DMF Cm^2. Ohm^1. mol^-	Calculate(Found)%			
								%C	%H	%N	%M
1	$[Co(L_1)_2Cl_2]$	838	Orange	65	252	4.98	18	60.14 (60.02)	6.20 (6.09)	6.68 (6.60)	7.04 (6.94)
2	$[Co(L_1)_2(H_2O)_2](NO_3)_2$	927	Orange	70	282	4.75	103	54.36 (54.21)	6.04 (5.95)	9.06 (8.89)	6.36 (6.12)
3	$[Ni(L_1)_2Cl_2]$	737.7	Orange	73	295	2.89	21	60.16 (60.07)	6.20 (6.09)	6.68 (6.43)	7.00 (7.02)
4	$[Ni(L_1)_2(H_2O)_2](NO_3)_2$	926.7	Brown	77	279	3.06	100	54.38 (54.17)	6.04 (5.91)	9.06 (8.79)	6.33 (6.17s)
5	$[Cu(L_1)_2Cl_2]$	842	Green	75	232	2.07	25	59.85 (60.00)	6.17 (6.07)	6.65 (6.50)	7.48 (7.37)
6	$[Zn(L_1)_2Cl_2]$	844	Yellow	77	>300	Dia	21	59.71 (60.00)	6.63 (6.47)	6.63 (6.40)	7.70 (7.43)
7	$[Cd(L_1)]Cl_2$	891	Yellow	69	263	Dia	97	56.56 (56.39)	5.83 (5.96)	6.28 (6.00)	12.57 (12.35)
8	$[Co(L_2)_2Cl_2]$	774	Green	70	288	4.98	15	67.74 (67.58)	6.71 (6.55)	7.52 (7.29)	7.93 (8.07)
9	$[Co(L_2)_2(H_2O)_2](NO_3)_2$	863	Brown	75	253	4.99	96	58.40 (58.66)	6.48 (6.41)	9.73 (9.95)	6.83 (7.00)

10	$[Ni(L_2)_2Cl_2]$	773.7	Green	68	264	3.28	28	67.76 (68.02)	6.99 (7.11)	7.52 (7.33)	7.89 (8.02)
11	$[Ni(L_2)_2(H_2O)_2](NO_3)_2$	862.7	Green	76	207	3.08	115	58.42 (58.67)	6.45 (6.53)	9.73 (9.50)	6.80 (7.03)
12	$[Cu(L_2)_2Cl_2]$	77.8	Green	66	290	2.15	21	64.78 (65.02)	6.68 (7.00)	7.19 (7.02)	8.09 (7.96)
13	$[Zn(L_2)_2 Cl_2]$	780	Yellow	64	271	Dia	14	64.61 (64.50)	6.67 (6.82)	7.18 (7.05)	8.33 (8.25)
14	$[Cd(L_2)_2] Cl_2$	829	Yellow	77	>300	Dia	125	60.94 (61.07)	6.28 (6.17)	6.77 (6.91)	13.54 (13.61)

5. Results and Discussion

Molar conductance values of complexes (1, 3, 5,6,8,10,12 and 13) in 10^{-3} M DMF are neutral .while complexes (2, 4,9,11 14) are 1:2 electrolytic nature for nitrate and chloride complexes. The molar conductance values and the metal contents are in a good agreement with given formulations (Geary ,1971),(Table 3).

1- Electronic Spectra and magnetic moment Measurements:

The electronic spectra of these two ligands $L_1$and L_2 and their complexes in DMF solution have been recorded giving ultraviolet spectra, d-d spectra and charge transfer spectra (Table 4).The ultraviolet spectra of the ligands exhibited two bands at (30487,36101) cm^{-1}, (27173,27777) cm^{-1} assigned to n $\rightarrow \pi^*$ (C=O) and $\pi \rightarrow \pi^*$ (C=N) transitions respectively. All these transitions were also found in the spectra of the complexes but they were shifted to lower frequencies confirming the coordination of the ligands to the metal ions (Lever,1968)· and these values agree with the magnetic moments obtained for cobalt(II) complexes 1,2,8 and 9 are 4.98,475,4.98 and 4.99 B.M. respectively ,and this values greater than theoretical value (3.87) B.M. due to orbital contribution (Nicholls ,1975,Suttan,1998). The electronic spectra of Co(II) complexes (1,2,8 and 9) exhibit three bands due to ν_1 at (11157-13177) cm^{-1} , ν_2 at (14816-15337) cm^{-1} and ν_3 at (16103-21141) cm^{-1} which are assigned to following transitions:

^4T_1g (F) $\longrightarrow$ ^4T_2g (F) ν_1 7500-11000 cm^{-1}
^4T_1g (F) $\longrightarrow$ ^4A_2g (F) ν_2 11000-16000 cm^{-}
^4T_1g (F) $\longrightarrow$ ^4T_1g (P) ν_3 18000-26000 cm$_{-}$

and these values agree with high spin octahedral configuration. While Ni(II) complexes (3,4,11and12) exhibit electronic spectrum bands at (11235 – 13071)cm^{-1} , (14104 – 18456) cm^{-1} and (20576 – 26737) cm^{-1} respectively , and assigned to transitions :

^3A_2g (F) $\longrightarrow$ ^3T_2g (F) ν_1
^3A_2g (F) $\longrightarrow$ ^3T_1g (F) ν_2
^3A_2g (F) $\longrightarrow$ ^3T_1g (P) ν_3

The octahedral geometry of Ni(II) ion in the complexes is confirmed by the measured magnetic moment values (2.89 – 3.28) B.M.(Mamata,et,al., 2008 and Sutton,1998).

The Cu(II) complexes (5 and 12) showed one band at (16780 , 18780) cm^{-1} respectively . This band may arise from the ^2Eg $^2E_{1g}$, This → band has been comparable both in position and width with the reported octahedral complexes , the magnetic moment obtained for these complexes are (2.07 , 2.15) B.M respectively (Baran,2013).(Table4).

Table 4 : Electronic spectra data of the complexes

Comp.No.	Electronic transition of d-d cm^{-1}			CT
	ν_1	ν_2	ν_3	
1	13177	14947	16103	33557
2	12297	15337	18103	31645
3	12195	16286	20576	38615
4	112235	14104	26737	35971
5	16780	-	-	32607
8	11157	14816	21141	33557
9	13333	16366	20491	36101
10	13071	18456	22123	31446
11	11627	16286	22935	37764
12	18796	-	-	31152

CT = Charg transfer band

2- Infrared Spectra :

The IR spectra of the free ligands (L_1 and L_2) showed band at 2956 cm^{-1} which is characteristic of stretching vibration (Mitu,et.al,2009) of the NH and other absorption band appeared at (1651, 1620) cm^{-1} which have been attributed to frequency of C=O amide group . This band in complexes is shifted to lower frequency indicating coordination of carbonyl oxygen atom to the metal (El-Faham,et.al.,2015) .A[so showed a strong band in the region (1589 , 1595) cm^{-1} , which is characteristic of the azomethine (stretching vibration of (C=N) group .This band in complexes is shifted to lower frequency indicating coordination of azomethine nitrogen atom to the metal (Al-Shaheen and Al-Bergas,2020). The spectra of L_1 and L_2 showed, a broad band at (1076, 1060) cm^{-1} due to N-N group shift towards lower frequency on complexation (Al-Shaheen,2017). The aqua complexes contain weak to medium abroad band at (3184 – 3392) cm^{-1} due to stretching vibration OH of water. Molecules are coordinated, confirmed by occurrence of additional strong band at (821 – 877) cm^{-1} due to OH rocking vibration (Gamo, 1960,Bellamy,1966). The spectra of the complexes showed bands in the region (536 – 628) cm^{-1} and (403 – 500) cm^{-1} are assigned to M-O and M-N stretching bands of the metal complexes(Sallomi and AI-Shaheen, 1994),) In addition the coordination of chloride could not be inferred from infrared spectra of the complexes because the band occurred beyond the range of our infrared spectrophotometer, whereas for Cl- ionic has been checked by $AgNO_3$. Chloride complexes 5 and 14 showed a band at 628 & 648 cm^{-1} has been attributed to ionic chloride (Cotton. and Wilkinson, 1980) On the other hand, the spectra of some complexes showed the presence of a band at (1373-1384) cm^{-1} due to ionic nature of nitrato group (Cotton. and Wilkinson, 1980) (Table 5).Fig. 2 and 3.

Table 5: Selected I.R. bands of the ligands and its complexes (in cm)$^{-1}$

Comp No.	υ (N-H)	υ (O-H)	υ (C=N)	υ (N-N)	υ (C=O)	υ (M-O)	υ (M-N)	Other
L_1	2956	3200-3300	1589	1076	1651	--	--	--
L_2	2956	-	1595	1060	1620	--	--	--
1	2958	3200-3300	1500	1031	1587	553	426	--
2	2960	3200-3300	1500	1022	1583	551	491	ν IonicNO$_3$ 1377, 761 ν-OH$_2$ 3300, 821
3	2958	3200-3300	1500	1020	1583	525	450	--
4	2953	3200-3300	1535	1010	1566	588	422	ν IonicNO$_3$ 1377, 761 ν-OH$_2$ 3300, 821
5	2954	3200-3300	1581	1018	1581	565	455	--
6	2958	3200-3300	1502	1015	1558	588	491	--
7	2950	3200-3300	1540	1037	1580	567	470	υ - ionic Cl - ,628
8	2956	--	1508	1029	1581	524	504	--
9	2954	--	1499	1008	1585	588	538	ν IonicNO$_3$ 1377, 761 ν-OH$_2$ 3300, 821
10	2958	-	1533	1031	1587	577	510	--
11	2951	--	1541	1049	1558	592	416	ν IonicNO$_3$ 1384, 823 ν-OH$_2$ 3392, 877
12	2954	--	1499	1012	1589	594	474	--
13	2958	--	1500	1022	1595	560	480	--
14	2954	--	1499	1022	1589	570	490	υ - ionic Cl - ,648

3- ^{1}HNMR spectra:

The proton nuclear magnetic resonance spectrum for the L_1 and L_2 was carried out using DMSO-d_6 as solvent and the following peaks were detected (Mohiuddina,2019) as shown in Table (6).

Table (6) : ^{1} H.NMR data of ligands L 1 and L2 , DMSO-d_6

Functional groups	) ppm(Chemical shift	
	L_1	L_2
$CH_3$2	0.81(d,6H) 0.85(d,6H)	0.82(d,6H) 0.85(d,6H)
CH_3	1.35(d,3H) 1.37(d,3H)	1.39(d,3H) 1.41(d,3H)
CH	1.75-183 (m,1H)	1.75-1.83 (m,1H)
N=C-CH_3	---	2.20(s,3H) 2.27(s,3H)
CH_2	2.37(d,2H) 2.41(d,2H)	2.37(d,2H) 2.40(d,2H)
CH-CO	3.64(q,1H) 4.57(q,1H)	3.98(q,1H) 4.70(q,1H)
OCH_3	3.80(s,3H) 3.85(s,3H)	---
Ar-H	6.77-7.28 (m,7H)	7.07-7.78 (m,9H)
N=CH	7.77(s,1H) 8.07(s,1H)	---
OH	9.47(s,1H) 9.53(s,1H)	---
NH	11.10(s,1H) 11.34(s,1H)	10.47(s,1H)

4- X-ray Powder Diffraction Analysis

The XRD patterns indicate a crystalline nature for metal complexes. Indexing of the diffraction patterns was performed using high Score Plus Software (Match program). For Co(II) and Cd(II) complexes, for example , their Miller indices (hkl) along with observed and calculated 2Ø angle , d values, and relative intensities , From the indexed data the unit cell parameters were also calculated and the powder XRD patterns of the compounds are completely different from those of the starting materials (Stou and. Jensen 1968), demonstrating the formation of coordination compounds. It is found that Co(II) and Cd(II) complexes have orthorhombic and tetragonal structure. Moreover , using diffraction data , the mean crystallite sizes of the complexes, D ,were determined according Scherrer equation ($D = 0.9\,\lambda\,/\,(\,\beta\ \cos\ \theta\,)$, where λ X -ray wavelength (1.5406 $A^{-\circ}$) , θ is

Bragg diffraction angle, and β is the full width at half maximum of the diffraction peak (Hong .. etal .,.2014) as shown in Table (7) .

Table 7: Crystal data and structure refinement for complexes

No	**Complex**	**1**
1	Molecular Formula	$CoC_{42}H_{52}N_4O_6Cl_2$
2	Molecular weight	838
3	Crystal system	Orthorhombic
4	Space group	P21 (42)
5	Unit cell parameters (A^{o})	a= 7.1692 A^{o} , b= 12.2813 A^{o} , c= 16.1892 A^{o}
6	Cell Volume (A^{o3})	962.33
7	Z	8
8	θ range , deg	4.17 – 42.17
9	Index ranges	$2 \le h \le 8$, $0 \le k \le 12$, $2 \le L \le 4$

No	**Complex**	7
1	Molecular Formula	$CdC_{42}H_{52}N_4O_6Cl_2$
2	Molecular weight	891
3	Crystal system	Tetragonal
4	Space group	P-4, 2,2
5	Unit cell parameters (A^{o})	a= 9.5142 A^{o} , b=9.5371 A^{o}, c= 8.2117 A^{o}
6	Cell Volume (A^{o3})	1017.2 A^{o}
7	Z	4
8	θ range , deg	95.72 – 22.34
9	Index ranges	$2 \le h \le 4$ $1 \le k \le 2$ $1 \le L \le 2$

CONCLUSION

From the FT-IR data showed that ligands L_1 and L_2 act as bidentate coordinated to the metal ions through oxygen carbonyl and azomethine nitrogen atoms. Also the above discussion of various physicochemical, spectral and according to the measurements of XRD analysis, the crystal geometries of some complexes has been established, and we concluded that the metal ions are hexacoordinate with most probable octahedral structure has been suggested for most complexes. Whereas cadmium complexes have tetrahedral geometries. Fig.(1).

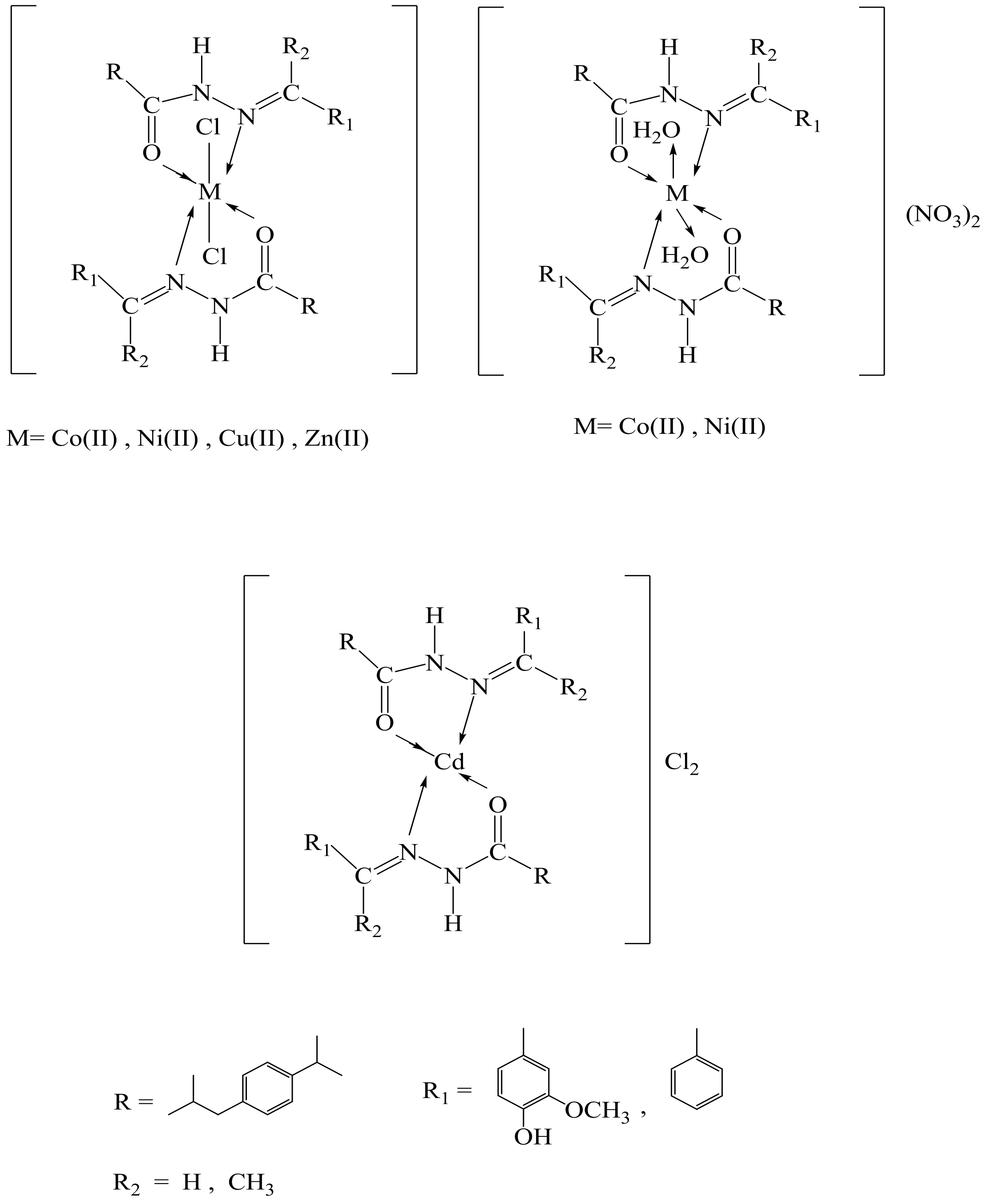


Fig.(1): The structure of the prepared complexes

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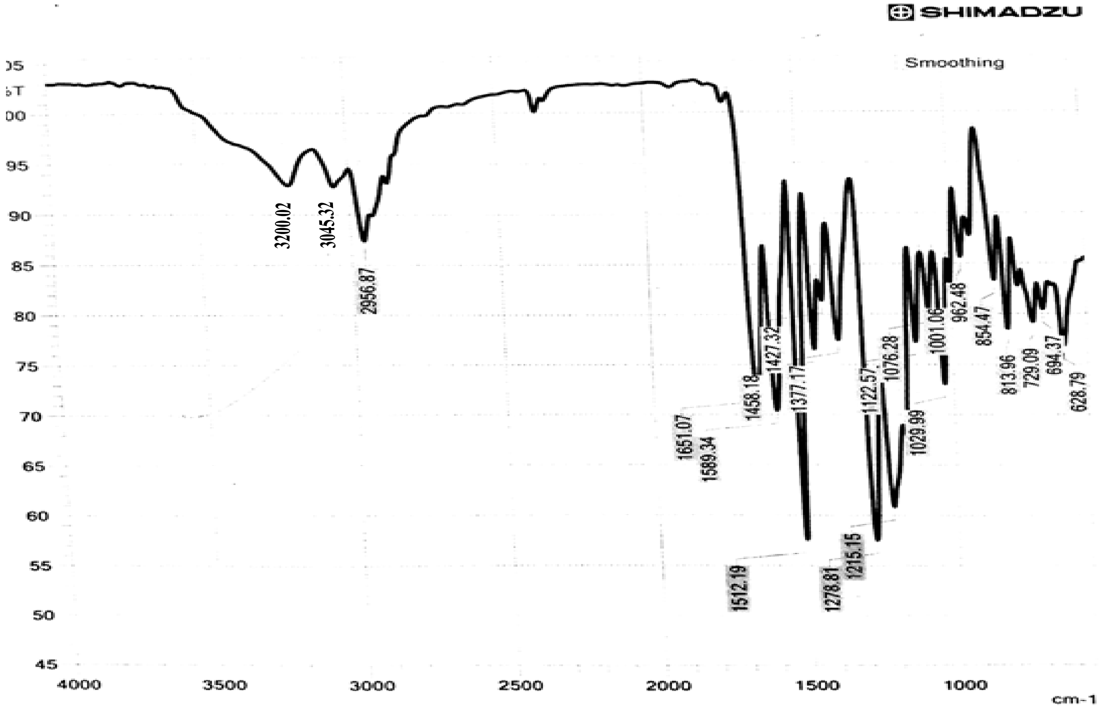

Fig (2): FT-IR Spectrum of (L_1)

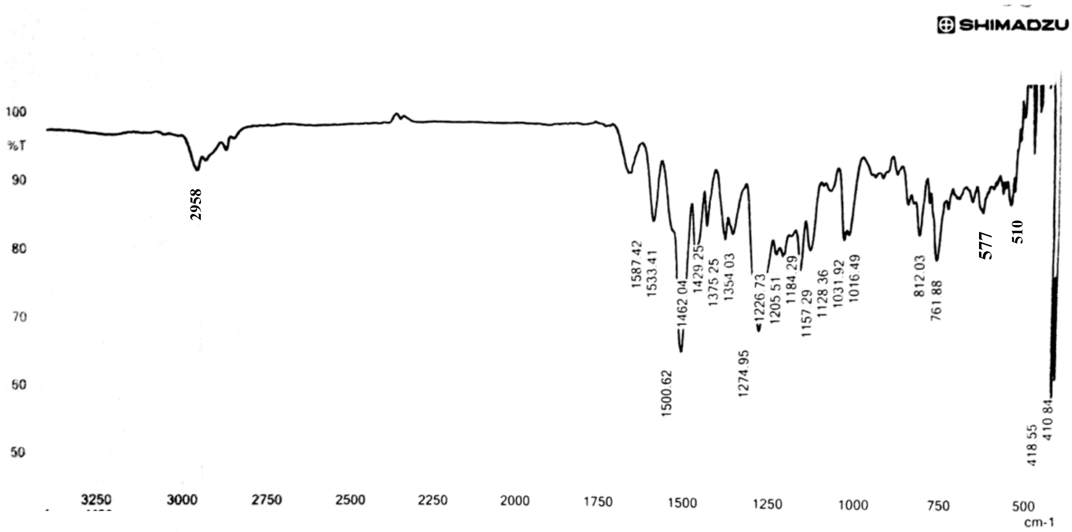

Fig (3): FT-IR Spectrum of complex (10)