

Preparation and Characterization of novel metal complexes containing hydrazone derived from camphor and 2,4-dinitrophenyl hydrazine

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Abstract

Ligand The development of 2,4-dinitrophenyl hydrazine with camphor under demonstrated conditions yielded another hydrazone ligand. The structures of metal (II) for example, copper, nickel, cobalt, zinc and iron with hydrazone have been composed from reacting between copper bromide, nickel chloride hexahydrate, cobalt chloride hexahydrate, zinc chloride, and iron bromide with hydrazone in 1:2 molar ratio. The ligand and their metal complexes have been detached in a solid state. The spectroscopic data of the structures proposes their 1:2 structures which are characterized by CHN, FT-IR and ¹H NMR spectroscopy. From spectroscopic reviews I proposed the octahedral structure for the all structures and the outcomes are spoken to and talked about underneath.

Keywords: 2,4-dinitrophenyl hydrazine, Complexes, hydrazone, metal complexes, camphor.

تحضير وتشخيص معقدات فلزية جديدة تحتوي هيدرازون مشتق من الكافور و 4,2- ثنائي نايتروفنيل هيدرازين

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الخلاصة:

تم تحضير الليكاند المستخدم في هذه الدراسة من تفاعل الكافور مع 4,2- ثنائي نايتروفنيل هيدرازين تحت ظروف تفاعل معينة ليتكون ليكاند الهيدرازون. معقدات العناصر الانتقالية حضرت من تفاعل املاح العناصر الانتقالية الثنائية مع الهيدرازون بنسبة مولية 1:2. الليكاند والمعقدات المحضرة انتجت بالحالة الصلبة وشخصت بواسطة التحليل العنصري الدقيق ومطيافية الاشعة تحت الحمراء والرنين النووي المغناطيسي, ومن هذه الدراسة الطيفية افترضنا تكون المعقدات بهيئة ثمانية السطوح, والبحث موضح في ادناه بالتفصيل.

الكلمات المفتاحية:

4,2- ثنائي نايتروفنيل هيدرازين, معقدات, هيدرازون, كافور, معقدات فلزية.

I. Introduction

Hydrazone subsidiaries have pulled in a lot of enthusiasm for manufactured science and extensive research on them in connection to their engineered

utility has been expert. Hydrazones are widely considered as reactants or response intermediates since they can promptly experience different ring conclusion reactions [Rollas *et al*, 2007] Hydrazones, RR-C=N-NR'R", are utilized as intermediates as a part of union

[Armbruster *et al*, 2006], as practical gatherings in metal carbonyls [Senturk *et al*, 2003], in natural compounds [Amr *et al*, 2003; Mohrle *et al*, 2003], hydrazones display physiological exercises in the treatment of a few sicknesses, for example, tuberculosis. This movement is ascribed to the arrangement of stable chelate buildings with move metals which catalyze physiological processes [Katyal *et al*, 1975; Mohan *et al*, 1988; Sinh *et al*, 1982]. They likewise go about as herbicides, bug sprays, nematocides, rodenticides, plant development controllers, sterilants for houseflies, among other applications [Katyal *et al*, 1975; Mohan *et al*, 1988; Molodykh *et al*, 1978]. In logical science hydrazones discover applications as multidentate ligands for move metals in colorimetric or fluorimetric determinations [Suez *et al*, 1994; Terra *et al*, 1999]. I report here the blend and portrayal of hydrazone Schiff base mixes. as of late the N-N linkage has been utilized as a key basic theme in different bioactive operators. Specifically, an expanding number of N-N bond-containing heterocycles and peptidomimetics have advanced into business applications as pharmaceutical and horticultural agents [Vicini *et al*, 2002; Mamolo *et al*, 2001]. The hydrazones have been exhibited to have antibacterial [Singh *et al*, 1979; Mohammed *et al*, 1991; El-Ashry *et al*, 2003; Almeida *et al*, 2008; Contelles *et al*, 2008; Kumar *et al*, 2009; Sherman *et al*, 2008]. Anticonvulsant [Saeed *et al*, 2008; Trofimov *et al*, 2008; Xia *et al*, 2008; Zheng *et al*, 2009]. what's more, antitubercular activities [Trofimov *et al*, 2008; Xia *et al*, 2008; Zheng *et al*, 2009; Sriram *et al*, 2006; Abou-Melha *et al*, 2008; El-Tabl *et al*, 2008; A- Joshi *et al*, 2008].

2. Experimental

2.1 Materials and Equipment:

All reagents were of analytical grade from Merck, Fluka, Sigma-Aldrich and BDH, all solvents used without further purification. ¹H NMR spectra were recorded in Bruker 300 MHz spectrometers with TMS as an inward reference using DMSO-d₆ dissolvable at Al- Albait University in Jordan. Infra-red spectra were recorded with KBr plates using a FTIR spectrophotometer Shimadzu show 8400 S in range 4000-400 cm⁻¹, all IR – charts recorded in Al-mustansiriyah university. Normal examination for Carbon, Hydrogen and Nitrogen were performed by using an Euro vector EA 3000A Elemental Analysis

(Italy) at Al- Albait university in Jordan. Dissolving motivations behind each and every solid compound were settled using a MPS10 electrically warmed melting point mechanical get together. Produced pathways are presented in Schemes 1-2 and physicochemical, apparition data for the as of late joined blends are given in Tables 1 and 2.

2.2 Preparation of (E)-1-(2,4-dinitrophenyl)-2-(1,7,7-trimethylbicyclo[2.2.1]heptan-2-ylidene)hydrazine:

To 2,4-dinitrophenyl hydrazine (0.01 mol) in ethanol (20 mL), camphor (0.01 mol) was included. The response blend was warmed under reflux for 4 h then poured onto a measuring glass containing an ice/water blend. The framed strong item was gathered by filtration and dried to give orange gems (from ethanol). Yield: 89 %, m.p. 188 - 189 °C.

2.3 Preparation of transition metal complexes:

The main products in all reactions were synthesis by this method:

the ligand of (E)- 1-(2,4-dinitrophenyl)- 2-(1,7,7-trimethylbicyclo[2. 2. 1] heptan-2-ylidene) hydrazine (0. 001 mol) was dissolved in (10 ml) of hot ethanol solution and mixed with CuBr₂, FeBr₂, ZnCl₂, NiCl₂.6H₂O and CoCl₂.6H₂O, or (0. 0005 mol) in (10 ml) of ethanol in mole ratio 1:2. The mixture was refluxed for 3 hrs. The mixture was cool at room temperature and the precipitated complex was then filtered off and dried. Then, the precipitate was recrystallized from methanol, those investigative What's more physical majority of the data of the edifices need aid recorded done table 1.

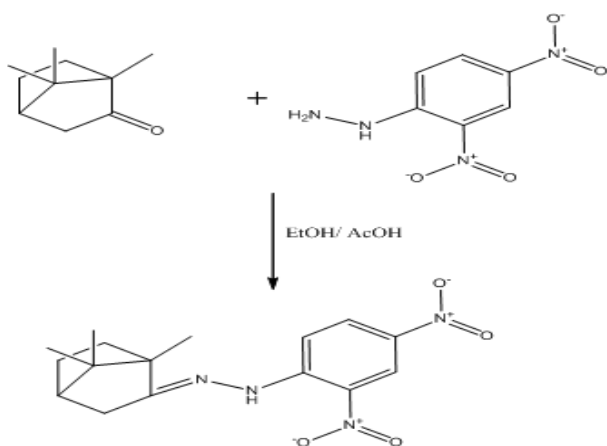
Table1. Analytical and physical data for new ligand and their complexes 1-5

Compounds	colour	Melting point (°C)	Analysis (%)			Yield %
			C	H	N	
Ligand (L)	Orange	188 – 189	57.82 (57.55)	6.07 (6.01)	16.86 (16.45)	89
Cu(L) ₂ (H ₂ O) ₂ [1]	Reddish-brown	181 – 183	50.42 (50.31)	5.55 (5.46)	14.70 (14.75)	81
Fe(L) ₂ (H ₂ O) ₂ [2]	Brown	152 – 153	50.94 (50.70)	5.61 (5.35)	14.85 (14.38)	78
Zn(L) ₂ (H ₂ O) ₂ [3]	Orange	171 – 173	50.30 (50.22)	5.54 (5.43)	14.66 (14.81)	76
Ni(L) ₂ (H ₂ O) ₂ [4]	Orange	191 – 192	50.74 (50.31)	5.59 (5.36)	14.79 (14.51)	66
Co(L) ₂ (H ₂ O) ₂ [5]	Orange	185 – 186	50.73 (50.34)	5.59 (5.63)	14.79 (14.63)	82

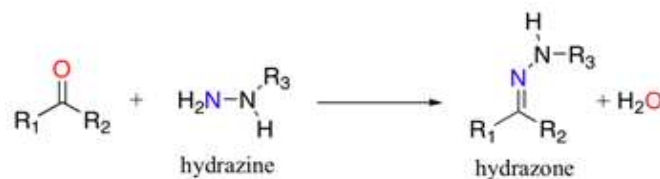
Calculated values are in parentheses

3. Results and discussion:

The compound (E)- 1-(2,4-dinitrophenyl)- 2-(1,7,7-trimethylbicyclo [2.2.1] heptan-2-ylidene)hydrazine is shaped in great yield by the reacting of 2,4-dinitrophenyl hydrazine with camphor. Basic scientific information (Table 1) of the mixes recommend that the response between 2,4-dinitrophenyl hydrazine and camphor happened in the 1:1 proportion as in Scheme 1. Also, the instrument of hydrazone arrangement response as Scheme 2. [A- Joshi *et al*, 2008]

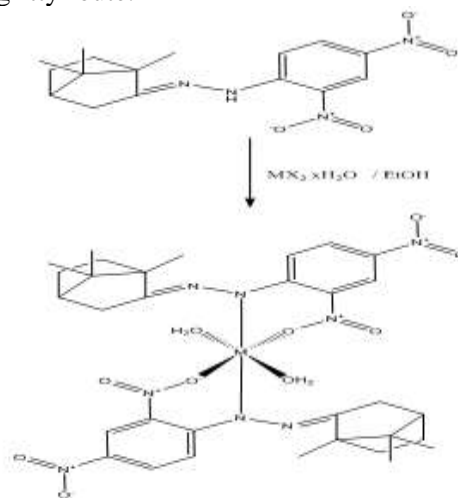


SCHEME 1. Preparative method for (E)-1-(2,4-dinitrophenyl)-2-(1,7,7-trimethylbicyclo[2.2.1]heptan-2-ylidene)hydrazine



SCHEME 2. General mechanism to form hydrazone compounds

(E)- 1-(2,4-dinitrophenyl)- 2-(1,7,7-trimethylbicyclo[2. 2. 1] heptan-2-ylidene)hydrazine may be crystalline to way What's more may be dissolvable done such as way regular solvents. Its confined constantly on around described Furthermore crystalline edifices for Copper (II), Nickel (II), Cobalt (II), Zinc (II) and iron (II) particles. The aftereffects of essential examination (CHN) with sub-atomic structure and the dissolving keeps tabs need aid presented to table 1. The conclusions obtained would over incredible simultaneousness for the individuals figured for the recommended mathematical statement and the softening concentrates need aid sharp, demonstrating the ethicalness of the orchestrated hydrazone and their edifices. Those structures of the hydrazone and their structures under investigation need aid provided for underneath (Scheme 3). Those structures about these mixes will be Moreover asserted Eventually IR and ¹H NMR spectra, which will a chance to be analyzed on a nitty gritty route.



Where $MX_2 \cdot xH_2O$ = $CuBr_2$, $FeBr_2$, $ZnCl_2$, $NiCl_2 \cdot 6H_2O$ and $CoCl_2 \cdot 6H_2O$.

SCHEME 3. Preparative techniques for edifices (1:2 molar proportion)

3.1 Infrared spectra :

The IR spectra of ligand and their complexes are presented in Table 2. The IR spectra of the ligands and complexes synthesized are shows strong band appeared at range (2958 - 2968) cm^{-1} and (3041 - 3107) cm^{-1} due to stretching aliphatic (C-H) and aromatic (C-H).²⁹
³⁰ The IR spectra of the complexes were compared with of the free ligand (L) in order to determine the coordination sites that may be involved in chelating. There was some guide peaks in the spectra of the ligands, which were helpful in achieving this goal. The position and/or the intensities of these peaks are expected to change upon chelating. New peak is also guide peak, as M-N and M-O in chelating.

Table 2. data of IR Spectra for Ligand and compounds 1-5

Ligand complexes IR (cm^{-1})	λ_{max} in nm (μm in $\text{M}^{-1}\text{cm}^{-1}$)
Ligand (L)	3086 (C-H) Ar., 2958 (C-H) aliph., 1489 (C=C) Ar., 3323 (N-H), 1739 (C=N), 1107 (C=N) aliph., 1570 (N=O), 1389 (N-O)
Cu(L) ₂ (H ₂ O) ₂ [1]	3088 (C-H) Ar., 2964 (C-H) aliph., 1419 (C=C) Ar., 3352 (O-H), 1612 (C=N), 1058 (C=N) aliph., 1535 (N=O), 1332 (N-O), 464 (M-N), 505 (M-O)
Fe(L) ₂ (H ₂ O) ₂ [2]	3088 (C-H) Ar., 2958 (C-H) aliph., 1421 (C=C) Ar., 3421 (O-H), 1612 (C=N), 1049 (C=N) aliph., 1539 (N=O), 1336 (N-O), 482 (M-N), 518 (M-O)
Zn(L) ₂ (H ₂ O) ₂ [3]	3041 (C-H) Ar., 2964 (C-H) aliph., 1419 (C=C) Ar., 3551 (O-H), 1610 (C=N), 1053 (C=N) aliph., 1541 (N=O), 1340 (N-O), 482 (M-N), 518 (M-O)
Ni(L) ₂ (H ₂ O) ₂ [4]	3107 (C-H) Ar., 2960 (C-H) aliph., 1419 (C=C) Ar., 3336 (O-H), 1614 (C=N), 1060 (C=N) aliph., 1543 (N=O), 1332 (N-O), 478 (M-N), 514 (M-O)
Co(L) ₂ (H ₂ O) ₂ [5]	3095 (C-H) Ar., 2968 (C-H) aliph., 1419 (C=C) Ar., 3336 (O-H), 1616 (C=N), 1058 (C=N) aliph., 1543 (N=O), 1332 (N-O), 478 (M-N), 514 (M-O)

Table 2 are showed this guide apex . Upon relationship These band was moved to higher or lower wave numbers in the structures, showing the participation of the nitrogen in coordination (M—N). [Silerstien *et al*, 2005; Shriner *et al*, 2004] Several medium appeared in the (464 - 482) cm^{-1} region of the spectra are a direct result of the stretching (M—N) bond. [Silerstien *et al*, 2005; Shriner *et al*, 2004] New aggregation are found in the spectra of the structures in the zones (505 - 518) cm^{-1} which are allotted to ν (M-O) amplifying vibrations for metal structures. [Silerstien *et al*, 2005; Shriner *et al*, 2004] The IR spectra of the ligand and buildings incorporated are shows two aggregation appeared at range (1049 - 1107) cm^{-1} and (1610 - 1739) cm^{-1} in light of augmenting aliphatic (C-N) and (C=N) respectively. [Silerstien *et al*, 2005; Shriner *et al*, 2004] Therefore; from the IR spectra it is assumed that the ligand carries on as a fair bidentate ligand encouraged to the metal molecule by method for

N and O particle in nitro aggregate. [Silerstien *et al*, 2005; Shriner *et al*, 2004].The IR spectra for ligand and it's complexes as shown in Fig. (1-6).

3.2 ¹H NMR spectra

¹H NMR spectra in deturated DMSO are given in Table 1 explain in Figure (7 – 13) and band data in Table 3, The ¹H NMR spectra of the hydrazone in DMSO indicates doublet signals inside the 7.48 ppm and 8.58 ppm are refers to H18 and H15 in complexes while in ligand appeared as doublet signals at 7.97 ppm and 8.67 ppm. H17 has been appeared in 7.99 ppm as doublet signals in ligand while in complexes has been showed at 8.45 as quartet signals. The spectra of the structures are differed in Comparison with those of the parent compound (hydrazone). The ¹H NMR spectra of the hydrazone united from camphor and 2,4-dinitrophenyl hydrazine revealed a banner at 9.63 ppm (singlet) in light of the proton NH and The proton NH has vanished in ¹H NMR spectra of structures hydrazone, demonstrating that the NH proton is removed by chelation with the metal ion. [Silerstien *et al*, 2005; Shriner *et al*, 2004]

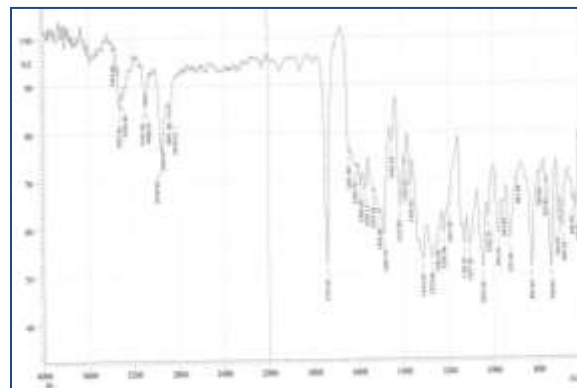


Figure (1): IR spectra for ligand

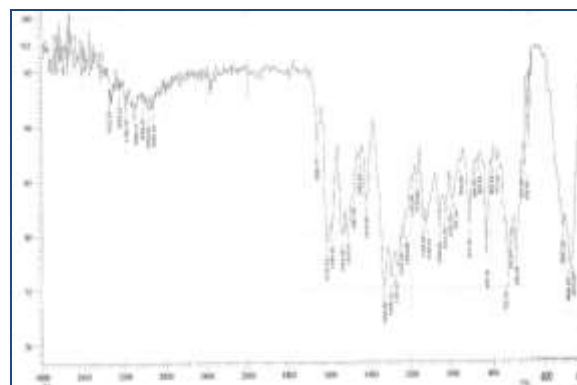


Figure (2): IR spectra for Complex 1

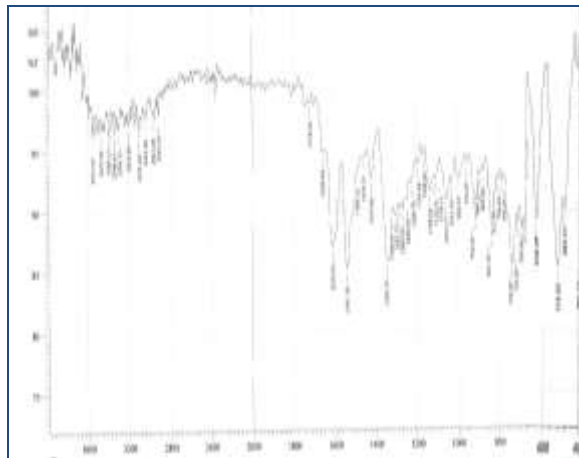


Figure (3): IR spectra for Complex 2

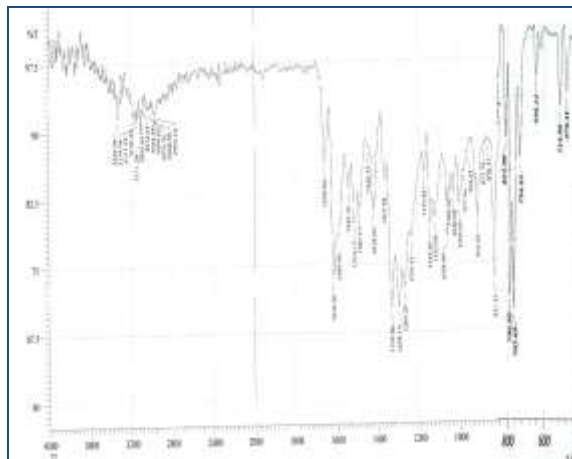


Figure (6): IR spectra for Complex 5

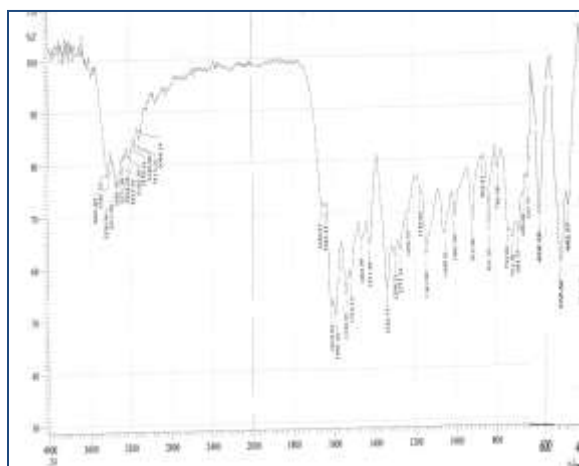


Figure (4): IR spectra for Complex 3

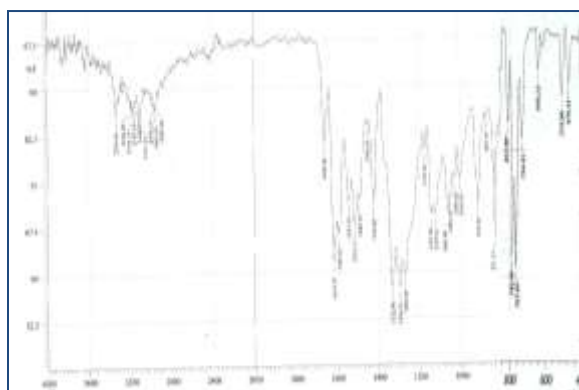


Figure (5): IR spectra for Complex 4

Table 3. ¹H NMR Spectroscopic data for Ligand and compounds 1-5

Ligand complexes	Chemical shift (ppm)
Ligand (L)	0.94 (s, 3H, 1), 0.95 – 1.10 (m, 6H, 10 & 11), 1.44 – 1.55 (m, 2H, 5), 2.69 – 2.73 (m, 4H, 7 & 8), 7.97 (d, 1H, 17), 8.67 (d, 1H, 18), 8.58 (d, 1H, 15), 9.63 (s, 1H, 12)
Cu(L) ₂ (H ₂ O) ₂ [1]	0.88 (s, 6H, 1), 0.93 (d, 12H, 10 & 11), 1.86 – 1.91 (m, 4H, 5), 1.95 – 2.03 (m, 4H, 7), 1.35 – 1.41 (m, 4H, 8), 7.48 (d, 2H, 17), 8.45 (q, 2H, 18), 8.58 (d, 1H, 15)
Fe(L) ₂ (H ₂ O) ₂ [2]	0.88 (s, 6H, 1), 0.93 (d, 12H, 10 & 11), 1.86 – 1.91 (m, 4H, 5), 1.95 – 2.03 (m, 4H, 7), 1.35 – 1.41 (m, 4H, 8), 7.48 (d, 2H, 17), 8.45 (q, 2H, 18), 8.58 (d, 1H, 15)
Zn(L) ₂ (H ₂ O) ₂ [3]	0.88 (s, 6H, 1), 0.93 (d, 12H, 10 & 11), 1.86 – 1.91 (m, 4H, 5), 1.95 – 2.03 (m, 4H, 7), 1.35 – 1.41 (m, 4H, 8), 7.48 (d, 2H, 17), 8.45 (q, 2H, 18), 8.58 (d, 1H, 15)
Ni(L) ₂ (H ₂ O) ₂ [4]	0.88 (s, 6H, 1), 0.93 (d, 12H, 10 & 11), 1.86 – 1.91 (m, 4H, 5), 1.95 – 2.03 (m, 4H, 7), 1.35 – 1.41 (m, 4H, 8), 7.48 (d, 2H, 17), 8.45 (q, 2H, 18), 8.58 (d, 1H, 15)
Co(L) ₂ (H ₂ O) ₂ [5]	0.88 (s, 6H, 1), 0.93 (d, 12H, 10 & 11), 1.86 – 1.91 (m, 4H, 5), 1.95 – 2.03 (m, 4H, 7), 1.35 – 1.41 (m, 4H, 8), 7.48 (d, 2H, 17), 8.45 (q, 2H, 18), 8.58 (d, 1H, 15)

The spectra of the critical number of mixes display a singlet signals at the degree 0.94 ppm and 0.88 ppm in light of the protons CH₃ (H1) of the ligand in hydrazone and it's structures respectively. The doublat signals at 0.93 ppm proposed the attribution of the proton of the CH₃- (H10 & H11) in all new complexes were prepared while the CH₃- (H10 & H11) in ligand has been appeared at the range (0.95 – 1.10) as multiple signals. [Silerstien *et al*, 2005] The spectra of the number of blends exhibit a different banner at the range 1.86 – 1.91 ppm, 1.95 – 2.03 ppm and 1.35 – 1.41 ppm due to the protons CH and CH₂- (H5, H7 & H8) of the hydrazone compound in complexes respectively but in hydrazone ligand has been showed at the range 1.44 – 1.55 ppm and 2.69 – 2.73 ppm for H5, H7 & H8 respectively. [Silerstien *et al*, 2005; Shriner *et al*, 2004] The ¹H NMR spectrum of ligand,

Fig. 8, showed a doublet signals at 7.97 ppm, 8.67 ppm and 8.58 ppm, corresponding to the phenyl protons (H17, H18 and H15). A doublet signal centered at 7.48 ppm and 8.58 ppm due to H17 & H15 in all complexes referring to phenyl ring while the quartet signal can be appeared at 8.45 ppm due to H18 in all complexes, [Silerstien *et al*, 2005; Shriner *et al*, 2004] Table 3. And Fig. (7 – 13). Observed through spectroscopic study that all new complexes prepared have the same frequency of the protons due to the similarity of ligand used in the preparation of these complexes.

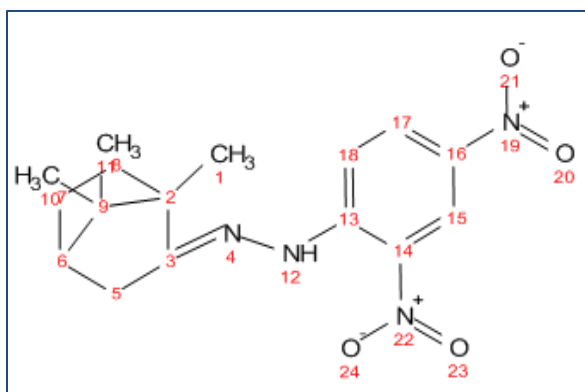
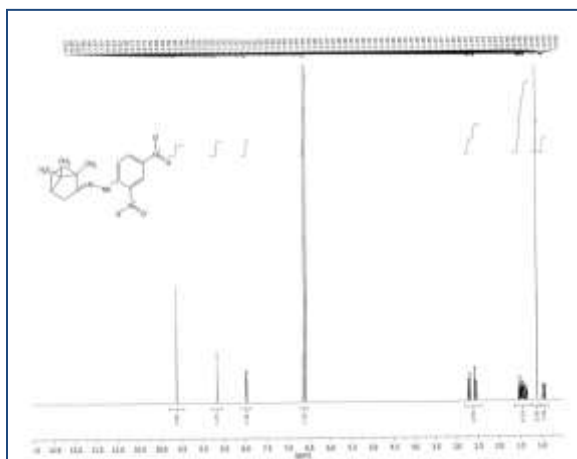
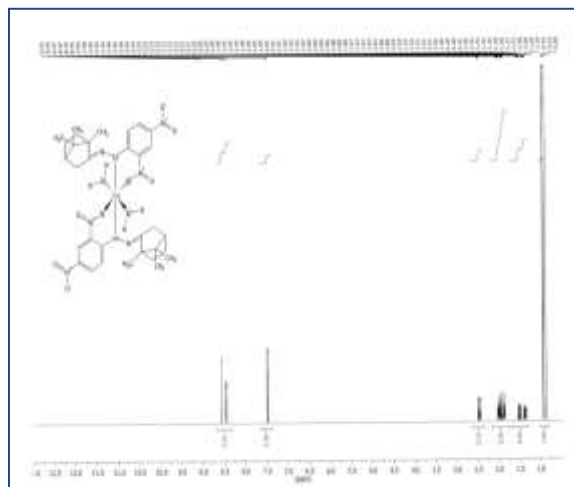
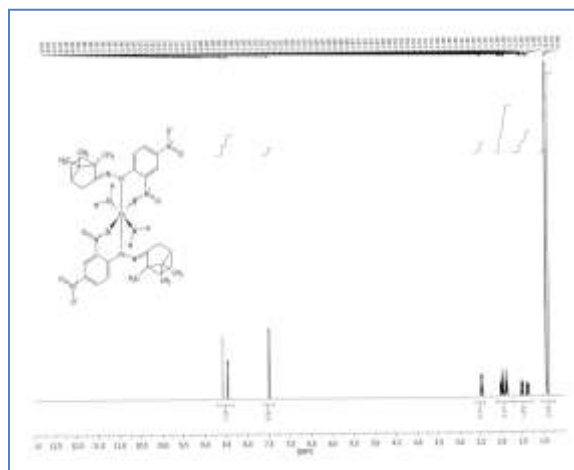
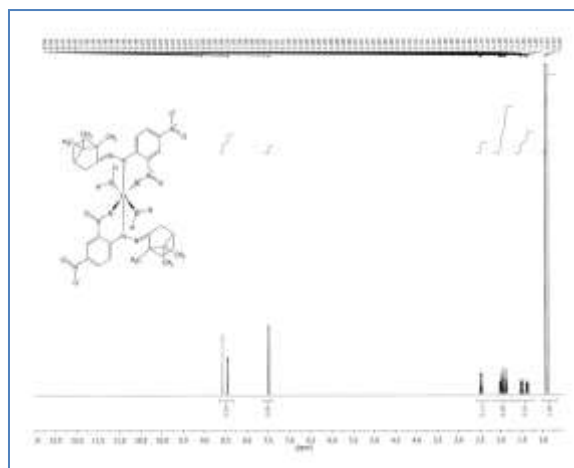
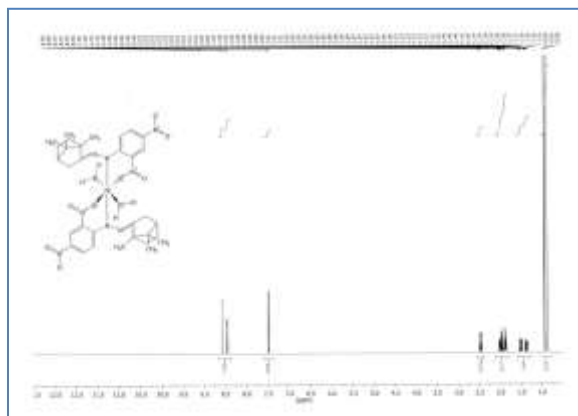
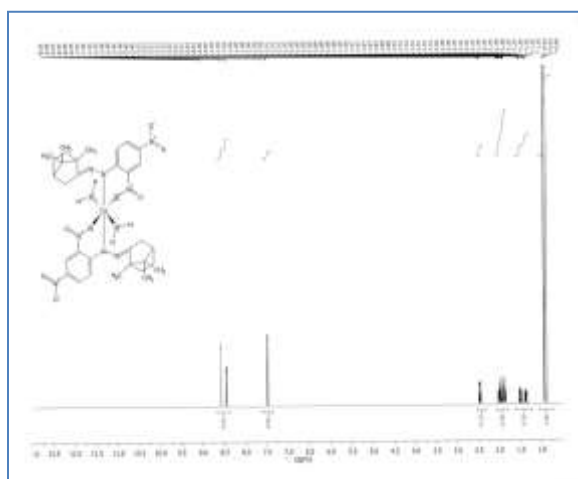


Figure (7): ligand form with tags

Figure (8): ^1H NMR spectra for ligandFigure (9): ^1H NMR spectra for complex 1Figure (10): ^1H NMR spectra for complex 2Figure (11): ^1H NMR spectra for complex 3

Figure (12): ¹H NMR spectra for complex 4Figure (13): ¹H NMR spectra for complex 5

Conclusion:

Another hydrazone ligand has been set up by the development of camphor with ,4-dinitrophenyl hydrazine. Logical IR, ¹HNMR and essential examination (CHN) revealed a 1:2 thing. Hydrazones and their subordinantes constitute an adaptable class of blends in common science. In another review that These blends have interesting natural properties, for instance, quieting, torment easing, anticonvulsant, antituberculous, antitumor, against HIV and antimicrobial development. Hydrazones are crucial blends for pharmaceutical arrangement, as could be permitted ligands for metal structures, organocatalysis besides for the amalgamations of heterocyclic blends.

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