Int. J. Chem. Sci.: 9(3), 2011, 1307-1318
ISSN 0972-768X
www.sadgurupublications.com

STUDIES ON THE COORDINATION COMPOUNDS OF THIAZOLIDIN-4-ONE DERIVED FROM SALICYLALDEHYDE-o-HYDROXYPHENYLUREA

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ABSTRACT

An equimolar mixture of salicylaldehyde and *o*-hydrxyphenylurea upon refluxing in MeOH forms the Schiff base **(1)**. The latter upon reacting with $HSCH_2COOH$ in dry C_6H_6 undergoes cycloaddition reaction and forms the corresponding thiazolidin-4-one, LH_3 **(2)**. A MeOH solution of 2 reacts with Cu (II), Co (II), Ni (II), MoO_2 (VI) and Zr $(OH)_2$ (IV) ions and forms the monomeric, non-electrolyte coordination compounds, $[Cu(OAc)(LH_2)]$ **(3)**, $[M(OAc)(LH_2)(MeOH)_2]$ **(4)** (here M = Co, Ni), $[MoO_2(acac)(LH_2)(MeOH)]$ **(5)** and $[Zr(OH)_3(LH_2)]$ **(6)**. A THF solution of a bidentate ON or NN donor ligand (AA) like ethanolamine (ea), *o*-aminophenol (oap), ethylenediamine (en), *o*-phenylenediamine (phen), 2,2'-bipyridyl (bipy) and 1,10-phenanthroline (ophen) reacts with a THF solution of **(6)** and forms the heterochelate, $[Zr(OH)_3(LH_2)(AA)]$ **(7)**. The coordination compounds have been characterized on the basis of elemental analyses, molecular weight, molar conductance, spectral (IR, reflectance, NMR) studies and magnetic susceptibility measurements. **(2)** behaves as a monobasic tridentate ONO donor ligand in these coordination compounds. A square-planar structure for **(3)**, an octahedral structure for **(4)** (M = Co, Ni) and **(6)**, and an eight-coordinate structure for **(5)** and **(7)** are suggested.

Key words: Thiazolidin-4-one, Coordination compounds, Spectral studies, Magnetic susceptibility, Heterochelates.

INTRODUCTION

Thiazolidin-4-ones belong to an important group of heterocyclic compounds with carbonyl group at fourth position[1,2]. They show broad spectra of biological activities due to their ready accessibility and diverse chemical reactivity[3]. They are involved in variety of applications such as, antimicrobial, antibacterial, anticonvulsant, antifungal, anti-HIV,

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antiproliferative, anti-inflammatory, antithyroid etc.[4-12]. Many drugs possess modified pharmacological properties in the form of the metal complexes[13,14]. These facts prompted us to explore the coordination behavior of thiazolidin-4-one derived from the Schiff base **(1)** towards Cu (II), Co (II), Ni (II), MoO_2 (VI) and Zr $(OH)_2$ (IV) ions.

EXPERIMENTAL

Materials

Ethylenediamine (Sarabhai); copper (II) acetate monohydrate (IDPL); cobalt (II) acetate tetrahydrate, *o*-aminophenol, hexadecaaquaoctahydroxotetrazirconium (IV) chloride (BDH); nickel acetate tetrahydrate (Fluka); ethanolamine, *o*-phenylenediamine, 2,2'-bipyridyl, 1,10-phenanthroline (Aldrich); ammonium molybdate tetrahydrate, $NaHCO_3$, MeOH, $HSCH_2COOH$, dry C_6H_6, THF, pet. ether (Ranbaxy) were used as supplied for the syntheses. Bis (acetylacetonato)dioxomolybdenum (VI) and hexadecaaquooctahydroxotetrazirconium (IV) acetate were prepared by adopting the published procedures[15,16].

Analyses and physical measurements

The metal contents of the complexes were estimated as per reported methods[17]. The C, H and N contents of LH_3 and its coordination compounds were estimated by CHN Eager analyzer model-300. The S contents was estimated gravimetrically[18] as $BaSO_4$. The molecular weights were determined by the Rast method using diphenyl as the solvent[19]. The molar conductivity measurements were carried out in DMF with the help of a Toshniwal conductivity bridge (CL01-02A) and a dip type cell calibrated with KCl solution The IR spectra were recorded in KBr pellets (4000-400 cm^{-1}) on a Beckman-20 spectrophotometer. The reflectance spectra were recorded on a Beckmann DU spectrophotometer attached with a reflectance arrangement. The ^1H NMR spectra of **(1)** and **(3)** (when M = Zn) were recorded on Bruker Avance II 400 NMR spectrometer. Magnetic susceptibility measurements were carried out at room temperature with a Gouy balance using mercury (II) tetraisothiocyanatocobaltate (II) ($\chi_g = 16.44 \times 10^{-6}$ cgs unit).

Synthesis of o–hydroxyphenylurea (ohypu)

o-Aminophenol (10.9 g, 0.1 mol) was dissolved in 20 mL of 1 : 1 HCl. To this solution, urea (24.0 g, 0.4 mol) in small quantity with constant stirring, distilled water (30 mL), HCl (1 mL) and glacial acetic acid (1 mL) were added. The brown solution obtained was refluxed on a water bath for 2 *h*. The light-brown compound separated on cooling was suction filtered, washed with distilled water and recystallised from 50% EtOH and dried *in vacuo*. m.p. = 154 °C, Yield = 90%; Anal. [$C_7H_8N_2O_2$; Found (calcd.) % C = 55.38 (55.26);

H = 5.32 (5.26); N = 18.23 (18.42)]; IR bands: ν(N–H) (3050 cm^{-1}), ν(C=O) (carbonyl) (1655 cm^{-1}), δ (NH_2) (1630 cm^{-1}) and ν(C–O) ϕ (1520 cm^{-1}).

Synthesis of (1)

An EtOH solution (50 mL) of *o*-hydroxyphenylurea (15.2 g, 0.1 mol) and salicylaldehyde (12.2 g, 0.1 mol) were refluxed on a water bath for 1 *h*. The excess of solvent was evaporated and the mixture was allowed to stand at room temperature. The brown compound separated out, was suction filtered, washed with and recrystallized from EtOH. The compound was dried as mentioned above. m.p. = 162°C, Yield = 70%; Anal. [$C_{14}H_{12}N_2O_3$; found (calcd) % C = 65.58 (65.62); H = 4.75 (4.69); N = 10.88 (10.94)]; IR bands: ν(N–H) (2950 cm^{-1}), ν(C=O) (carbonyl) (1665 cm^{-1}), ν(C=N) (azomethine) (1620 cm^{-1}) and ν(C–O) ϕ (1526 cm^{-1}).

Synthesis of (2)

A dry C_6H_6 solution of **(1)** (2.56 g, 10 mmol) and $HSCH_2COOH$ (0.92 g, 10 mmol) were refluxed for 12 *h* on a water bath. The mixture was cooled to room temperature and then washed with 10% $NaHCO_3$ solution. C_6H_6 layer was separated using a separating funnel. The partial evaporation of C_6H_6 layer gave a solid product, which was filtered, washed with and recrystallized from pet. ether. The compound was dried as mentioned above. Yield = 15%. [*Anal*: $C_{16}H_{14}N_2O_4S$ found (calcd) % C = 58.67 (58.18); H = 4.36 (4.24); N = 8.35 (8.48), S = 9.57 (9.70)]; IR bands (KBr): ν(O–H) (intramolecular H-bonding) (2870 cm^{-1}), ν(C=O) (thiazolidinone ring) (1710 cm^{-1}), ν(C=O) (amide) (1665 cm^{-1}), ν(C–N) (thiazolidinone ring) (1575 cm^{-1}), ν(C–O) (phenolic) (1528 cm^{-1}) and ν(C–S) (thiazolidinone ring) (832 cm^{-1}).

Syntheses of (3-6)

A MeOH solution (30-50 mL) of the appropriate metal salt (5 mmol) was added to a MeOH solution (50 mL) of **(2)** (1.65 g, 5 mmol) and the mixture was then refluxed for 3-4 *h*. The solid products formed were suction filtered, washed repeatedly with MeOH and were then dried as mentioned above. Yield = 50-65%.

Syntheses of (7)

A THF solution (~20 mL) of appropriate AA (2 mmol) was added to a THF solution of 6 (0.94 g, 2 mmol). The mixture was refluxed on a water bath for 1 *h*. The excess of the solvent was evaporated and the separated compound was suction filtered. It was washed with and recrystalised from THF and dried as mentioned above. Yield = 70-85%.

RESULTS AND DISCUSSION

The cycloaddition reaction of the Schiff base **(1)** with $HSCH_2COOH$ in dry C_6H_6 results in the formation of LH_3 **(2)**. The reaction of a MeOH solution of **(2)** with appropriate metal salts in 1 : 1 molar ratio on refluxing produces respective stable coordination compounds **(3-6)**. The reaction of **(6)** with AA (where AA = ON or NN donor ligands) in THF results in the formation of its respective heterochelates **(7)**. The compounds, **(4)** (M = Co, Ni) and **(5)** do not lose weight on heating at 120°C for hours, indicating that MeOH molecule (s) are not lost but are coordinated to metal ion. The coordination compounds and heterochelates are monomers in diphenyl. They are insoluble in H_2O, MeOH, EtOH and soluble in DMSO and DMF. Their molar conductance measurements (Λ_M = 3.3-9.6 mho cm^2 mol^{-1}) (Table 1) indicate their non-electrolytic nature[20]. The heterochelates are more stable than the precursor **(6)**.

Table 1: Analytical, molar conductance and molecular weight of compounds

Compd.	Mol. formula	Λ_M (mho cm^2 mol^{-1})	Mol. wt. obs. (calcd.)	Found (calcd.) %				
				M	C	H	N	S
1	$C_{14}H_{12}N_2O_3$	–	256[a] (256)	–	65.58 (65.62)	4.75 (4.69)	10.88 (10.94)	–
2	$C_{16}H_{14}N_2O_4S$	–	330[a] (330)	–	58.67 (58.18)	4.36 (4.24)	8.35 (8.48)	9.57 (9.70)
3	$CuC_{18}H_{16}N_2O_6S$	7.3	467.4 (451.5)	13.94 (14.06)	47.65 (47.84)	3.63 (3.54)	6.31 (6.20)	7.15 (7.09)
4 (M = Co)	$CoC_{20}H_{24}N_2O_8S$	4.3	523.6 (510.9)	11.64 (11.53)	46.83 (46.98)	4.56 (4.70)	5.62 (5.48)	6.37 (6.26)
4 (M = Ni)	$NiC_{20}H_{24}N_2O_8S$	6.8	522.6 (510.7)	11.37 (11.49)	46.87 (46.99)	4.85 (4.70)	5.59 (5.48)	6.41 (6.26)
5	$MoC_{22}H_{24}N_2O_9S$	9.6	565.6 (587.9)	16.37 (16.31)	44.67 (44.91)	4.13 (4.08)	4.61 (4.76)	5.57 (5.44)
6	$ZrC_{16}H_{16}N_2O_7S$	5.3	495.5 (471.2)	19.18 (19.35)	40.93 (40.75)	3.28 (3.40)	5.77 (5.94)	6.63 (6.79)

Cont…

Compd.	Mol. formula	Λ_M (mho cm^2 mol^{-1})	Mol. wt. obs. (calcd.)	Found (calcd.) %				
				M	C	H	N	S
7 (AA = ea)	$ZrC_{18}H_{23}N_3O_8S$	8.2	548.4 (532.2)	17.09 (17.14)	40.51 (40.59)	4.37 (4.32)	7.83 (7.89)	6.08 (6.01)
7 (AA = oap)	$ZrC_{22}H_{23}N_3O_8S$	4.5	568.3 (580.2)	15.67 (15.72)	45.55 (45.50)	3.91 (3.96)	7.29 (7.24)	5.47 (5.52)
7 (AA = en)	$ZrC_{18}H_{24}N_4O_7S$	6.3	548.8 (531.2)	17.22 (17.17)	40.61 (40.66)	4.56 (4.52)	10.59 (10.54)	5.97 (6.02)
7 (AA = phen)	$ZrC_{22}H_{24}N_4O_7S$	5.5	558.4 (579.2)	15.79 (15.74)	45.52 (45.58)	4.09 (4.14)	9.63 (9.67)	5.57 (5.52)
7 (AA = bipy)	$ZrC_{26}H_{24}N_4O_7S$	4.6	645.2 (627.2)	14.58 (14.54)	49.69 (49.74)	3.88 (3.83)	8.96 (8.93)	5.06 (5.10)
7 (AA = ophen)	$ZrC_{28}H_{24}N_4O_7S$	3.3	638.8 (651.2)	14.06 (14.00)	51.54 (51.60)	3.72 (3.68)	8.54 (8.60)	4.94 (4.91)

[a]Mass spectral data

IR spectra

The IR spectra of the compounds **(1-7)** were recorded in KBr and the important peaks are presented in Table 2. The Schiff base **(1)** exhibits the ν(C=O) (carbonyl), ν(C=N) (azomethine) and ν(C–O) ϕ stretches at 1665, 1620 and 1528 cm^{-1}, respectively. The disappearance of the ν(C=N) (azomethine) stretch (1620 cm^{-1}) of **(1)** and the appearance of a new band at 1575 cm^{-1} due to the ν(C–N) (thiazolidinone ring) stretch[21] in **(2)** suggests the formation of the corresponding thiazolidin-4-one. The formation of **(2)** is further supported by the appearance of a new band at 832 cm^{-1} due to the ν(C–S) (thiazolidinone ring) stretch[22]. Both these bands remain unchanged in the coordination compounds indicating the non-involvement of both N and S atoms of thiazolidin-4-one moiety towards the coordination. The ν(C=O) (thiazolidinone ring) stretch[23] of **(2)** occurring at 1710 cm^{-1} undergoes a negative shift by 26-38 cm^{-1} in **(3-7)** indicating the coordination through carbonyl O atom of thiazolidin-4-one moiety. **(2)** occurs in the keto form as evident by the presence of a strong band at 1665 cm^{-1} due to the ν(C=O) (amide) stretch[24]. This band remains almost at the same energy in **(3-7)**, which rules out the involvement of keto O atom towards coordination. The strong band occurring at 2870 cm^{-1} in **(2)** due to the intramolecular H-bonded OH group of the phenolic moiety disappears in **(3-7)** indicating the breakdown of H-bonding and the subsequent deprotonation of OH group followed by the involvement of phenolic O atom towards coordination[25]. The ν(C–O) (phenolic) stretch of **(2)** occurring at 1528 cm^{-1}, undergoes a positive shift by ≤ 10 cm^{-1} in **(3-7)** supports the involvement of phenolic O atom towards coordination[26]. The magnitude of the above shift of the ν(C–O) (phenolic) stretch supports the monomeric structures of **(3-7)** as suggested by molecular weight measurements. Free acetate ion exihibits ν_{as}(OAc) and ν_{s}(OAc) stretches at 1560 cm^{-1} and 1416 cm^{-1}, respectively[27]. On complexation, the former band undergoes a positive shift and the latter to a negative shift.

Table 2: IR, reflectance spectral data (cm^{-1}) and magnetic moments of the coordination compounds

Compd.	**ν(C=O) (thiazolidinone)**	**ν(C–O) Φ**	**ν_{as} (OAc)**	**ν_{s} (OAc)**	**ν(C–O) (MeOH)**	ν_{max}	Mag. Moment (B.M.)
2	1710	1528	–	–	–	–	–
3	1680	1537	1570	1345	–	17300	1.82
4 (M = Co)	1676	1535	1572	1352	966	8650, 17340, 19500	4.78
4 (M = Ni)	1680	1536	1564	1340	972	8670, 15860, 24500	3.32
5	1672	1532	–	–	970	–	Diamagnetic
6	1675	1534	–	–	–	–	Diamagnetic
7 (AA = ea)	1678	1537	–	–	–	–	Diamagnetic
7 (AA= oap)	1674	1537	–	–	–	–	Diamagnetic
7 (AA = en)	1684	1536	–	–	–	–	Diamagnetic
7 (AA= ophen)	1680	1534	–	–	–	–	Diamagnetic
7 (AA = bipy)	1677	1538	–	–	–	–	Diamagnetic
7 (AA = phen)	1679	1535	–	–	–	–	Diamagnetic

The appearance of two new bands at 1564-1572 cm^{-1} [ν_{as}(OAc)] and 1340-1352 cm^{-1} [ν_s(OAc)] stretches indicates the presence of the coordinated acetato group in **(3)** and **(4)**. The energy difference ($\Delta\nu$ = 220-225 cm^{-1}) between these stretches is > 210 cm^{-1} which indicates the monodentate nature of the acetato moiety. The presence of a broad band between 3345-3435 cm^{-1} due to the ν(O–H) (MeOH) stretch and the decrease of the ν(C–O) (MeOH) stretch[27] from 1034 cm^{-1} to lower energy by 62-68 cm^{-1}, respectively in **(4)** (M = Co, Ni) and **(5)** indicates the presence of coordinated MeOH molecule(s). The appearance of bands at 945 and 914 cm^{-1} due to the ν_s(O=Mo=O) and ν_{as}(O=Mo=O) stretches respectively, in **(5)** indicates the presence of a *cis*-MoO_2 structure[28]. The appearance of two new bands, one at 1578 cm^{-1} and other at 1395 cm^{-1} due to the ν_s(C–O) and ν_{as}(C–O) stretches, respectively in **(5)** suggests the presence of coordinated acetylacetonato moiety in a monobasic bidentate manner[29,30]. The absence of a new band between 850-950 cm^{-1}, due to the ν(Zr=O) stretch favours the formulation of **(6)** and **(7)** as $[Zr(OH)_3(LH_2)]$ and $[Zr(OH)_3(LH_2)(AA)]$ and not as $[ZrO(OH)(LH_2)(H_2O)]$ and $[ZrO(OH)(LH_2)(H_2O)(AA)]$, respectively. The presence of a broad band between 3400-3450 cm^{-1} and the appearance of a new band between 1135-1155 cm^{-1} due to the δ(Zr–OH) bending mode also support the proposed structures of the present compounds[31,32]. The δNH_2 vibrations of AA (AA = ea, oap, en, phen) occurring between 1600-1640 cm^{-1} shifts to lower energy by 20-35 cm^{-1} in **(7)** indicating coordination of their N atoms to the metal ion. The ν(C–O) (alcoholic/phenolic) stretches of ethanolamine and *o*-aminophenol occurring at 1230 and 1520 cm^{-1}, respectively, shift to 1205 cm^{-1} and 1550 cm^{-1} in respective heterochelate conforming the participation of alcoholic/phenolic O atom towards coordination[31,32]. The ν(C=N) (ring) stretch of 2,2'-dipyridyl and 1,10-phenanthroline occurring at 1585 and 1590 cm^{-1}, respectively undergoes higher shift (~25 cm^{-1}) in **(7)**, indicating the coordination of their ring N atoms to the respective metal ion[31,32].

Reflectance spectra

The reflectance spectrum of **(3)** exhibits an asymmetric broad band at 17300 cm^{-1} due to the $^2B_{1g} \rightarrow {}^2A_{1g}$, $^2B_{2g}$ and ^2E_g transitions suggesting a square-planar geometry[33]. The absence of a band in the range 8000-10000 cm^{-1} precludes the presence of a tetrahedral structure[34]. **(4)** (M = Co) exhibits three bands at 8650, 17340 and 19500 cm^{-1} due to the $^4T_{1g}(F) \rightarrow {}^4T_{2g}(F)(\nu_1)$, $^4T_{1g}(F) \rightarrow {}^4A_{2g}(F)(\nu_2)$, $^4T_{1g}(F) \rightarrow {}^4T_{1g}(P)(\nu_3)$ transitions, respectively in an octahedral symmetry[35]. The ν_3/ν_1 value in the present compound is 2.25 and it lies in the usual range (2.0-2.8) reported for majority of octahedral Co (II) compounds[35]. The spectral parameters are: Dq = 978.69 cm^{-1}, B = 799.1, β = 0.82, β^0 = 18.0% and CFSE = –93.7 kJ mol^{-1}. The reduction of Racah parameter from the free ion value (971 cm^{-1}) to 799.1 cm^{-1} and the % covalence value (18.0) in **(4)** (M = Co) are indicative of the presence of covalent nature in the present case[35]. The appearance of three bands at 8980, 16410 and 24500 cm^{-1} in **(4)** (M = Ni) is due to the $^3A_{2g}(F) \rightarrow {}^3T_{2g}(F)$ (ν_1), $^3A_{2g}(F) \rightarrow {}^3T_{1g}(P)$ (ν_2) and $^3A_{2g}(F) \rightarrow {}^3T_{1g}(F)$ (ν_3)

transitions, respectively in an octahedral symmetry[35]. The ν_2/ν_1 value in the present compound is 1.8, which occurs in the usual range (1.6-1.82) reported for the majority of octahedral Ni (II) compounds[36]. The spectral parameters are: Dq = 898 cm^{-1}, B = 811.74, β = 0.79 and β^0 = 21.0%, CFSE = –128.91 $kJmol^{-1}$. The reduction of Racah parameter from the free ion value (1030 cm^{-1}) to 811.74 cm^{-1} and the % covalence value (21.0) in **(4)** (M = Ni) are indicative of the presence of covalent nature in the present case[36]. The 10Dq value (9786 cm^{-1}) of Co (II) compound is greater than the corresponding Ni (II) compound, (8980 cm^{-1}). This is in line with the spectrochemical series of metal ions for a given ligand, given stoichiometry and a given stereochemistry[36]: Ni (II) < Co (II). The β^0 values of **(4)** (M = Ni) is comparable to the corresponding Co (II) compound : 21% ~ 18%. This is in line with the nephelauxatic metal ion series in terms of B and β^0 for a given ligand, given stoichiometry and a given stereochemistry[36]. The higher negative CFSE value in the Co (II) coordination compound in comparison to that of the corresponding Ni (II) coordination compound is as expected.

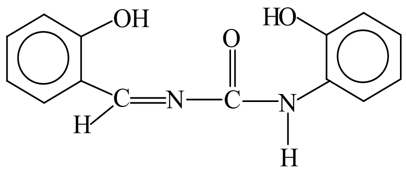
^{1}H NMR spectra

The ^{1}H NMR spectra of **(1)**, **(2)**, **(6)** and **(7)** (where AA = ea and oap) have been recorded in DMSO-d_6. The chemical shifts (δ) are expressed in ppm from TMS[37]. **(1)** exhibits signals at δ 6.82-8.00 ppm due to aromatic protons[32], δ 7.32 ppm due to azomethine proton[27], a broad signal at δ 10.91 ppm due to –OH protons[32] and a signal at δ 11.7 ppm due to –NH proton[32]. **(2)**, **(6)** and **(7)** exhibit signal at δ 3.83, δ 3.3, δ 3.04 ppm (AA = ea) and at δ 3.02 ppm, (AA = oap) ppm respectively due to the > CH_2 protons of thiazolidin-4-one moiety[38]. The presence of the signal at δ 5.89, δ 5.82, δ 5.78 ppm (AA = ea) and δ 5.73 ppm (AA = oap) ppm is due to the proton of –CHNS moiety[38]. The appearance of –NH proton in the spectra of **(2)**, **(6)** and **(7)** (where AA = ea and oap) at δ 11.54, δ 11.08, δ 11.2 ppm and 11.05 ppm, respectively suggests the presence of keto form of the thiazolidin-4-one in these compounds[32]. The signals at δ 10.72, δ 10.83, δ 10.81 ppm (AA = ea) and δ 10.78 (AA = oap) ppm in **(2)**, **(6)** and **(7)**, respectively are due to the presence of the phenolic –OH protons in **(2)**, **(6)** and **(7)** (when) [27]. The presence of multiplet in the range 6.48-7.45 ppm is due to the aromatic protons in **(2)**, **(6)** and **(7)**[32]. The appearance of the signals at δ 2.3 ppm and δ 1.8 ppm in **(7)** (AA = ea) are due to the –OH and –NH_2 protons, respectively, and it indicates the neutral bidentate ON donor nature of the ethanolamine in the heterochelate. **(7)** (AA = oap) exhibits signal at δ 7.3 ppm due to –NH_2 group[39] of coordinated *o*-aminophenol.

Magnetic susceptibility measurements

The Cu (II) ion belongs to the $S = \frac{1}{2}$ system and since its spin-orbit coupling constant is negative[40], the magnetically dilute Cu (II) complexes are expected to exhibit magnetic moments higher than the spin-only value (1.73 B.M.) due to the presence of orbital contribution[40]. The magnetic moment of **(3)** is 1.82 B.M., which lies in the usual range (1.75-2.20) reported for the magnetically dilute Cu (II) compounds[36]. The magnetic moment of **(4)** (M = Co) is 4.78 B.M., which is consistent with high-spin octahedral structure[36]. The magnetic moment of **(4)** (M = Ni) is 3.32 B.M., which suggests six-coordinate structure[36]. The coordination compounds of MoO_2 (VI) and Zr $(OH)_2$ (IV) ions are diamagnetic as expected.

Proposed structures of the compounds



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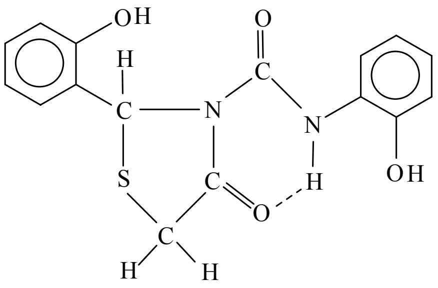

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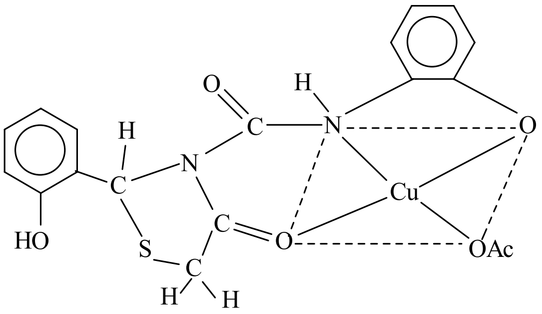

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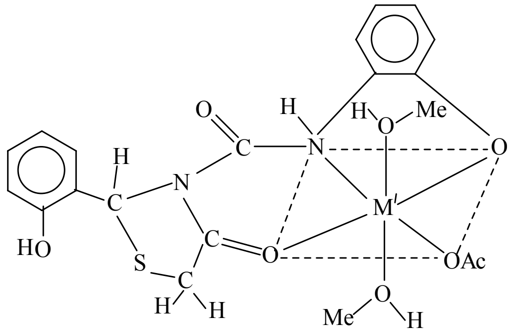

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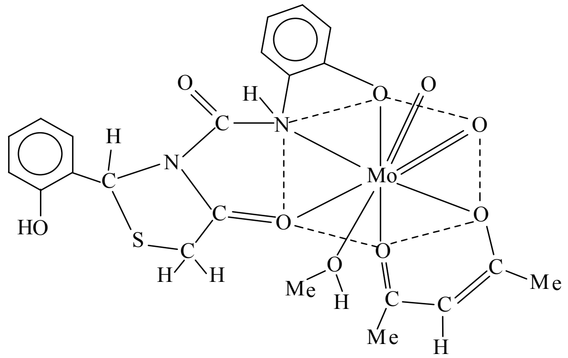

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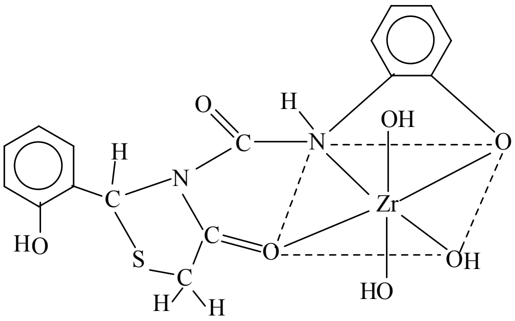

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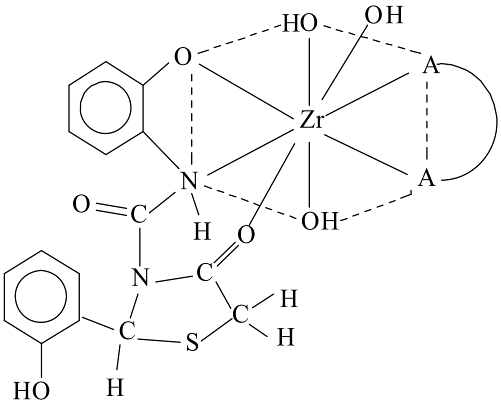

(7)

CONCLUSION

On the basis of analytical data, conductance measurements, molecular weight, spectral, magnetic susceptibility measurements, We propose a square-planar structure for **(3)**, an octahedral structure for **(4)** (M = Co, Ni) and **(6)**, and an eight-coordinate structure for **(5)** and **(7)**.

ACKNOWLEDGEMENT

The corresponding author is highly indebted to Director of his institute for granting Cumulative Professional Development Allowances for the preparation of this manuscript.

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Accepted : 10.05.2011