



Research Paper

Synthesis, Characterization and Antibacterial Activity of Novel Schiff Base Derived From 4-Acetyl-3-Methyl-1-(4'-Methyl-Phenyl)-2-Pyrazolin-5-One and its Transition Metal Complexes

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Abstract- The novel Schiff base has been synthesized from 4-acetyl-3-methyl-1-(4'-methylphenyl)-2-pyrazolin-5-one with 2-amino-4-phenyl-5-methyl thiazole and was characterized by elemental analysis, mass spectra, ¹H NMR spectra, ¹³C NMR spectra and FT IR spectra. This Schiff base forms complexes of type ML₂.2H₂O (Where M= Mn, Fe, Co, Ni and Cu) and this complexes were characterized by elemental analysis, Magnetic susceptibility, electrical conductance, electronic spectra, infrared spectra and TGA analysis. All the compounds were tested for their antimicrobial activity. The result indicates that the growth of the tested organism was inhibited by most of the compounds.

Keywords: Pyrazolin-5-one, 2-Amino thiazole, Schiff base, Transition metal complexes, Spectroscopy, Antibacterial activity.

Introduction

The chemistry of pyrazolone derivatives has attracted much attention because of their interesting structural properties and applications in diverse areas [1-4]. They are useful reagents for the extraction and separation of various metal ions [5-6]. They can also be used in laser materials, in chromatographic study, in the petrochemical industry [7-8] and as ¹H NMR shift reagents. Many of these ligands exhibited keto-enol tautomerism and because of this they show interesting structural and spectroscopic properties. [9] β-diketones have played and continued to play a key role in coordination compounds that have found wide application in several fields, from new materials to catalysts [10], as precursors for CVD in the microelectronic industry and as potential antitumourals [11]. Even the simplest pyrazolone-5 derivatives for instance, antipyrine and amidopyrine are well-known analgesics and widely used as medicine. Pyrazolone derivatives are also used as starting materials for the synthesis of biologically active compounds and for the construction of condensed heterocyclic systems [12].

Among these ligands, acyl pyrazolones have been studied extensively owing to their effective properties with respect to extracting metal ions [3]. On the other hand pyrazolone-based Schiff base chemistry is less extensive. In continuation of our work [13-20] on the metal complexes of Schiff base ligands we reported here the study of Mn(II), Fe(II), Co(II), Ni(II) and Cu(II) metal complexes of Schiff base derived from 4-acetyl-3-methyl-1-(4'-methylphenyl)-2-pyrazolin-5-one[AMP] with 2-amino-4-phenyl-5-methyl thiazole [APMT]. Synthesis characterization and antimicrobial activity of above metal complexes are reported here.

Material and Methods

All the chemicals used in the present study were of A.R. grade. Propiophenone, Iodine, hydrochloric acid, sulfuric acid, thiourea, ethylacetoacetate, sodium acetate, calcium hydroxide, acetyl chloride, 1:4 dioxane and methanol (SD's fine chemical Ltd., and Merck chemicals, Mumbai) were used without further purification. The Mn(II), Co(II), Ni(II), Cu(II) metal acetate and Fe(II) sulphate (SD's

fine chemical Ltd, Qualigens-Glaxo, Mumbai and Merck chemicals., Mumbai) were used for the preparation of Schiff base metal complexes.

Melting points were taken in one side open capillaries on a Melting point apparatus having model number VMP-D of a make VEEGO. Electronic Spectra were recorded in DMF solution on LAMBDA 19, UV/VIS/NIR ("SICART-CVN" at Vallabh Vidyanagar, Gujarat, India). The Mass spectrum of ligand was recorded on the instrument named LCMS-2010A of make Shimadzu. Carbon, Hydrogen and Nitrogen were estimated on a Thermo fisher (Thermo electron corporation Limited), Flash Elemental Analyzer-1112. The ^1H NMR and ^{13}C NMR spectra of ligand [in Deuterated Chloroform(CDCl_3)] were recorded on a AVANCE-II 400 of make BRUKER spectro-photometer using TMS [$(\text{CH}_3)_4\text{Si}$] as internal standard. The Infrared spectra of the ligand studied in the present work were recorded on the model FT-IR-8300 of Shimadzu in KBr (Zydu Research Center, Ahmedabad, India).

The Schiff base ligand was prepared by condensation of equimolar amount of AMP^[21] and APMT^[22] in minimum quantity of methanol. The reaction mixture was refluxed in rotamental for about three hours. On cooling the yellow solid compound obtained was filtered, washed with methanol and dried in air. For the preparation of complexes, an aqueous solution of metal acetate/metal sulphate (0.05 M) and 1:4 dioxane solution of ligand (0.05M) were mixed in presence of acetate buffer ($\text{p}^{\text{H}}=6.5$) and the mixture was digested on sand bath for 30 minutes, cooled and filtered the precipitates and then washed with water and then with 1:4 dioxane to remove excess metal ions and unreacted Schiff bases respectively.

Results and discussion

Schiff base ligand was characterized by elemental analysis, mass spectra, ^1H NMR, ^{13}C NMR and FT IR spectra. The analytical data of ligand and metal complexes, together with physical properties are given in Table 1. The analytical data of the complexes correspond to the general stoichiometry $[\text{ML}_2 \cdot 2\text{H}_2\text{O}]$ where M= Mn(II), Fe(II), Co(II), Ni(II) and Cu(II), Ligand L=AMPAPMT. The values of molar conductance (λ_{M}) of the complexes in DMSO indicate that the $[\text{ML}_2 \cdot 2\text{H}_2\text{O}]$ are non-electrolytes. Magnetic moments lie in the range 5.50-5.76 B.M., 4.80-5.02 B.M., 4.36-4.48 B.M., 2.75-2.95 B.M., and 1.90-1.99 B.M., for Mn(II), Fe(II), Co(II), Ni(II) and Cu(II) complexes respectively.

Mass Spectral data

The Positive ion mass spectral analysis of AMPAPMT observed at m/z 403.19 (M^+), Confirms the theoretical molecular weight i.e. 402.51.

^1H NMR Spectra (CDCl_3)

(400 MHz, CDCl_3) $\delta=13.90(\text{s}, 1\text{H}, \text{OH})$, $7.34(\text{s}, 1\text{H}, \text{Thiazole ring})$, $7.20-7.84(\text{m}, 9\text{H}, 2\text{Ar})$, $2.88(\text{s}, 3\text{H},$

$\text{CH}_3)$, $2.55(\text{s}, 3\text{H}, \text{CH}_3)$, $2.45(\text{s}, 3\text{H}, \text{CH}_3)$, $2.36(\text{s}, 3\text{H}, \text{CH}_3)$.

^{13}C NMR Spectra (CDCl_3)

(400MHz, CDCl_3) $\delta=12.72(\text{C}_4)$, $17.90(\text{C}_{16})$, $18.32(\text{C}_{12})$, $21.11(\text{C}_{23})$, $103.21(\text{C}_{13})$, $119.52-129.46$ (Tertiary Aromatic Carbon), $134.54(\text{C}_{20})$, $134.56(\text{C}_5)$, $136.27(\text{C}_{17})$, $137.47(\text{C}_1)$, $147.65(\text{C}_{14})$, $148.46(\text{C}_2)$, $159.31(\text{C}_{15})$

FT-IR Spectra

Interpretation: From the recorded IR spectrum, the wave numbers of corresponding groups are shown below.

2987.84cm^{-1} ($\nu_{\text{C-H}}$ stretching of Aromatic), 2920cm^{-1} ($\nu_{\text{O-H}}$ stretching of saturated hydrocarbon), 1626.05cm^{-1} , ($\nu_{\text{C=N}}$ stretching of azomethine), 1608.69cm^{-1} , $1485.24, 1128\text{cm}^{-1}$ (characteristic bands of pyrazolin ring), 1510.31cm^{-1} (Characteristic band of thiazole ring).

The infrared spectra of the ligand show $\nu_{\text{O-H}}$ (weakly H-bonded) band at 2920cm^{-1} ^[23]. The absence of this band in all the metal complexes indicates the removal of proton of hydroxyl group of pyrazolin ring during the chelation. This is further supported by the shift of C-O frequency from 1320cm^{-1} (in ligand-L) to $1371-1321\text{cm}^{-1}$ (in complexes). The sharp intense band at 1626.05cm^{-1} in the ligand can be assigned to $\nu_{\text{C=N}}$ (azomethine). A downward shift ($\Delta\nu=06-31\text{cm}^{-1}$) in $\nu_{\text{C=N}}$ (azomethine) is observed upon coordination indicating that the nitrogen of azomethine group is involved in coordination. All the complexes show broad band in the region 3200cm^{-1} to 3450cm^{-1} which may be assigned to $\nu_{\text{O-H}}$ of coordinated water^[24]. To account for the octahedral stereochemistry of the metal complexes, the coordination of two water molecules is expected.

The bands present at $\sim 507\text{cm}^{-1}$ in Mn(II) complex, $\sim 501\text{cm}^{-1}$ in Fe(II) complex, $\sim 516\text{cm}^{-1}$ in Co(II) complex, $\sim 563\text{cm}^{-1}$ in Ni(II) complex and $\sim 511\text{cm}^{-1}$ in Cu(II) complex may be due to metal-nitrogen stretching vibration^[25,26]. A less intense band at $\sim 1620\text{cm}^{-1}$ in the spectra of ligand may be assigned to $\nu_{\text{C=N}}$ (ring)^[27]. All the metal complexes do not show shifting in $\nu_{\text{C=N}}$ compared to its respective ligand. This suggests that the nitrogen atom of the thiazole ring has not participated in the coordination. However, in water containing metal complexes, this band is observed as a broad band with some fine structures this may be due to coupling of the bending mode of coordinated water molecules with $\nu_{\text{C=N}}$ ^[28].

Electronic spectra

The ligand shows two absorption bands at $38,314\text{cm}^{-1}$ and $27,397\text{cm}^{-1}$. No absorption was observed in the visible region for the ligand. In the absence of Quantum mechanical calculation, it is not possible to assign the absorption bands to definite electronic transitions with complete certainty.

However, it appears reasonable to assign the bands to $\pi \rightarrow \pi^*$ transitions^[29]. The electronic spectra of Mn(II) complex exhibited three very low intense bands, one at 14850cm^{-1} , which may rise due to ${}^6A_{1g} \rightarrow {}^4T_{1g}$ (G) transition, another band at 17670cm^{-1} assigned to ${}^6A_{1g} \rightarrow {}^4A_{1g}$ (G) transition and the third band at 25492cm^{-1} may be assigned to ${}^6A_{1g} \rightarrow {}^4A_{1g}, {}^4E_g$ (G) transition for Mn(II) ion in octahedral environment. The μ_{eff} (Table-1) value of the complex suggests the spin $3d^5$ configuration^[30]. The electronic spectra of Fe(II) complex show broad band at 28553cm^{-1} and 12490cm^{-1} which may be assigned to the ${}^5T_{2g} \rightarrow {}^5E_g$ transition. The magnetic moment value 4.81 BM indicates that the complex is spin-free and it has octahedral geometry^[31]. The electronic spectra of Co(II) complex exhibit absorption bands in the region 8350cm^{-1} to 9650cm^{-1} corresponding to ν_1 and ν_3 transitions ${}^4T_{1g}(F) \rightarrow {}^4T_{2g}(F)(\nu_1)$; ${}^4T_{1g}(F) \rightarrow {}^4T_{1g}(P)(\nu_3)$. In the present investigation, Co(II) complex exhibited the absorption bands at 8590cm^{-1} and 18924cm^{-1} , corresponding to ν_1 and ν_3 transitions respectively. These bands were the characteristics of high spin octahedral Co (II) complexes. However, ν_2 band is not observed because of its proximity to strong ν_3 transition. The magnetic measurement of Co (II) complex display magnetic moment value of 4.39 B.M. which is in the octahedral range 4.40 to 4.53 B.M. The Ni(II) complex exhibited three bands at 8540cm^{-1} , 18739cm^{-1} and 26312cm^{-1} which was attributed to the ${}^3A_{2g} \rightarrow {}^3T_{2g}(F)$ (ν_1); ${}^3A_{2g} \rightarrow {}^3T_{1g}(F)$ (ν_2) and ${}^3A_{2g} \rightarrow {}^3T_{1g}(P)$ (ν_3) transitions respectively indicating octahedral geometry around Ni(II) ion. Ni(II) complex exhibit the magnetic moment value of 2.79 B.M which is in the range of 2.75 to 3.02 B.M suggesting consistency with their octahedral environment^[32]. For the Cu(II) complex with D_{4h} symmetry, three spin allowed transitions ${}^2B_{1g} \rightarrow {}^2A_{1g}$ (ν_1), ${}^2B_{1g} \rightarrow {}^2B_{2g}$ (ν_2) and ${}^2B_{1g} \rightarrow {}^2E_g$ (ν_3) are possible but the electronic spectra of Cu(II) complex display two bands at 16987cm^{-1} and 23952cm^{-1} . There should be third transition but we could not observe the same which may be due to very close energy values of different states. Absence of any spectral band below 10000cm^{-1} rules out the possibility for tetrahedral structure of the present complex and also suggests distorted octahedral geometry of the complex^[33]. The low molar conductance values in DMF solution for all metal complexes (Table-1) indicate that the complexes are nonelectrolytes.

TGA analysis

Thermo gravimetric analysis of Schiff base ligand and its metal complexes are used to:

- (i) Get information about the thermal stability of these new complexes,
- (ii) Decide whether the water molecules (if present) are inside or outside the inner coordination sphere of the central metal ion and
- (iii) Suggest the general scheme for thermal decomposition of metal complexes.

In the present investigation, heating rates were suitably controlled at $10^\circ\text{C min}^{-1}$ under nitrogen

atmosphere and the weight loss was measured from the ambient temperature up to $\sim 1000^\circ\text{C}$ ^[34-37]. The data are provided in the Table: 2.

Number of coordinated or lattice water molecule/molecules present in the complexes were calculated from the percentage weight loss of the complexes from the thermograms.

Generally, the loss of lattice water will be present at a lower temperature than that of coordinated water^[38-39]. From the nature of the thermograms and percentage weight-loss, the such type of complexes studied in the present work can be classified in three groups^[40-41]. The thermogram of this group of metal complexes show three stage decomposition.

- (1) The first stage decomposition is obtained in the temperature range $140-210^\circ\text{C}$. The % weight loss in this range corresponds to the loss of two coordinated water molecules.^[42-44]
- (2) The second stage decomposition is obtained in the temperature range $210-400^\circ\text{C}$. The % weight loss in this range corresponds to % weight loss of two Schiff base ligands.
- (3) The third stage decomposition range is obtained in the temperature range $400-900^\circ\text{C}$. The % weight loss in this range corresponds to % weight loss of metal oxide residue. All the metal complexes do not show weight loss below 120°C indicates the absence of lattice water in the metal complexes. On the basis of TGA and analytical data of all Mn(II), Fe(II), Co(II), Ni(II) and Cu(II) complexes studied in the present work correspond to type $[\text{ML}_2.2\text{H}_2\text{O}]$.

Antibacterial activity

The antibacterial activity was estimated against E.coli, B. subtilis and S. aureus and evaluated by using of agar disc diffusion method on the basis of the size of inhibition zone formed around the paper discs. For each concentration, the mean diameter (mm) of inhibition zone developed was calculated. The test compounds in measured quantities were dissolved in DMF to get concentrations of 200 and 100 ppm of compounds. Twenty five milliliter nutrient agar media was poured in each Petri plates. After solidification, 0.1mL of test bacteria spread over the medium using a spreader. The discs of Whatmann no. 1 filter paper having the diameter 5.00 mm were placed at four equidistant places at a distance of 2cm from the center in the inoculated Petri plates. Filter paper disc treated with DMF served as control and Amoxicillin used as a standard drug. These Petri plates were kept in refrigerator for 24 hours for pre diffusion. Finally, Petri plates were incubated for 24 hours 30°C . The zone of inhibition was calculated in millimeters carefully.

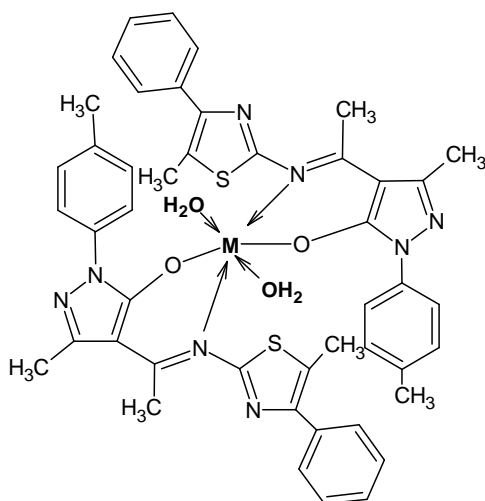
The Schiff base ligand was found to be biologically active (Table: 3). It is known that chelation tends to make ligands act as more powerful and potent bactericidal agent^[45]. The values indicate that the metal complexes had a higher antibacterial

activity than the free ligand. Such increased activity of the metal complexes can be explained on the basis of the overtone concept^[46] and chelation theory^[47]. According to the overtone concept of cell permeability, the lipid membrane that surrounds the cell favors the passage of only lipid soluble materials, due to which liposolubility is an important factor controlling the antimicrobial activity. On chelation, the polarity of the metal ion is reduced to a great extent due to the overlap of the ligand orbital and the partial sharing of the positive charge of the metal ion with donor groups. Furthermore, it increases the delocalization of electrons over the whole chelate ring and enhances the lipophilicity of the complex. This increased lipophilicity enhances the penetration of the complex into the lipid membrane and blocks the metal binding sites on the enzymes of the microorganism.

Conclusion

On the basis of these results obtained from elemental analysis, infrared spectra, electronic spectra, TGA analysis and magnetic susceptibility measurements the following structures are proposed for the Schiff base metal complexes.

Structure of Complexes of Schiff base ligand AMPAPMT



Where M= Mn(II), Fe(II), Co(II), Ni(II) and Cu(II).

Antibacterial activity leads to the following conclusions:

1. The metal complexes show more activity than the ligand against tested bacteria.
2. Antibacterial activity of Ni(II) complex has higher activity than the other complexes.

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Table.1: Analytical data and physical property of Ligand and their metal complexes

Compound	MW	Color	MP (°C)	λ_M ($\Omega^{-1}cm^{-1}mol^{-1}$)	Yield (%)	Elemental analysis (%) Found (Calculated)					
						C	H	N	S	M	μ_{eff} B.M.
AMPAPMT	374.46	Brown	222	-	75	67.19 (68.63)	5.34 (5.51)	13.37 (13.92)	7.72 (7.92)	-	-
Mn (AMPAPMT) ₂ .2H ₂ O	893.98	Brown	-	11.23	65	61.71 (60.80)	5.01 (5.19)	12.47 (12.53)	7.11 (7.17)	6.21 (6.15)	5.51
Fe (AMPAPMT) ₂ .2H ₂ O	838.78	Brown	-	11.12	59	61.87 (61.74)	5.16 (5.18)	12.76 (12.52)	7.09 (7.17)	6.34 (6.24)	4.81
Co (AMPAPMT) ₂ .2H ₂ O	841.86	Dark Brown	-	12.73	60	61.37 (61.53)	5.18 (5.16)	12.51 (12.48)	7.24 (7.14)	6.55 (6.56)	4.39
Ni (AMPAPMT) ₂ .2H ₂ O	841.62	Green	-	10.74	61	61.57 (61.54)	5.18 (5.16)	12.51 (12.48)	6.55 (6.54)	7.12 (6.97)	2.79
Cu (AMPAPMT) ₂ .2H ₂ O	846.48	Brown	-	15.16	61	61.22 (61.21)	5.05 (5.14)	12.53 (12.41)	7.19 (7.04)	7.19 (7.04)	1.97

Table.2. Thermo Analytical Results of Metal Complexes of Ampapmt

Compounds	Stage-I [140-210°C]	Stage-II [210-400°C]	Stage-III [400-900°C]
	Mass Lose Obs. (Calc.)	Mass Lose Obs. (Calc.)	Mass Lose Obs. (Calc.)
[Mn(AMPAPMT) ₂ (H ₂ O) ₂]	3.98 (4.03)	87.31 (87.36)	8.89 (8.83)
[Fe(AMPAPMT) ₂ (H ₂ O) ₂]	3.81 (4.02)	87.22 (87.28)	8.97 (8.92)
[Co(AMPAPMT) ₂ (H ₂ O) ₂]	3.87 (4.01)	87.79 (87.87)	8.39 (8.34)
[Ni(AMPAPMT) ₂ (H ₂ O) ₂]	3.96 (4.01)	90.01 (90.07)	6.55 (6.53)
[Cu(AMPAPMT) ₂ (H ₂ O) ₂]	4.01 (3.99)	87.46 (87.42)	8.23 (8.81)
Assignment	Loss of two coordinated water molecules	Loss of two Schiff base ligand molecules	Metal Oxide/Metal

Table: 3

compound	Antibacterial activity of ligand and its metal complexes Zone of inhibition in mm(concentration in ppm)					
	E. coli		B. subtilis		S. aureus	
	100	200	100	200	100	200
AMPAPMT	8	15	6	11	7	14
[Mn(AMPAPMT) ₂ (H ₂ O) ₂]	10	17	8	15	10	16
Fe(AMPAPMT) ₂ (H ₂ O) ₂]	10	19	9	16	11	18
Co(AMPAPMT) ₂ (H ₂ O) ₂]	9	15	10	14	9	15
Ni(AMPAPMT) ₂ (H ₂ O) ₂]	12	20	13	16	13	21
Cu(AMPAPMT) ₂ (H ₂ O) ₂]	10	16	9	15	10	17
Amoxicillin	17	28	16	22	18	29

Route of synthesis

