



# Synthesis, Characterization and Antimicrobial activity of some novel schiff Base 3d Transition Metal Complexes Derived from Dihydropyrimidinone and 4- Aminoantipyrine

Mohanambal D.<sup>1</sup> and Arul Antony S.<sup>2</sup>

<sup>1</sup>Sriram Engineering College, Department of chemistry, Perumalpattu, Chennai-602024, Tamil Nadu, INDIA

<sup>2</sup>PG and Research Department of Chemistry, Presidency College, Chennai-600 005, Tamil Nadu, INDIA

Available online at: [www.isca.in](http://www.isca.in), [www.isca.me](http://www.isca.me)

Received 14<sup>th</sup> May 2014, revised 28<sup>th</sup> May 2014, accepted 10<sup>th</sup> July 2014

## Abstract

Novel Schiff base and its 3d transition metal complexes of Mn(II), Fe(III) and VO(IV) has been designed and synthesized from 4-aminoantipyrine and Ethyl 4- methyl -oxo-6-phenylhexahydro pyrimidine-5-carboxylate . The ligand and complexes were characterized by physico chemical studies like solubility, melting point, elemental analysis, magnetic susceptibility, conductivity, and spectral studies like IR, UV/VIS spectra. From the data it has been observed that the complexes has the composition of ML<sub>2</sub> type and paramagnetic nature, the complexes were coordinated through azomethine nitrogen and pyrazole oxygen of the ligand which was further supported by the appearance of new bands in IR spectra due to  $\nu(M-N)$ ,  $\nu(M-O)$  and  $\nu(M-Cl)$ . The ligand and its complexes had been screened for their antifungal and antibacterial activities against the fungi (*Aspergillus niger*, *Candida*), and the bacteria (*E.coli*, *Salmonella typhi*, *Bacillus subtilis*, *Staphylococcus aureus*.) and showed good antimicrobial activities against the tested bacteria and fungi at 1000, 750 and 500 mg/ml. Therefore possible use of the complexes as antibiotic can be suggested.

**Keywords:** 4-aminoantipyrine, antimicrobial, DHPMs, Schiff base, transition metal complex.

## Introduction

The co-ordination chemistry of organic transition metal complexes of Schiff base ligand with donor groups like nitrogen, oxygen gained importance for more than two decades because of their uses as models of biological systems<sup>1-3</sup>, including antibacterial<sup>4</sup>, antifungal<sup>5</sup>, antitumour<sup>6</sup>, and anti-inflammatory activities<sup>7</sup>. Now a day's chemist are very much focused on the Schiff base derived from heterocyclic ring with carbonyl compounds as it important special centre of attraction in many areas like biological clinical, medicinal, analytical and pharmacological field etc<sup>8-10</sup>. Among them 4-aminoantipyrine based heterocyclic had a great importance as it is abundant in nature and have wide pharmacological activities<sup>11</sup>. The coordinating property of Schiff base with 4-aminoantipyrine and Dihydropyrimidinones (DHPMs) have attracted considerably because of their pharmaceutical and therapeutic properties. The synthesis of dihydropyrimidinones was first reported by Biginelli in 1893; consist of one-pot condensation of an aldehyde,  $\beta$ -ketoester and urea under strongly acidic conditions. Thorough literature survey reveals that more attention has been given to Schiff's base derived from 4-aminoantipyrine with several aldehydes. But less work had been reported on the condensation process of 4-aminoantipyrine comprising with Ethyl 4-methyl-2-oxo-6-phenylhexahydropyrimidine-5-carboxylate. Hence, in the present content a new bidentate Schiff base derived from Ethyl 4-methyl-2-oxo-6-phenylhexahydropyrimidine-5-carboxylate (DHPMs) and 4-aminoantipyrine had been synthesized and characterized by elemental analysis, melting

point, and molar conductance and further confirmed by Mass, Uv-Vis, FTIR, <sup>1</sup>H-NMR, <sup>13</sup>C-NMR, and ESR spectroscopic studies.

## Material and Methods

All the chemicals and solvents were purchased from Spectro chem. Pvt, Ltd, Mumbai. The completion of the reactions were monitored by TLC performed on silica gel plates. Melting points were measured in conventional method (open capillary tubes). The elemental analyses of the complexes were carried out by Perkin Elmer 2400. Molar conductance measurements were performed on ELICO-CM180 by using 10<sup>-3</sup> mol/L solutions of DMSO as the solvent at room temperature. Magnetic susceptibility was determined by Lake Shore Model 7410 VSM using standard Ni as calibrate. The IR spectra were recorded JASCO FT/IR-4100 spectrometer in the range of 4000-400 cm<sup>-1</sup> using KBr pellets. The electronic spectra were reordered by JASCO UV Vis / V-650 spectrometer in the range of 200-800nm. An NMR spectrum was recorded in CDCl<sub>3</sub> using Bruker 300MHz spectrometer. The ESI mass spectra of the ligand and its complexes were recorded using Mass QTOF micro mass spectrometer using nitrogen environment (CE-8e<sup>-</sup> V, CV -278e<sup>-</sup> V). The antimicrobial activities were carried out determined the minimum inhibition concentration (MIC).

**Synthesis of Ethyl 4-methyl-2-oxo-6-phenylhexahydropyrimidine-5-carboxylate (DHPMs):** The Ethyl 4-methyl-2-oxo-6-phenylhexahydropyrimidine-5-carboxylate has

been synthesized according to the reported procedure<sup>12</sup>. The product (yield 62%, literature yield 78.5%)<sup>13</sup> was obtained as a white solid, typical mp 198°C (lit. mp 202 to 204°C)<sup>14</sup>. The spectral data matched with reported data<sup>15</sup>.

**Preparation of Schiff base:** A mixture of 4-aminoantipyrine (2.03 g, 0.01 mol, and 20 ml) and Ethyl 4-methyl-2-oxo-6-phenylhexahydropyrimidine-5-carboxylate (2.6 g, 0.01 mol 20 ml) were refluxed for 4 hours at 60°C. Then the precipitated washed, filtered and re-crystallized by ethanol, and dried in desiccator over anhydrous calcium chloride vacuum. The general structures of ligand obtained from chemical analysis and spectral methods, were shown in Scheme (1).

**Synthesis of complexes: Manganese (II) complex:** The mixture of ligand (0.02 mol) and Mn(II) chloride (0.01 mol) were dissolved in 50ml ethanol and was refluxed for 5 h. Then, the dark brown solution obtained was evaporated to half of its original volume and allowed to crystallize. The complex that precipitate was filtered washed with ethanol and dried in a desiccator over calcium chloride vacuum.

**Iron (III) complex:** About 30ml of hot ethanolic solution of the ligand (0.02 mol) was added with 20 ml of the ferric chloride solution (0.01 mol). Then the solution was refluxed on a water-bath for 5h. It was then evaporated to half of its initial volume and allowed to crystallize. The dark brown complex separated was filtered, washed with ethanol and dried in vacuum.

**Vanadyl (IV) complex:** A solution of (0.01 mol) VOSO<sub>4</sub> and Schiff base ligand (0.02 mol) were mixed together, refluxed for 4 hrs at 50°C. On cooling colored solid metal complex was precipitated, washed with ethanol and dried. Recrystallization from hot ethanol gave the pure product.

**Antimicrobial activity assay:** *In vitro* antibacterial (*E.coli.*, *Staphylococcus aureus*, *Bacillus spp.*, *Salmonella spp.*) and *in vitro* antifungal (*Candida*, *Aspergillus niger*), tested by Mueller Hinton Agar solid media (MHA) and Sabouraud's dextrose

(SDA) media.<sup>16-18</sup> Agar diffusion assay was carried out to evaluate the antimicrobial activity of ligand and its metal complexes synthesized. The plates were incubated at 37°C for 24 hr, during which activity was evidenced by the presence of a zone of inhibition surrounding the well and antibacterial and antifungal activity was expressed as the mean diameter of inhibition zones (mm) produced by the synthesized compounds when compared to controls.

## Results and Discussion

The physical properties as well as data of micro analysis of the ligand and the complexes are summarized in table 1. All metal complexes are colored, stable in air at room temperature. These are insoluble in polar solvent and soluble in organic solvents such as DMF, DMSO. The molar conductance data of the complexes shows that the complexes are 1:2 non electrolytes in DMSO<sup>19</sup>.

**IR spectra:** The coordination character of the ligand to the metal ion has been determined by IR spectra and presented in table 2 and figure 1. The IR spectra showed a broad band around 3425 cm<sup>-1</sup> indicating the presence of stretching vibration of NH group present in the free ligand. The frequency of the band was not change to much as the NH group not involved in the chelation<sup>20</sup>. The stretching frequency of carbonyl group of ligand  $\nu(\text{C}=\text{O})$  at 1702 cm<sup>-1</sup> is shifted to lower frequency in the range 1687-1668 cm<sup>-1</sup> in complexes. Similarly the frequency corresponding to  $\nu(\text{C}=\text{N})$  at 1647 cm<sup>-1</sup> shifted to lower frequency in the range of 1610-1588 cm<sup>-1</sup> in complexes. The shifting in wave number and their intensities of referenced bands led to predict the chelation behavior. In addition, the spectra of metal complexes show some new bands in between 608-642 cm<sup>-1</sup>, 544-583 cm<sup>-1</sup> and 386-326 cm<sup>-1</sup> which are probably due to the formation of  $\nu(\text{M}-\text{O})$ ,  $\nu(\text{M}-\text{N})$  and  $\nu(\text{M}-\text{Cl})$  bond respectively<sup>21</sup>. In addition to other bands, the Vanadyl complex showed an additional band at 982 cm<sup>-1</sup> attributed to  $\nu(\text{V}=\text{O})$  frequency<sup>22</sup>.

Table-1  
Micro analysis and Physical properties of the ligand and its complexes

| Compound  | Molecular weight | Color        | % yield | M.Pt (°C) | Found (Calc). (%) |                |                  |                | Molar conductance<br>Ohm <sup>-1</sup> cm <sup>2</sup><br>mol <sup>-1</sup> x 10 <sup>-3</sup> |
|---|------------------|--------------|---------|-----------|-------------------|----------------|------------------|----------------|--|
|   |                  |              |         |           | C                 | H              | N                | M              |  |
| L<br>C <sub>25</sub> H <sub>27</sub> N <sub>5</sub> O <sub>3</sub>  | 445              | Yellow       | 62.9    | 233       | 66.89<br>(67.40)  | 6.08<br>(6.11) | 15.89<br>(15.72) | -              | -  |
| [MnL <sub>2</sub> ]Cl <sub>2</sub><br>[Mn(C <sub>25</sub> H <sub>27</sub> N <sub>5</sub> O <sub>3</sub> ) <sub>2</sub> Cl <sub>2</sub> ]      | 1016             | Light yellow | 70.2    | 252       | 59.06             | 5.35           | 13.77            | 5.40           | 3  |
| [FeL <sub>2</sub> Cl <sub>2</sub> ]Cl<br>[Fe(C <sub>25</sub> H <sub>27</sub> N <sub>5</sub> O <sub>3</sub> ) <sub>2</sub> Cl <sub>2</sub> ]Cl | 1053             | Blood red    | 62.8    | 256       | 57.35<br>(57.02)  | 5.24<br>(5.17) | 13.27<br>(13.30) | 5.07<br>(5.77) | 1.4  |
| [VOL <sub>2</sub> ]SO <sub>4</sub><br>[VO(C <sub>25</sub> H <sub>27</sub> N <sub>5</sub> O <sub>3</sub> ) <sub>2</sub> ]SO <sub>4</sub>       | 1054             | Yellow       | 72.4    | 268       | 56.95<br>(56.98)  | 5.23<br>(5.16) | 13.17<br>(13.29) | 4.95<br>(4.83) | 1.6  |

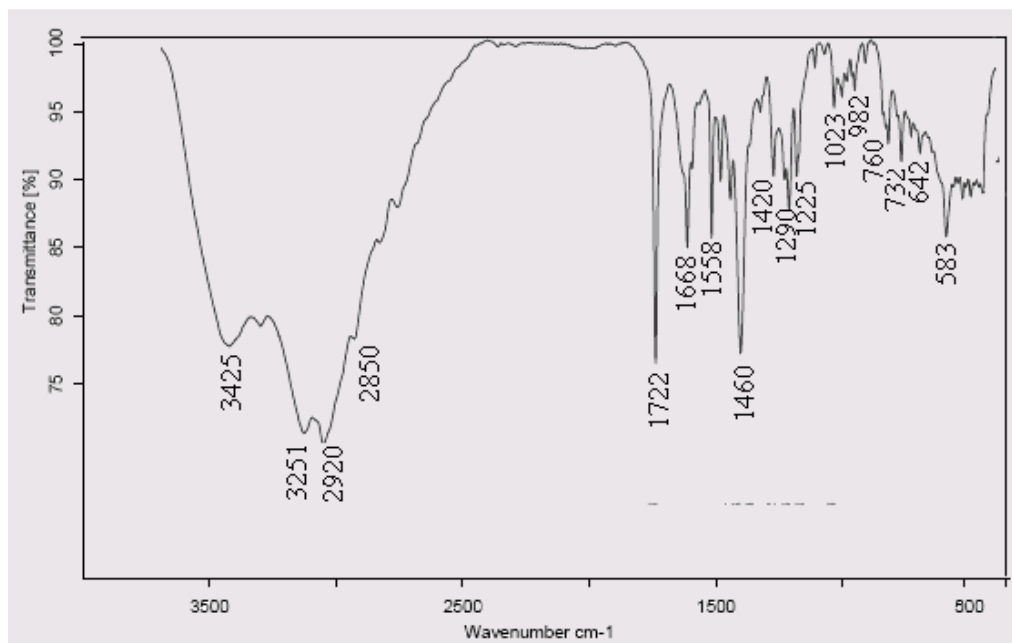


Figure-1  
FTIR spectrum of VO (IV) complexes

Table-2  
FTIR spectral data of ligand and complexes (in  $\text{cm}^{-1}$ )

| Compound                               | $\nu\text{C=O}$ | $\nu\text{C=N}$ | $\nu\text{M-O}$ | $\nu\text{M-N}$ | $\nu\text{M-Cl}$ | $\nu\text{V=O}$ | Phenyl Ring |
|--|-----------------|-----------------|-----------------|-----------------|------------------|-----------------|-------------|
| L                                      | 1702            | 1647            | -               | -               | -                | -               | 1026 & 766  |
| [MnL <sub>2</sub> Cl <sub>2</sub> ]    | 1687            | 1610            | 608             | 544             | -                | -               | 1025 & 764  |
| [FeL <sub>2</sub> Cl <sub>2</sub> ].Cl | 1669            | 1590            | 623             | 574             | 326              | -               | 1025 & 766  |
| [VOL <sub>2</sub> ].SO <sub>4</sub>    | 1668            | 1588            | 642             | 583             | -                | 982             | 1023 & 732  |

**Magnetic moment:** The  $\mu_{\text{eff}}$  for Mn (II) complex is found to be 3.6 B.M. This is quite close to partial spin pairing state ( $S=3/2$ ). The experimental value more or less approaches the expected value ( $\mu_{\text{eff}}=3.87$ )<sup>23</sup>. Hence, the value of  $\mu_{\text{eff}}$  noted for the Mn(II) complex is suggestive of the fact that the complex has a square-planar environment with ground term  $^4A_{1g}$  ( $b_{2g}^2 e_g^2 a_{1g}^1$ ). The magnetic moment values are found to be 1.73, 1.75 BM for Fe (III), and VO (IV), complexes, respectively, suggesting an octahedral geometry has been assigned to the complexes, except VO (IV) complex which has square-pyramidal geometry. These values are comparable with other reported complexes<sup>24-25</sup>

**Electronic spectra:** The spectrum of Mn(II) complex which is shown in figure 2, displays a broad shoulder like absorption band near 560nm and a very weak shoulder around 383nm in ethanol. The high energy band may be due to charge-transfer whereas the band at 560nm may be assigned to d-d transition from the partially spin-paired ground term  $^4A_{1g}$  ( $b_{2g}^2 e_g^2 a_{1g}^1$ ) to the  $b_{1g}$  ( $d_{xy}$ ) orbital i.e. ( $b_{2g}^2 e_g^2 a_{1g}^1$ )  $\rightarrow$  ( $b_{2g}^2 e_g^1 a_{1g}^1 b_{1g}^1$ ). This may be consistent with a square-planar environment for Mn (II) complex. The electronic spectrum of Fe (III) complex shows new bands at 656nm, 563nm and 376nm, which are attributed

to  $^6A_{1g} \rightarrow ^4T_{1g}$  4(G),  $^6A_{1g} \rightarrow ^4E_{1g}$ ,  $^4A_{1g}$  4(G),  $^6A_{1g} \rightarrow ^4T_{2g}$  4(D) respectively<sup>26</sup>. The electronic spectra of the VO(IV) complex showed at 826nm, 635nm, 459nm are assigned to  $^2B_{2g} \rightarrow ^2E_g$ ,  $^2B_{2g} \rightarrow ^2B_{1g}$  and  $^2B_{2g} \rightarrow ^2A_{1g}$  transition. These values suggested a square pyramidal geometry<sup>27</sup> as shown in figure 3.

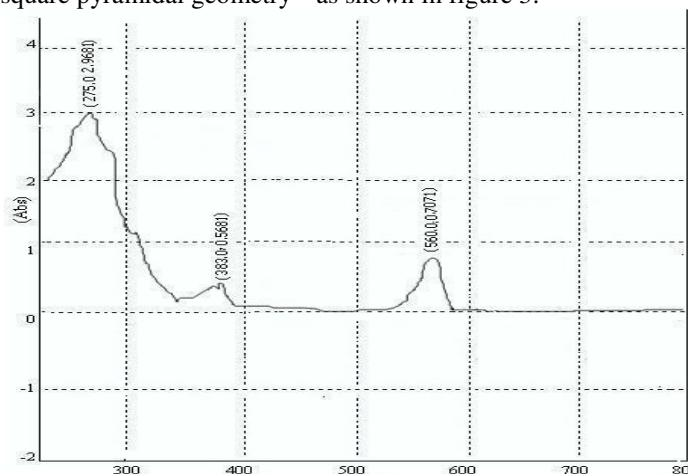


Figure-2  
Electronic spectrum of Mn (II) complexes

**Antimicrobial activities:** The MIC values of antimicrobial activities of ligand and metal complexes by well disk method in DMSO solvent were presented in table-3 and 4 (figure 4 and 5). The antimicrobial activities of tested compounds were stronger than the free ligand and it is increased with increase of concentration of test solution. The antimicrobial activity can be explained by Overtone's concept and the Tweedy's Chelation theory<sup>28-29</sup>. According to the Overtone's cell permeability concept, the liposolubility factor favours the antimicrobial activities of tested compound, since lipid soluble material that only allowed to pass into the membrane of the cell. Based on

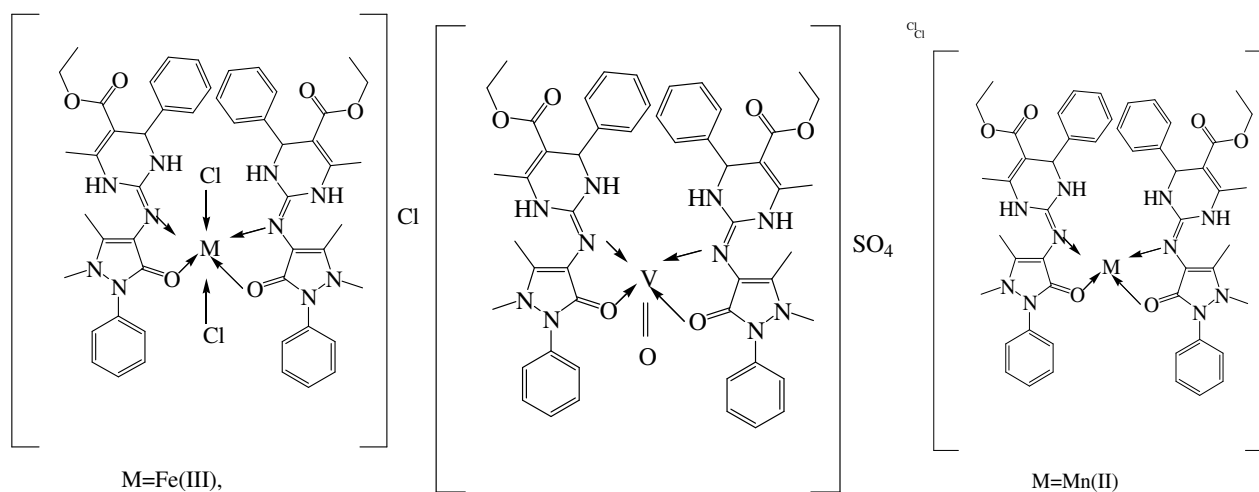
Chelation theory, delocalization of  $\pi$ -electrons (which reduce the polarity of the metal) over the ligand orbital and positive charge of the metal with donor groups enhances the lipophilicity of the complexes. As the lipophilicity increases which in turn enhance the penetration of the complexes into lipid membranes and blocking of the metal binding sites in the enzymes of microorganisms<sup>30</sup>. These complexes also disturb the metabolic activities like cell respiration, synthesis of protein etc, of microorganism. The other factors which affect the antimicrobial activity are solubility, stability, conductivity, and geometry of the complexes.

**Table-3**  
**Antibacterial Activity of Schiff Base and its Complexes**

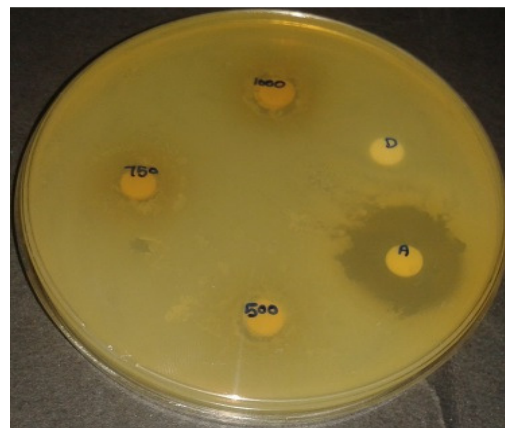
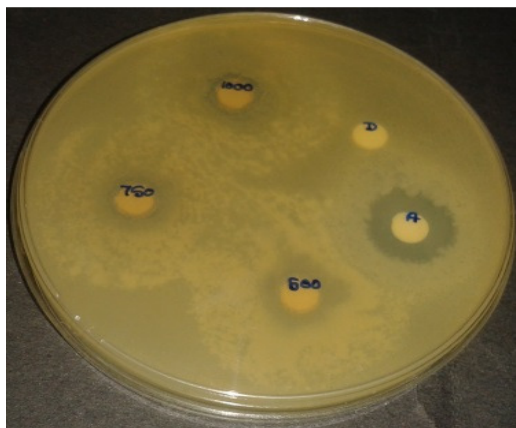
| Name of the bacterial strains | Diameter of Zone Inhibition (mm) |     |     |                                     |     |     |                                       |     |     |                                    |     |     | Solvent DMSO | Standard Drug |
|-------------------------------|----------------------------------|-----|-----|-------------------------------------|-----|-----|---------------------------------------|-----|-----|------------------------------------|-----|-----|--------------|---------------|
|                               | Ligand                           |     |     | [MnL <sub>2</sub> Cl <sub>2</sub> ] |     |     | [FeL <sub>2</sub> Cl <sub>2</sub> ]Cl |     |     | [VOL <sub>2</sub> ]SO <sub>4</sub> |     |     |              |               |
|                               | 1000                             | 750 | 500 | 1000                                | 750 | 500 | 1000                                  | 750 | 500 | 1000                               | 750 | 500 |              |               |
| <i>E.coli</i>                 | 7                                | 6   | 5   | 10                                  | 8   | 7   | 8                                     | 6   | 4   | 7                                  | 6   | 4   | -            | 14            |
| <i>Salmonella typhi</i>       | 3                                | 3   | 3   | 12                                  | 10  | 6   | 8                                     | 7   | 7   | 7                                  | 6   | 6   | -            | 14            |
| <i>Bacillus subtilis</i>      | 7                                | 5   | 4   | 8                                   | 8   | 6   | 6                                     | 5   | 5   | 5                                  | 4   | 3   | -            | 7             |
| <i>Staphylococcus aureus</i>  | 6                                | 4   | 3   | 11                                  | 9   | 6   | 6                                     | 6   | 6   | 5                                  | 5   | 5   | -            | 13            |

**Table-4**  
**Antifungal Activity of Schiff Base and its Complexes**

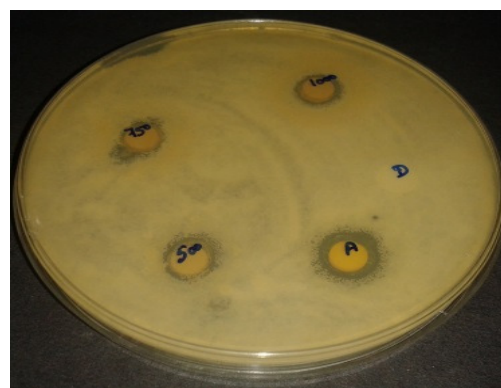
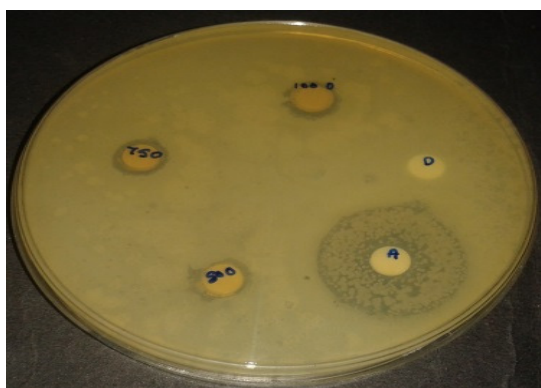
| Name of the fungal strains | Diameter of Zone Inhibition (mm) |     |     |                                     |     |     |                                       |     |     |                                    |     |     | Solvent DMSO | Standard Drug |
|----------------------------|----------------------------------|-----|-----|-------------------------------------|-----|-----|---------------------------------------|-----|-----|------------------------------------|-----|-----|--------------|---------------|
|                            | Ligand                           |     |     | [MnL <sub>2</sub> Cl <sub>2</sub> ] |     |     | [FeL <sub>2</sub> Cl <sub>2</sub> ]Cl |     |     | [VOL <sub>2</sub> ]SO <sub>4</sub> |     |     |              |               |
|                            | 1000                             | 750 | 500 | 1000                                | 750 | 500 | 1000                                  | 750 | 500 | 1000                               | 750 | 500 |              |               |
| <i>Aspergillusniger</i>    | 5                                | -   | -   | 7                                   | 6   | 5   | 5                                     | 5   | 4   | 5                                  | 5   | 5   | -            | 7             |
| <i>Candida</i>             | 7                                | -   | -   | 8                                   | 7   | 7   | 5                                     | 5   | 5   | 5                                  | 5   | 4   | -            | 10            |



**Figure-3**  
**The suggested structure for the ML<sub>2</sub> complexes**



**Figure-4**  
Antibacterial activities of Fe (III) and Mn (II) Complexes



**Figure-5**  
Antifungal activities of VO (IV) and Mn (II) Complexes

## Conclusion

In this paper, the synthesis of novel schiff base ligand and its 3d transition metal complexes derived from condensation of 4-aminoantipyrine comprising with dihydropyrimidinones have been described. The Schiff base behaves as a bidentate ligand and it coordinates through its azomethine nitrogen and the carbonyl group of the five member ring to the central metal atom. This is supported by infrared spectral data. The electronic spectral data confirmed the geometry of Mn(II), Fe(III), VO(IV) as Square planar, Octahedral and Square pyramidal geometry respectively. The complexes are in  $ML_2$  composition, non electrolytes and paramagnetic in nature. The biological activity of ligand is lower than the metal complexes. This means that metal chelation is significantly effective than the antimicrobial behavior of the organic ligand.

## Acknowledgement

The authors acknowledge the financial support rendered by UGC IX plan and also thank Sri Ram Engineering College for extending support.

## References

1. Israa A. Hassan, Transition Metal Complexes of Bidentate Ligand N-aminoquinolino-2-one and Anthranilic Hydrazide, *Research Journal of Chemical Sciences*, **3(12)**,50-53,(2013)
2. Butrus H. Nabeel and Saeed T. Synthesis and Structural Studies on Some Transition metal complexes of Bis-(benzimidazole-2-thio) ethane, propane and butane ligands, *Research Journal of Chemical Sciences*,**2(6)**,43-49,(2012)
3. Z.-L.You and H.-L. Zhu, Syntheses, crystal structures, and antibacterial activities of four schiff base complexes of copper and zinc, *Zeitschrift fur Anorganische und Allgemeine Chemie*, **630(15)**, 2754–2760, (2004)
4. Kabeer A S, Baseer M A, Mote N A, Synthesis and antimicrobial activity of some Schiff bases from Benzothiazoles, *Asian. J. Chem.*, **13(2)**, 496–500, (2001)
5. Rajasekar K, Ramachandramoorthy T. and Balasubramanian S. Synthesis, Spectral characterization and Crystal structure of  $[Cd(4-AAP) 2(NO_2)_2]$  (4-AAP=

- 4-Aminoantipyrine), *Research Journal of Chemical Sciences*, **3(3)**, 48-51 (2013)
6. Desai S B, Desai P B, Desai K R, Synthesis of Some Schiff Bases, Thiazolidinones and Azetidiones Derived From 2,6-Diaminobenzo[1,2-D:4,5-D'] Bisthiazole and their Anticancer Activities, *Heterocycl. Commun.* **7(1)**:83-90, (2001)
  7. Zahid H C, Mohammad S I, Hummara S I, Claudiu T S, Transition Metal Acetylsalicylates and Their Anti-inflammatory Activity, *Journal of Enzyme Inhibition and Medicinal Chemistry*, **17(2)**, 87, (2002)
  8. Sindhukumarn B, Raijuulal G., and Mohanan K, Microwave assisted synthesis, spectroscopic, thermal and biological studies of some Lanthanide (III) chloride complexes with a heterocyclic Schiff's bases, *Synthesis And Reactivity In Inorganic Metal Organic And Nano-Metal Chemistry*, **39**:24, (2009)
  9. Thankamony M. and Mohanan K, Synthesis, spectral studies, thermal decomposition kinetics, reactivity and antibacterial activity of some lanthanide (II) nitrate complexes of 2-(N-indole-2-one) amino-3-carboxyethyl-4-5-6-7-tetra-drobenzo[b]thiophene, *Indian journal of chemistry A*, **46**, 247-251, (2007)
  10. Raman N, Dhavethu Raja J, and Sakthivel A, Synthesis spectral characterization of Schiff base transition metal complexes DNA cleavage and antimicrobial activity studies, *Journal of Chemical Sciences*, **119(4)**; 303-310, (2007)
  11. Manjula B. and Arul Antony S, Preparation, Characterization, Antimicrobial Activities & DNA Cleavage Studies of Schiff Base Complexes Derived From 4-Amino Antipyrine, *Asian Journal of Biochemical and Pharmaceutical Research*, **3(1)**, 168-178 (2013)
  12. Chatterjee N R, Sharma D C, Jadhav G R & Chandak B G, An efficient and Facile Synthesis of Some Ethyl-1-Carbamoyl-4-(2 or 4-Substituted Phenyl)-6-Methyl-2-Oxo-1,2,3,4-Tetrahydropyrimidine-5-Carboxylate Through Ultrasound Accelerated Vilsmeier-Haack Reaction and their Screening as Antiplatelet and Anticardiac Activity, *International Journal of Pharma and Bio Sciences*, **1(3)** (2010)
  13. Tu S, Xiao-Tong Z, Fang F, Xiao-Jing Z, Song-Lel Z, Tuan-Jie L, Da-Qing S, Xiang-Shan W, Shun-Jun J, One-pot synthesis of 3,4-dihydropyrimidine-2-1(H)-ones using boric acids a catalyst, *Tetrahedron Lett*, **44**; 6153-6155 (2003)
  14. Wang Y, Yu J, Miao z, Chen R, Bifunctional primary amine-thioure-TfOH (BPAT-TfOH) as a chiral phase-transfer catalyst; the asymmetric synthesis of dihydropyrimidines, *Organic & Biomolecular chemistry*, **9(8)**, 3050-3054 (2011)
  15. Electronic supplementary Material (ESI) for RSC Advances, The Royal society of chemistry (2012)
  16. Jayabalakrishnan C, and Natarajan K, Ruthenium (II) carbonyl complexes with tridentate Schiff bases and their antibacterial activity, *Transition.Met.Chem*, **27(1)**; 75-79, (2002)
  17. Santha Lakshmi S, Syed Tajueen S, and Kannapan Geetha, Studies on antimicrobial activities of Schiff base complex derived from Dapsone, *Journal of Pharmacy Research*, **4(5)**; 1531, (2005)
  18. Syed Tajueen S, Santha Lakshmi S, and Kannapan Geetha, Studies on antimicrobial activity of some hydrazones and its copper complexes, *Journal of Pharmacy Research*, **3(11)**, 2759, (2010)
  19. Geary W G, The use of conductivity measurements inorganic solve for the characterization of coordination compounds, *Coord. Chem Rev*, **7(1)**; 81-122, (1971)
  20. Raman, A Sakthivel, and K.Rajasekaran, Synthesis and Spectral Characterization of Antifungal sensitive Schiff base transition Metal Complexes, *Mycobiology*, **35(3)**, 150-153, (2007)
  21. Elemike E E, Oviawe A P, and Otuokere I E, Potentiation of the Antimicrobial Activity of  $\beta$ -Benzylimino-2, 3-Dimethyl-1-Phenylpyrazal-5-One by Metal Chelation, *Research Journal of Chemical Sciences*, **1(8)**, 6-11 (2011)
  22. Mishra A.P, and Pandey, L.R, Synthesis, structure and reactivity of oxovanadium(IV) schiff base, *Ind.J .Chem*, **44A**, 1800-1805, (2005)
  23. Wahid U.Malik, G.D.Tuli, R.D.Madan, Selected Topics in Inorganic Chemistry, First Edition 1976, pp390, Reprint (2004)
  24. Kettle S F A, Physical inorganic chemistry oxford, UK; spekturm, (1996)
  25. Suman Malik, Suparna Ghosh, Bharati Jain, Archana Singh, and Mamta Bhattacharya, Synthesis, Characterization and Biological Evaluation of some 3d-metal Complexes of Schiff base Derived for Xipamide Drug, *International journal of Inorganic Chemistry*, 1155, 549805 (2013)
  26. Roy S, Nath Mandal T, and Kumar Bariq A, Metal complexes of pyrimidine derived ligands - Syntheses, characterization and X-ray crystal structures of Ni(II), Co(III) and Fe(III) complexes of Schiff base ligands, *Polyhedron*, **26**, 2303 (2007)
  27. Misbah ur Rehman, Muhammad Imran, Muhammad Arif, Synthesis, Characterization and in vitro Antimicrobial studies of Schiff-bases derived from Acetyl acetone and Amino acids and their Oxovanadium (IV) complexes, *American Journal of Applied Chemistry*, **1(4)**, 59-66 (2013)

28. Thimmaiah K N, Lloyd W D and Chandrappa G T, *Extractive Spectrophotometric determination of molybdenum (V) in molybdenum steels*. *Inorg Chim Acta*, **81**, 106, (1985)
29. Tweedy B G, Plant extracts with metal ions as potential antimicrobial agents, *Phytopathology*, **55**; 910-914, (1964)
30. Manjula B., Arul Antony S. and S. Arul Antony, Studies on DNA Cleavage and Antimicrobial screening of Transition Metal complexes of 4-aminoantipyrene Schiff base, *Research Journal of Chemical Sciences*, **3(12)**, 22-28, (2013)