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Synthesis and Versatile Applications of Schiff Base Transition Metal Complexes

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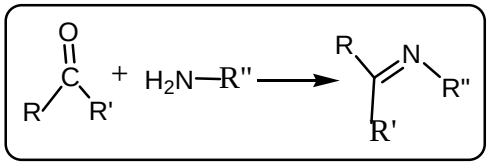
Abstract: ***Schiff bases and their metal complexes plays a very important role in modern co-ordination chemistry. It possesses novel structural features and find number of applications in various fields including food industries,dyes, agricultural ,catalysis, polymer science. The most important feature of Schiff bases is complexation with various metals which lead to the formation of new structures which exhibit wide range of biological activities. Transition metal complexes acts as antimicrobial, antifungal, antiulcer, anticancer agents. This review mainly focuses on the research contributing to the preclinical screenings as well as versatility in various fields. .***

Keywords: Metal complexes, antimicrobial, antifulgal, biological activities

I. INTRODUCTION

In the recent years , considerable research is being undertaken to synthesise a ligand which shows absolute specificity. One such promising agent in this group is Schiff base which are also called as anils, imines or azomethines. These are named as Schiff bases after the scientist Hugo Schiff who first synthesized such compounds.[1] These are the organic compounds formed by condensation of primary amines with an active carbonyl group of aldehyde or ketones and possess a azomethine group (>C=N-).



R,R',R" may be alkyl , aryl , cycloalkyl , heterocyclic radicals which may be variously substituted.

Schiff bases combines with different metals to form coordination or complex compounds and contribute a lot in the development of coordination chemistry. Schiff bases are easy to synthesis. Schiff bases form stable complexes with almost all metal ions including transition , nontransition , inner transition metals. The Schiff base metal complexes are very much important in bioinorganic chemistry, biomedical chemistry , pharmaceutical industries , catalysis ,polymer chemistry, supramolecular chemistry, catalysis, material science.

Some of the important classes of Schiff bases that has been studied extensively are hydrazones ,carbaxones , thiocabazones , thiosemicarbazones , thiocarbazones and oximes etc.[2] Transition metal complexes with heterocyclic Schiff Base ligands possess a variety of applications in pharmacological and agrochemical activities .Among biological activities anticancer ,antimalarial , antitubercular, antiHIV ,antifungal,antimicrobial,antidepressant[3,4] of these complexes were studied. `The synthesis and characterization of schiff base transition metal complexes having nitrogen and oxygen has increased manifold in the last few years.[5,6] There are various methods to synthesize metal complexes[7.] But the most commonly used method is addition of alcoholic solution of metal ions to the alcoholic solution of ligands in presence of aqueous or alcoholic solution of ammonia or sodium acetate. Some times product may obtained in the neutral medium.P^H of the reaction medium may affect the product formation. It also determines the ultimate product formation. It also determines the ultimate compostion , constitution geometry and structure of the resulting complex..Schiff base ligands may be mono, bidentate or polydentate.. Large No. of bidentate ligands are reported.[8] These complexes because of their unique properties are very useful in biochemistry and biomedicine [9,10] These complexes find wide application in the analytical field ,medical field ,catalytic field , industrial field .The synthesis and versatile applications of the Schiff base complexes are summerised here.

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II. SYNTHETIC METHODS OF SCHIFF BASES AND ITS METAL COMPLEXES

A. Scheme 1

Schiff bases o-Vanillidine-2-amino-benzothiazole (VBT), o-Vanillidine-2-amino-4-methyl benzothiazole (VMBT), o-Vanillidine-2-amino-6-chlorobenzothiazole (VCBT), and o-Vanillidin-2-amino-6-bromobenzothiazole (VBBT)were synthesized from substituted 2 aminobenzothiazoles and o-vanilline. These newly synthesized Schiff bases reacted with Copper ions at room temperature (pH 5.89) forming 1:2 [Metal: Ligand] complexes (Fig.1) and hence were used as chromogenic reagents in the spectrophotometric determination of Copper.[11]

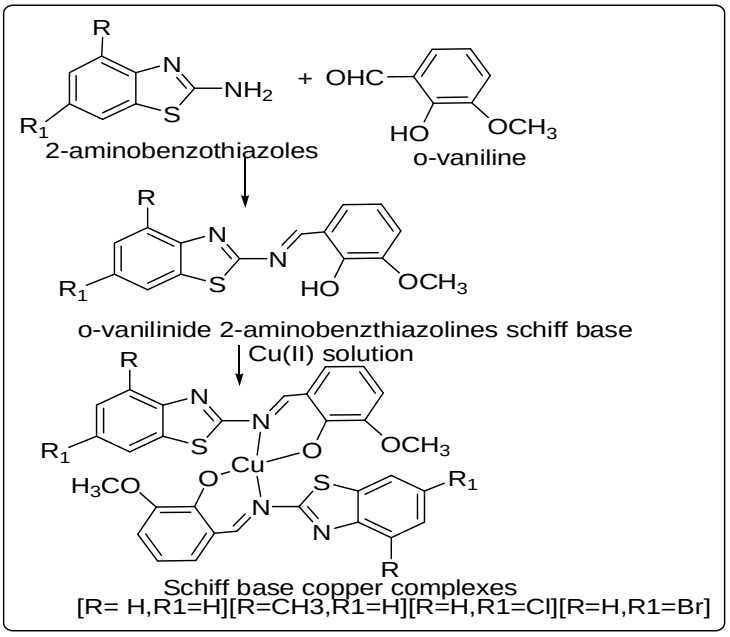

Figure 1

B. Scheme 2 :

Two ligands HL_1 and HL_2 were synthesized by condensation of N-(3-aminopropyl)imidazole and 3,5- dichloro-salicylaldehyde /3,5-dibromo salicylaldehyde using glacial acetic acid in catalytic amount .Complex 1and 2 were synthesized using Cu(OAc)2·H2O in the acetone–methanol mixture with excess aqueous ammonia.[12]

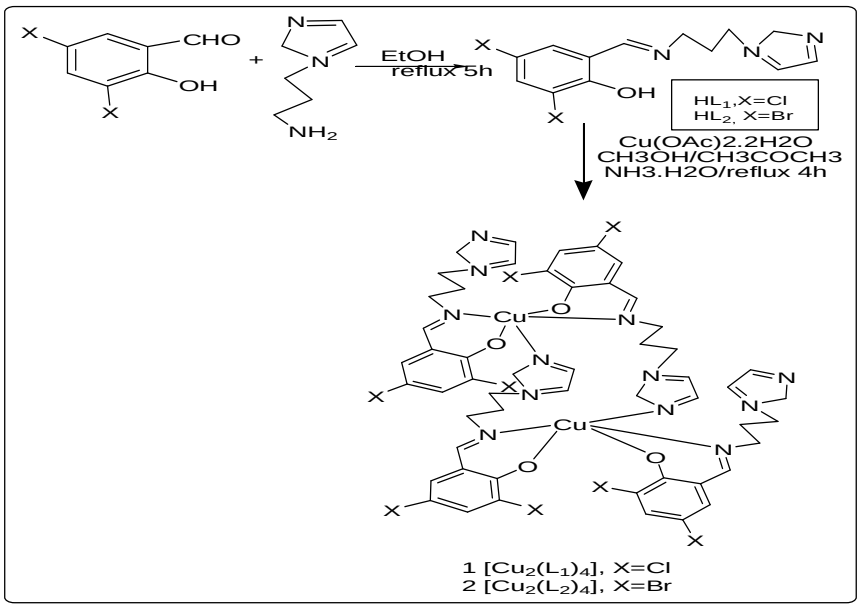

Figure 2

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C. Scheme 3

A new series of Co(II), Ni(II), Cu(II) and Zn(II) metal complexes of a novel ligand 3-(2- (1-(2,4-DihydroxyPhenyl)ethylidene)hydrazinyl)-2H-benzo[b][1,4]oxazin-2-one, (DPE-HBO) were prepared by conventional as well as microwave irradiation. First the ligand DPE-HBO was synthesised by dissolving 3-Hydrazino-1,4-benzoxazin-2-one (HBO) (0.01 M) and 2,4-Dihydroxyacetophenone (0.01 M) in hot water (25 ml) using piperidine in catalytic amount.. The contents were refluxed on a heating mantle for 3 h. A dark yellow colored solid was separated out.In Microwave assisted method equimolar mixture of 3-hydrazino-1,4-benzoxazin-2-one (HBO) and 2,4-dihydroxyacetophenone was mixed thoroughly in a grinder. And then irradiated by the microwave oven by taking 4–5 ml of distilled water as the solvent and two drops of piperidine catalyst. A dark yellow product was separated in 4-5 min. To synthesise the metal complex (Fig.4) , ligand DPE-HBO (0.01 M) was dissolved in hot DMSO (20 ml) and methanolic solution of the respective divalent metal chloride [Co(II), Ni(II), Cu(II) and Zn(II)] was added slowly. pH of each solution was adjusted to 7-8 range by adding 10% methanolic ammonia solution and heated under reflux for 3 h on hot mantle. The bright colored metal complexes were separated.[13]

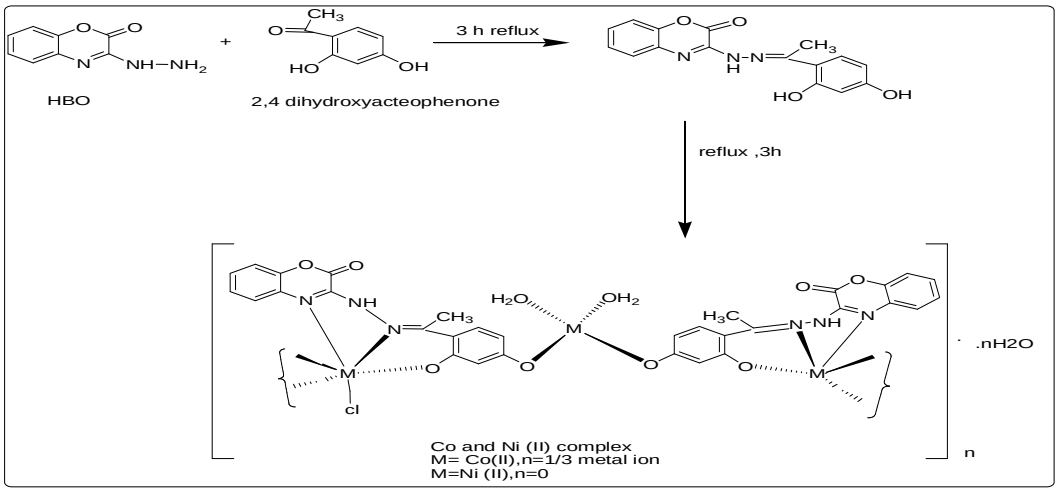

Figure 3

D. Scheme 4

A ligand (1E)-N-((5-((E)-(2,3-dimethyl-1-phenyl-4-pyrazolineimino)methyl)thiophen-2-yl)methylene)-2,3-dimethyl-1-phenyl-4-pyrazolineamine(Fig.4) was synthesised by adding a hot solution of 4-aminoantipyrine in ethanol to a hot solution of 2,5-thiophenedicarboxaldehydein the same solvent and 2:1 proportion and refluexeing the resulting solution for 18 h at 85◦C.Yellow coloured crystals separates out by cooling the solution overnight at 0◦C

The complex of this ligand(Fig.5) was synthesised by adding hot ethanolic solution of the corresponding salts of chloride, nitrate or sulphate of nickel and copper to the hot ethanolic solution of ligand slowly and then refluxed for 26-30 h at 80◦C .The product was obtained after cooling.[14]

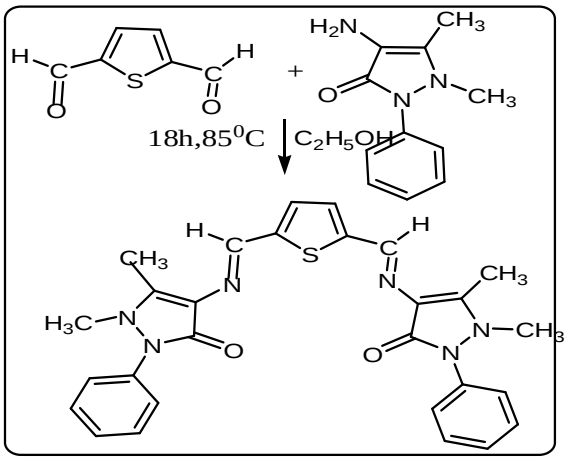

Figure 4

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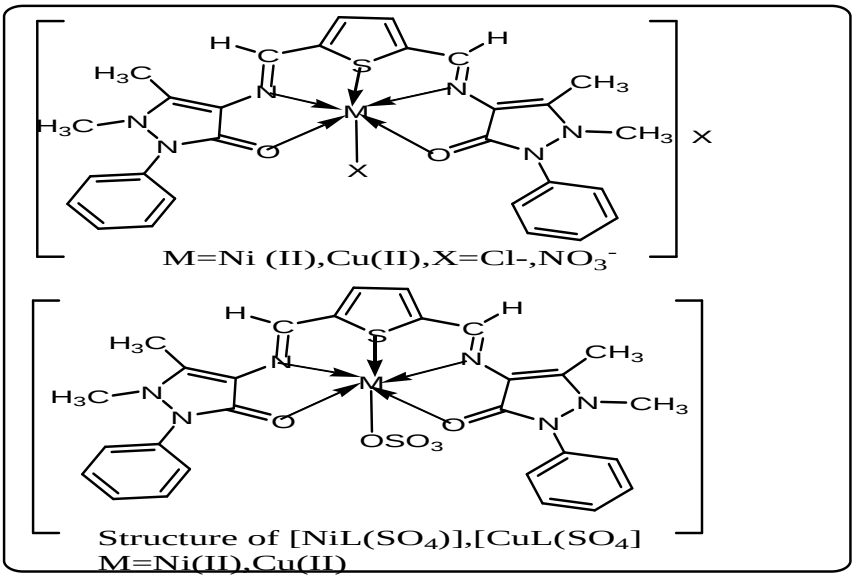

Figure 5

E. Scheme 5

A new Schiff base ligand was synthesized from benzil. firstly the nitration of benzil was carried out using 1 : 1 conc. HNO_3 : H_2SO_4 by refluxing for 6 h. Then the nitro group was reduced by using freshly prepared solution of $SnCl_2$.The obtained product was then dissolved in methanol and salicylaldehyde was added dropwise .The desired ligand (Fig.6)was obtained after evaporating the solvent under vacuum.Copper complex of this ligand(Fig.7) was prepared by adding methanolic solution of Cu-acetate in small portion to methanolic solution of the ligand with constant stirring .thr stirring was continue for 3 h to get the complex product.[15]

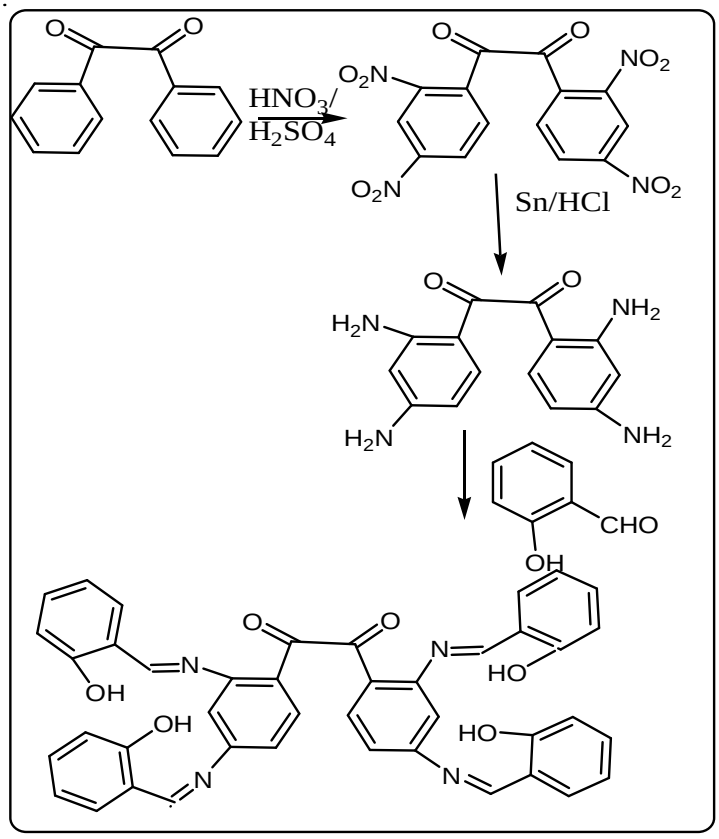

Figure 6

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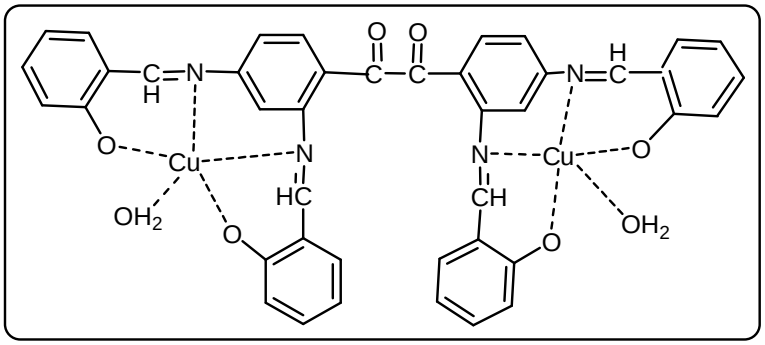

Figure 7

F. Scheme 6

The Schiff-base ligand 1 (E)-2-(((5-methyl-4-phenylthiazol-2-yl)imino)methyl)phenol was synthesized from 2-amino-4-phenyl-5-methyl thiazole and salicyladehyde .Firstly 2-amino-4-phenyl-5-methyl thiazole (1 mmol) was dissolved in about 20 mL absolute ethanol. This ethanolic solution was then added slowly to a magnetically stirred solution of salicyladehyde (1 mmol).Few drops of glacial acetic acid was added to enhance the reaction .This solution mixture was then refluxed for few hours ,concentrated to its half volume ,then cooled and finally n-Hexane was added dropwise to precipitate the desired ligand.The Co(II),Ni(II),Cu(II) and Zn(II) complex of this ligand(Fig.8) was synthesized by refluxing the ethanolic solution of metal salts and ligand for three hours.[16]

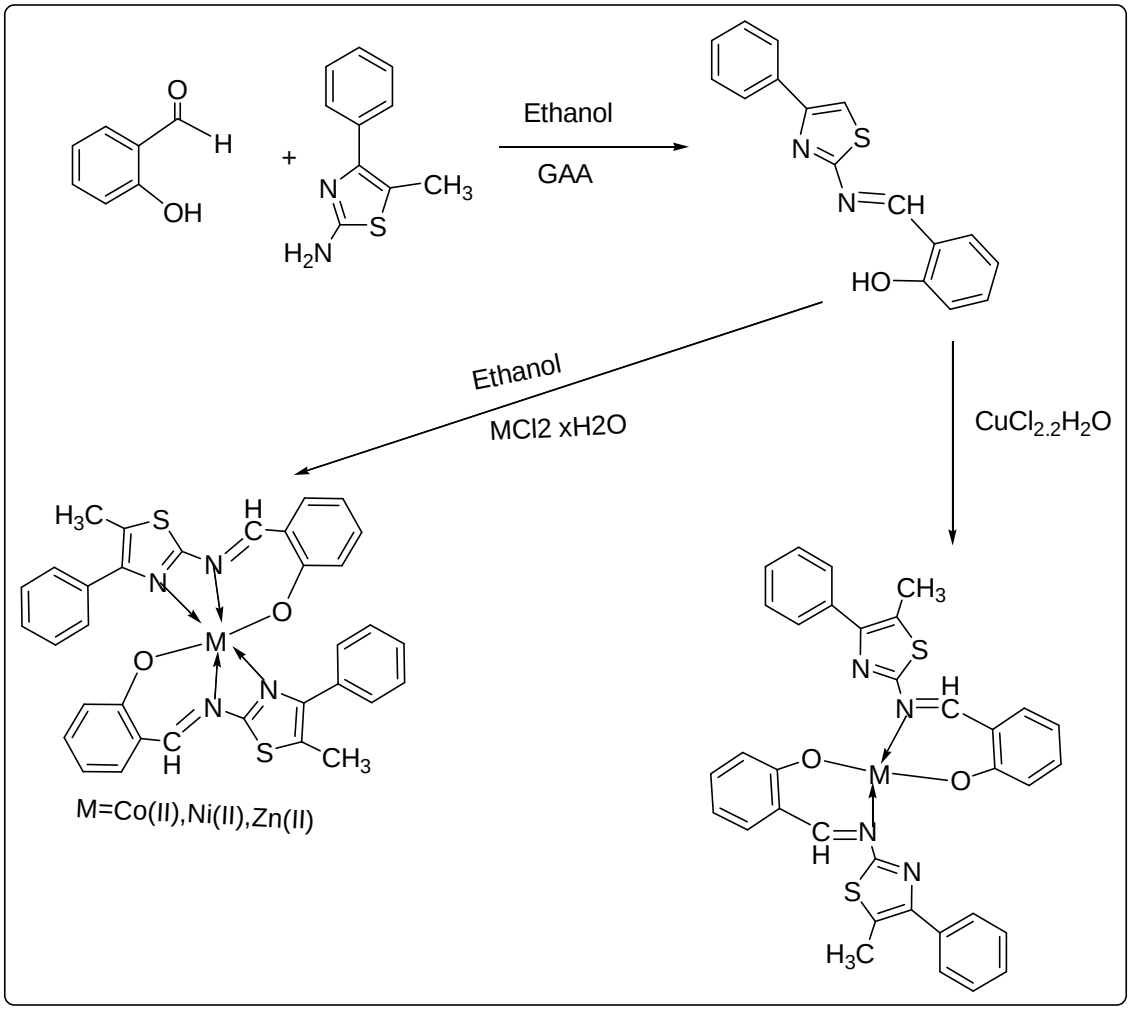

Figure 8

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G. Scheme 7

An ethanolic solution of 2-aminophenol was added to quinoline-2-carboxaldehyde dissolved in 50 ml of DMF in a 1:1 molar ratio. The mixture was refluxed for 2 h. A dark brown precipitate was developed, which was washed with a small amount of ethanol, then filtered, dried and weighed. The synthesis of the Schiff base ligand is presented in fig.10

2.3.2 | Synthesis of mixed ligand complexes An ethanolic solution of metal chloride salt ofCr(III) ,Mn(II) ,Fe(III) ,Co(II) ,Ni(II) ,Cu(II) ,Zn(II) and Cd(II) was added to a mixture of Schiff base ligand dissolved in 50 ml of DMF and of 1,10-phen dissolved in 20 ml of ethanol with a molar ratio of 1:1:1. The final mixture was refluxed for 2 h.Then the precipitates were washed with ethanol, filtered, dried and weighed.[17] The synthesis of the mixed ligand complexes isfig.10

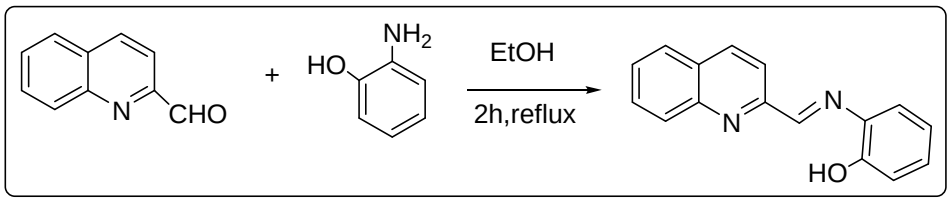

Figure 9

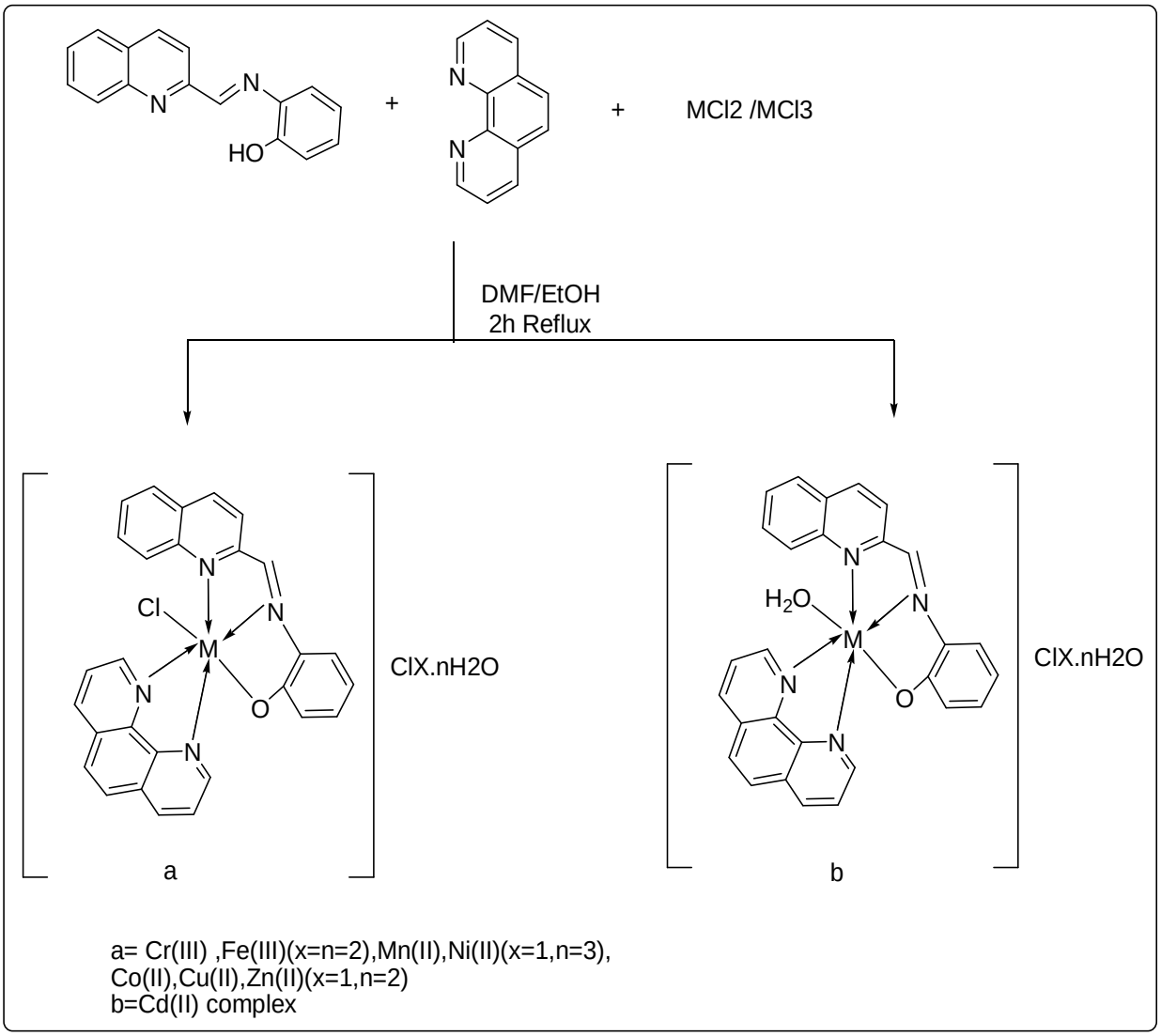

Figure 10

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III. VERSATILE APPLICATIONS OF SCHIFF BASE METAL COMPLEXES

Transition metal complexes of Schiff base ligand found to possess many applications in industrial,medical ,biological, analytical field.

A. In analytical field

The development of use of Schiff bases in analytical method mainly depends on the formation as well as stability of complex which depends mainly on pH, temperature, cation size, and the structure of the ligand. Some of the references explaining the use of Schiff base metal complexes in qualitative and quantitative analysis are given below.

G. Tantaru[18] et al. prepared Salen-type Schiff's base, 1-ethyl- salicylidene-bis-ethylene diamine by condensing ethyl-o-hydroxyphenyl ketone with ethylene diamine and formed brown complex with Mn(II) ion and used for spectrophotometric determination of Mn(II) in pharmaceutical products .

Fakhari[19] et.al synthesized N,N′-bis(3-methylsalicylidene)-ortho-phenylene diamine (MSOPD).This Schiff base formed a 1:1 complex with Ni ion at room temp. and used in the spectrophotometric determination of nickel in some natural food samples .

Potentiometric sensors for Dy(III) based on a Bis-pyrrolidene Shiff`s bases have been prepared by Ganjali[20] et al. which are used in the potentiometric determination of Fluoride ions in mouthwash by the titration against Dy(III).

Kim et al [21]. used salophen having phenyldiamine backbone for trace determination of Ni(II), Co(II) and Cu(II) in water samples by solvent extraction method. Similar study using salophen has been carried out for trace determination of Fe(II) and Fe(III) in water samples[22].

B. In Catalytic Field

Schiff base transition metal complexes act as efficient catalysts both in homogeneous as well as heterogeneous reactions due to their high thermal and moisture stabilities. Schiff base complexes catalyses no.of reactions like oxidation[23] , epoxidation[24-26], oligomerisation of ethylene [27,28] oxidative addition reactions[29] , reduction of thionyl chloride [30,31], epoxidation of olefins [32,33].

Nanosized Cu and Ni Schiff-base complexes, obtained from the condensation of 2-amino-3-hydroxypyridine, with either 3-methoxysalicylaldehyde (ahpv) or 4-nitrobenzaldehyde (ahpnb), were synthesized . These nanosized Schiff-base Cu/Ni complexes and their oxides showed remarkable catalytic activity towards the selective oxidation of benzyl alcohol in aqueous H_2O_2/ dimethylsulfoxide (DMSO) solution. And yielded about 94% and 98% benzldehyde resp. with 100% selectivity[34].

Catalytic activities of five ternary copper (II)-Schiff base complexes (Fig.11)of ligands derived from 3 methoxy salicylaldehyde (MS) or 4 diethylamino salicylaldehyde (DS) and amino acids {L-phenylalanine (Phe), L histidine (His) or DL-tryptophan (Trp)}, as primary ligands, and 2,4 0-bipyridyl (DP) as a secondary ligand were studied by Adam[35]et al in the oxidation of benzyl alcohol to benzaldehyde by an aqueous H2O2 in different reaction conditions.

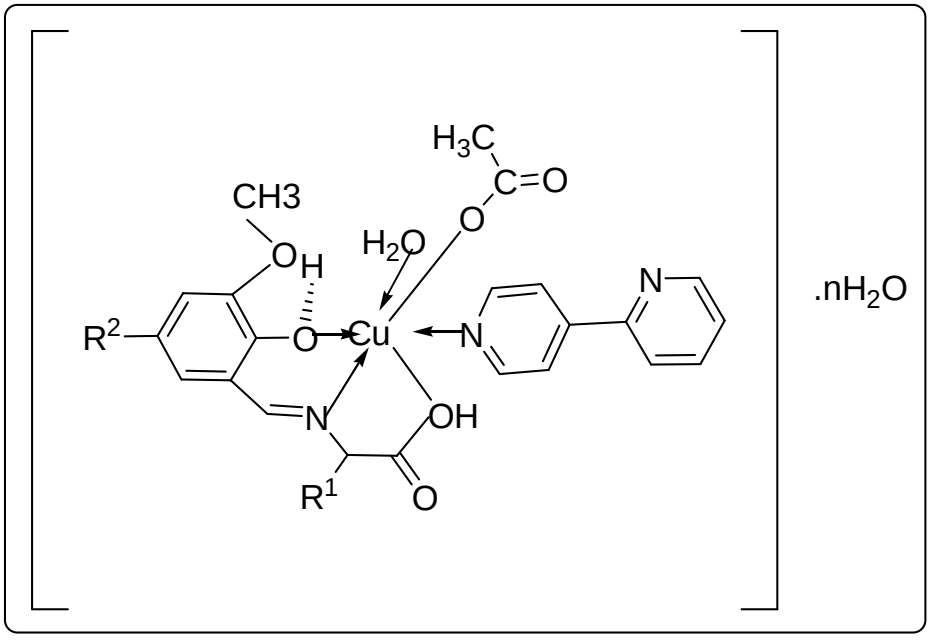

Figure 11

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Ahmadi[36] et al studied the Catalytic properties of Co(II) and Cr(III) Schiff base complexes (Fig.12) obtained from thiourea in oxidation of toluene using hydrogen peroxide (H_2O_2) as the oxidant. The Co(II) complex was catalytically active towards toluene, but Cr(III) complex did not show any catalytic activity.

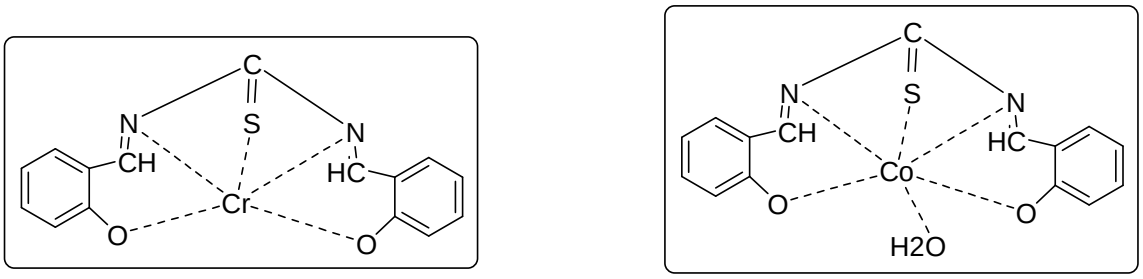

Figure 12

C. Biological Field

Schiff bases and their metal complexes are biologically active compounds and used as metallo-drugs. Most of the Schiff base complexes mainly heterocyclic Schiff base complexes acts as effective anticancer agents. Cancer is the second most leading cause of human death in which a group of cells display uncontrolled growth .[37-39]

A Schiff base ligand was synthesized from 4-chloro-*o*-phenylenediamine and 3,5-dichloro-2-hydroxyacetophenone and its homo and hetero binuclear oxygen bridged Cu (II) and Ni(II) complexes were prepared.(Fig.13) The Schiff base and its complexes were screened for their antimicrobial and anticancer activity. Homobinuclear Cu complexes were found to be more active than other mono and binuclear complexes. Similarly the anticancer activity was studied by treating the complexes with MCF -7 cell line (human breast cancer cell) and found that homobinuclear Cu(II) complex showed high cytotoxicity indicating as potential anticancer agent.[40]

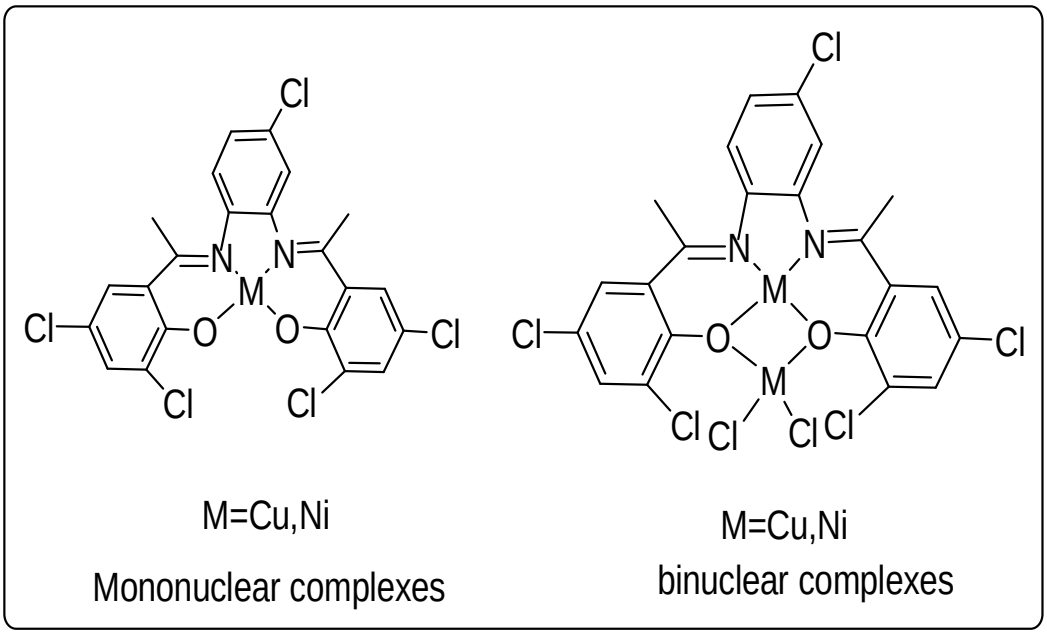

Figure 13

A new Schiff base ligand (HL)was synthesized from quinoline-2-carboxaldhyde with 2-aminophenol and by using 1,10-phenan as second ligand,. its mixed ligand complexes of Cr(III), Mn(II), Fe(III), Co(II), Ni(II), Cu(II),Zn(II) and Cd(II) were synthesized.(Fig.14) All these ligands and mixed metal complexes were screened for their anticancer activity against two cell lines: breast cancer cell line(HCT-116) and colon cancer cell line (MCF-7) and found that the metal complexes showed higher activity than that of HL, especially theCu(II) complex which showed the highest IC50 against breast cell line.[41]

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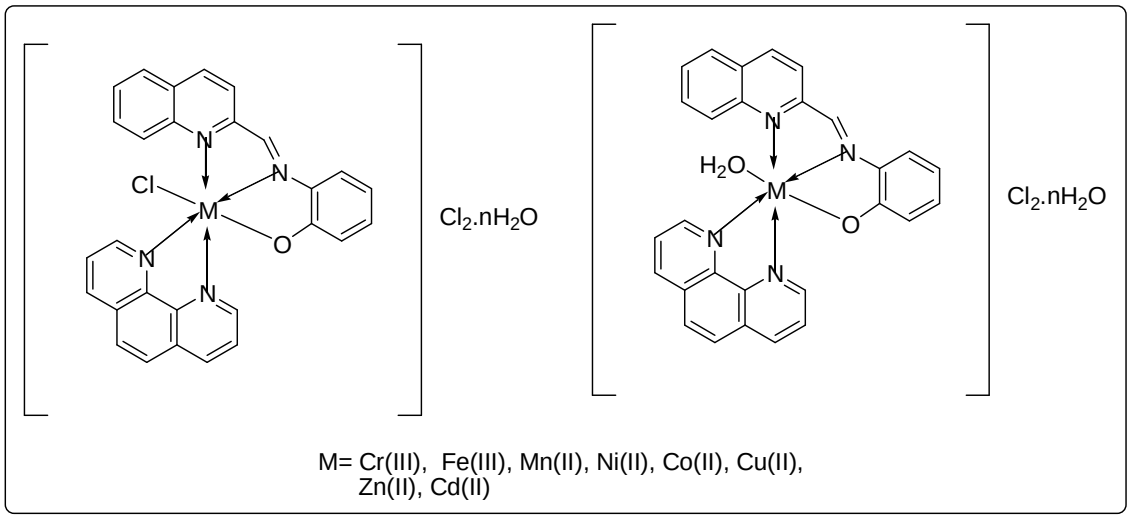

Figure 14

$[Mn(penh)_2]$, $[Co(penh)_2]$, $[Cu(penh)_2]$,and [Cd $(penh)_2$] complexes of (E)-N'-(1-(pyridin-2-yl)ethylidene) nicotinohydrazide (penh) were synthesized and their cytotoxic activity against three cancer cell lines A549 human lung cancer, BGC823 human gastric cancer and Eca109 human esophageal cancer were studied and found that these complexes acts as a novel therapeutic reagent in the treatment of cancer.[42]

Copper(II) complex, viz. cis-[dichloro (N1-(2-benzyloxybenzylidene) pyridine-2-carboxamidrazone) copper(II)] The in vitro antitumor activity of this complex and the parent ligand is determined against the human breast cancer cell-line MCF-7 which revealed that copper complexation renders a highly antiproliferative compound with IC50 value of 3 lM [43].

A series of four new mixed-ligand copper(II) complexes (**1–4**) of the type [Cu(L)(diimine)] (ClO_4) [where L is 2-((1*H*-imidazol-2-yl)methylene)-*N*-phenylhydrazinecarbothioamide and the diimines are 1,10-phenanthroline (phen, **1**), 2,2′-bipyridine (bpy, **2**), 4,4′-dimethyl-2,2′-bipyridyl (dmbpy, **3**), and 2,2′-dipyridylamine (dpa, **4**)] have been successfully synthesized.(Fig.15) These complexes showed efficient antibacterial activities against *Staphylococcus aureus* (Gram positive) and *Pseudomonas aeruginosa* (Gram negative). Furthermore, studies of their *in vitro* cytotoxicity against AGS cancer cells indicated promising antitumor activity with significant IC_{50} values [44].

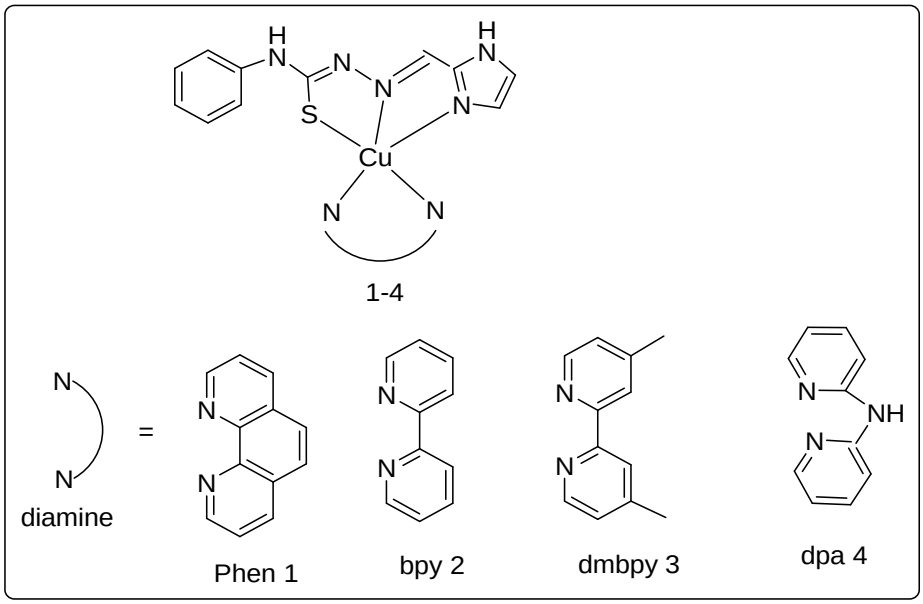

Figure 15

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Mononuclear copper(II) complexes $[Cu(L1)(NO_3)_2]$ (**1**) and $[Cu(L2)(CH_3O)NO_3]$ (**2**) OF Two hydrazone ligands 8-quinolinecarbaldehyde 3-methoxybenzoylhydrazone (L1) and 8-quinolinecarbaldehyde benzoylhydrazone (L2) were synthesized.(Fig.16)It was found that these complexes exhibited significant cytotoxicity against a human cervical cancer cell line (HeLa), and showed better activity than cisplatin. The complexes arrested HeLa cell cycle at the G2 phase.[45]

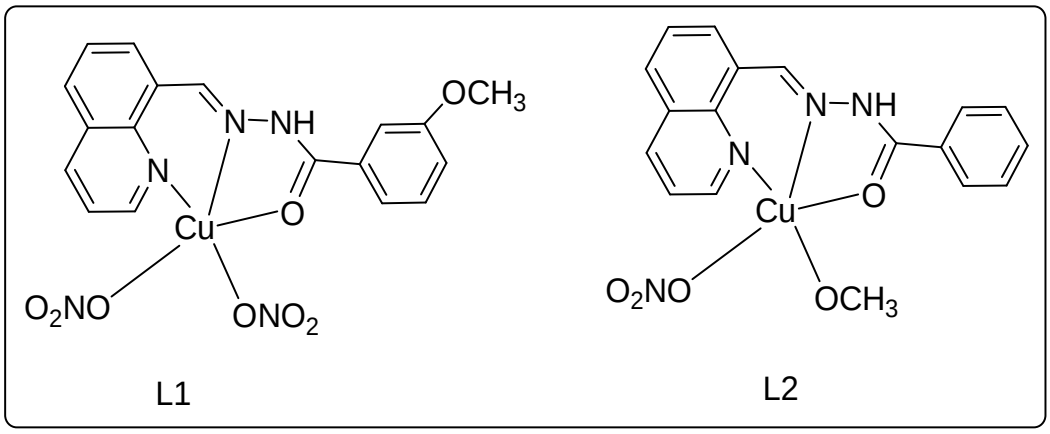
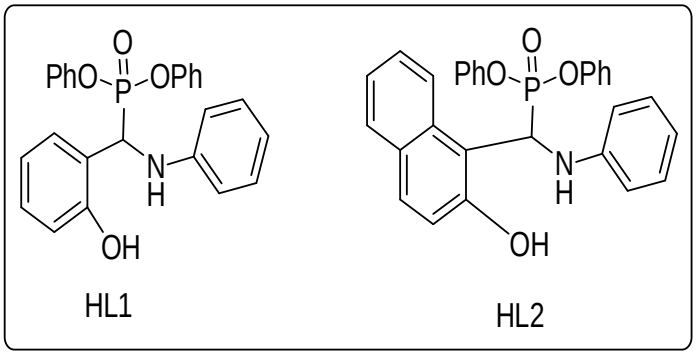


Figure 16

A one pot procedure was used to synthesize two new derivatives of a-aminophosphonates. Novel copper(II) complexes of a-aminophosphonates were synthesized by coordinating different coppersalts with the newly synthesizeda-aminophosphonates. Their structures were characterized by dif-ferent spectral and analytical techniques. Evaluation of the metal-free ligands HL1, HL2, and theirCu(II) complexes(Fig.17) against human colon carcinoma HT-29 cell lines was performed, using cisplatinas a reference drug. The results indicated that the complexes of the ligand HL1exhibitedenhanced anticancer activity, while ligand HL2complexes showed decreased anticancer activity.[46]



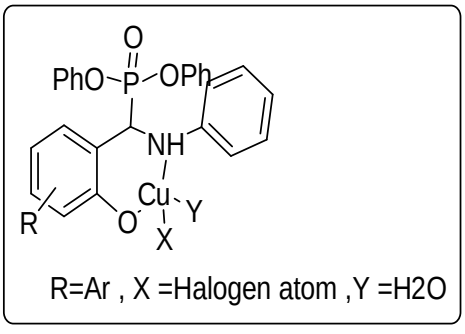


Figure 17

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D. Industrial Field

Schiff base metal complexes are extensively used in various types of industries like dyes, platic ,leather industries.[47-51]

A new Cu(II) complex of monomeric Schiff base 2–((E)–(2–hydroxypropylimino) methyl)–4– nitrophenol] was synthesized. (Fig.18) Both the Schiff base and its copper complex showed luminescent properties which revealed its use as novel potential candidates for applications in textile such as UV-protection, antimicrobial, laundry and functional bleaching treatments[52].

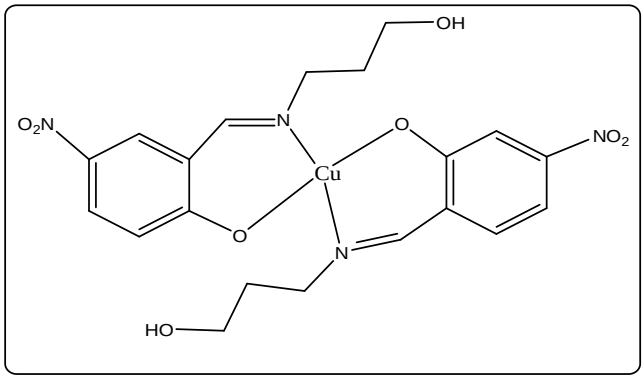

Figure 18

The development of new light-emitting materials has attracted significant attention due to their diverse applications, including the fabrication of light-emitting devices [53-55]. Many Schiff bases and their complexes shows electroluminescent properties and hence used as light emitting layers ine electrolumicescent devices. [56-60]

A Schiff base ligand of pyrimidine amine with 2-hydroxy-1-naphthaldehyde and its Co(II), Ni(II), Cu(II) and Pd(II) metal complexes were synthesized.(Fig.19)These synthesized compounds showed strong photoluminescence and found to be useful as an auxillary or active layers in the fabrication of electroluminescent devices like solar cells ,LED, chemical sensor, biosensor, super capacitors etc[61].

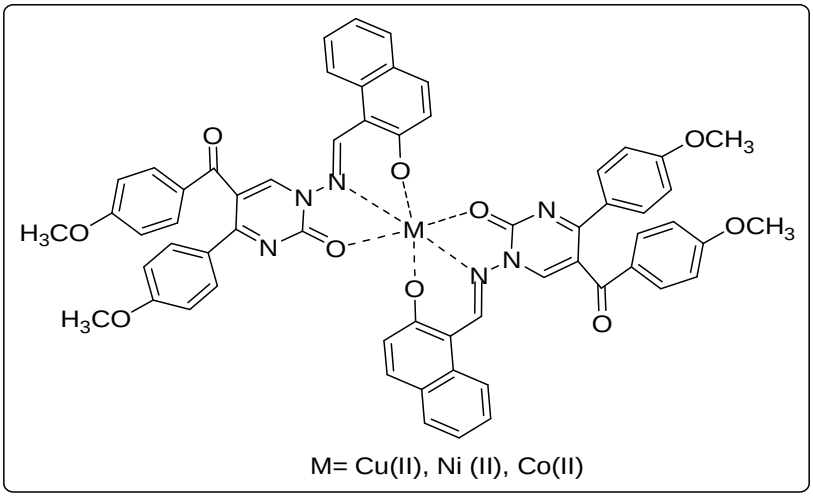

Figure 19

Copper(II) and zinc(II) complexes of the Schiff base ligands N-[(2-pyridyl)-methyl]-salicylimine (Hsalampy),N-[2-(N,N-dimethyl-amino)-ethyl]-salicylimine (Hsald-men), andN-[(2-pyridyl)-methyl]-3-methoxy-salicylimine (Hvalampy) were synthesised and subjected to their luminescent properties. It was found that Zn complexes exhibited interesting luminescent properties.[62]

IV. CONCLUSIOS

Schiff bases are the versatile ligand which coordinate to the metal forming complexes. These complexes have wide applications in every field.But the most promising is the biological field. Heterocyclic Schiff base metal complexes act as excellent antimicrobial as well as anticancer agent. But still more research is needed in the development of these metal complex drugs.

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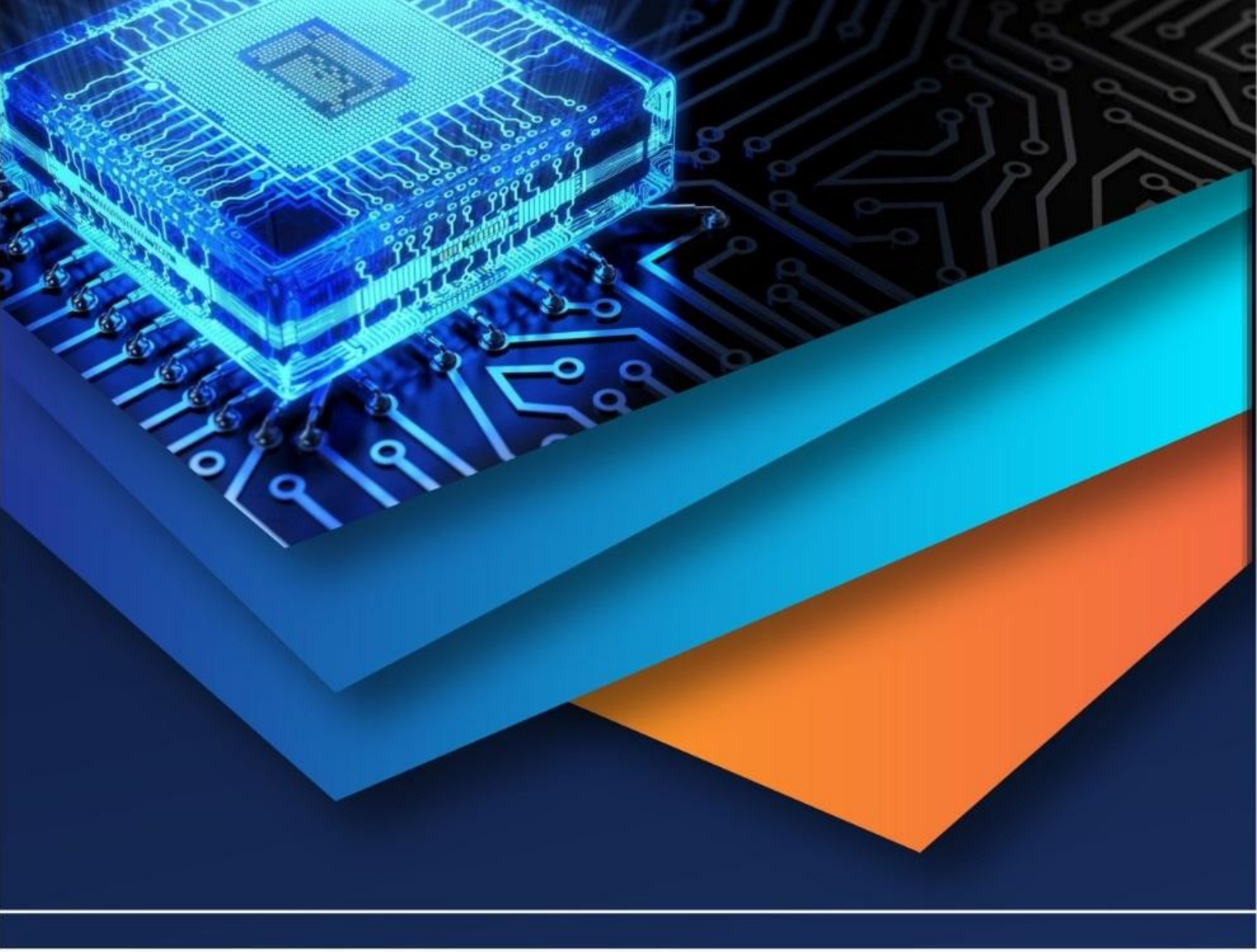

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