



Cu(II) COMPLEXES WITH SOME TRIDENTATE N₂O DONOR SCHIFF BASE LIGANDS OF ETHYLENEDIAMINE DERIVATIVES: A COMPREHENSIVE REVIEW

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ABSTRACT :

The proposed review work focuses on the systematic study of the polynuclear Cu(II) complexes containing tridentate N₂O donor Schiff base ligands as N,O donor derived from ethylenediamine derivatives in presence of several auxiliary bridging groups. The nuclearity, geometry around Cu(II) centers, bridging mode of ligands and magnetic properties of complexes are describe briefly.

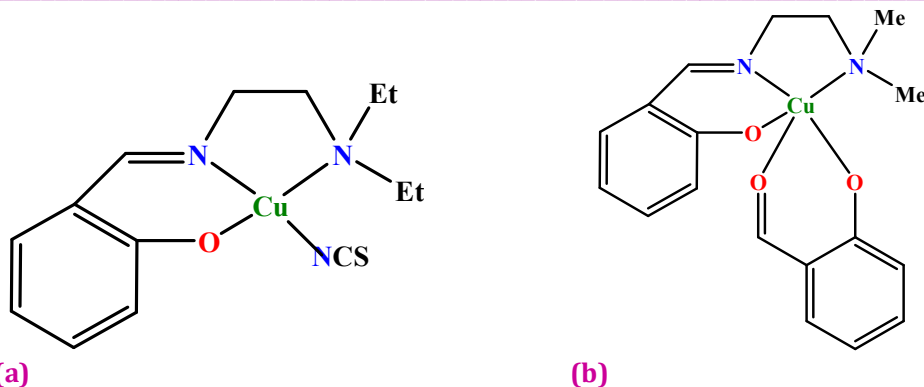
KEYWORDS : Crystal structure; Copper(II); Schiff base; Ethylenediamine; Dinuclear, Polynuclear, Coligands.

INTRODUCTION:

The coordination chemistry of copper(II) complexes with multidentate ligands, mainly with Schiff bases, has always been an important area of research due to their diverse nature. A large number of Cu(II)-Schiff base complexes have been synthesized and characterized in recent time for their important catalytic, magnetic and biological properties [1-18]. The potential role played by copper(II), present in the active sites of many metalloproteins [19-20], has stimulated the design of new Schiff bases and their copper complexes as models. We have discussed about the structural and magnetic properties of the Cu(II) complexes of tridentate N₂O donor Schiff base ligands derived from the 1:1 condensation of N-substituted ethylenediamine with different kind of carbonyl compounds.

MONONUCLEAR COMPLEXES

The crystal structures of several mononuclear Cu(II) complexes of these type of Schiff base ligands have been reported in the literature. Most of the mononuclear Cu(II) complexes possesses square planar geometry, in which the three donor atoms of the Schiff base coordinate meridionally and various anionic coligands (*e.g.* azide, halide, thiocyanate, acetato *etc.*) being coordinated to the remaining site (Scheme 1a) [21]. A series of similar type of mononuclear Cu(II) complexes have been synthesized by changing the substituents in the amine or aldehyde part of the tridentate Schiff base ligands [22].

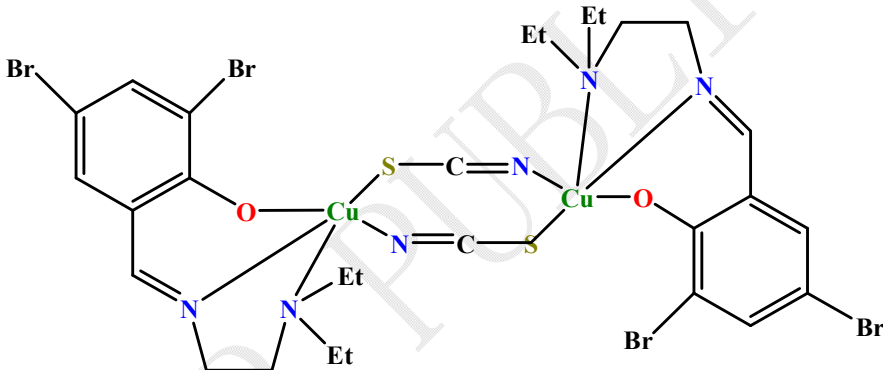


Scheme I. Schematic view of (a) square planar and (b) (4+1) square pyramidal mononuclear Cu(II) complexes of tridentate N₂O donor schiff base ligands.

G. Das *et al.* reported a mononuclear (4+1) elongated square planar Cu(II) complex in which one tridentate Schiff bases being coordinated to the metal center and other two sites occupied by another coligand (Scheme 1b) [23]. A series of similar type of mononuclear Cu(II) complexes have been synthesized by changing the substituents in the amine or aldehyde part of the tridentate Schiff base ligands or by changing the coligands [24].

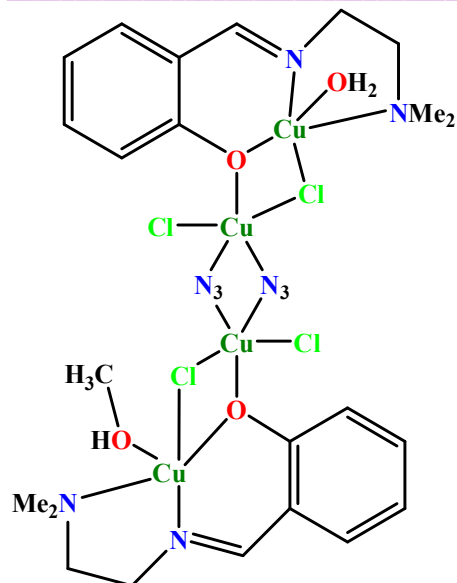
Dinuclear complexes

Z. Hong *et al.* reported a dinuclear Cu(II) complex of tridentate Schiff base ligand with thiocyanate as bridging coligand (Scheme 2) [25].



Scheme 2. Schematic view of dinuclear Cu(II) complexes of tridentate N₂O donor schiff base ligands.

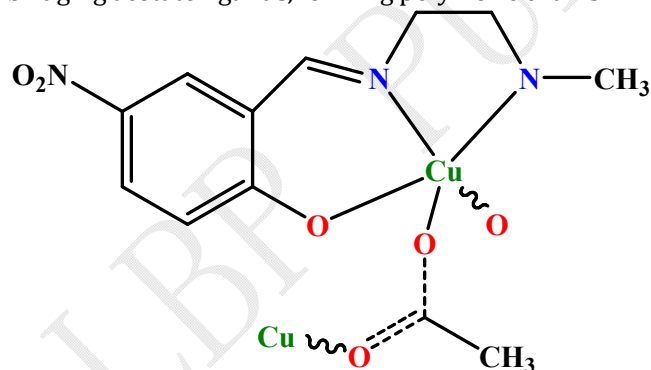
Z.-L. You *et al.* reported a complex {bis(μ₂-Azido-N,N)-bis(μ₂-chloro)-bis(μ₂-2-(2-((dimethylamino)ethylimino)methyl)phenolato-N,N',O,O)-aqua-dichloro-methanol-tetra copper(II)} using a N₂O donor Schiff base ligand. The crystal structure of the complex consists of [Cu₄L^c(N₃)₂Cl₄(OH₂)(CH₃OH)] (where L^c=2-[(2-dimethylaminoethylimino)methyl]phenol) (Scheme 3) [26].



Scheme 3. Schematic view of polynuclear Cu(II) complexes of tridentate N₂O donor schiff base ligands with azido and chlorido as bridging coligands.

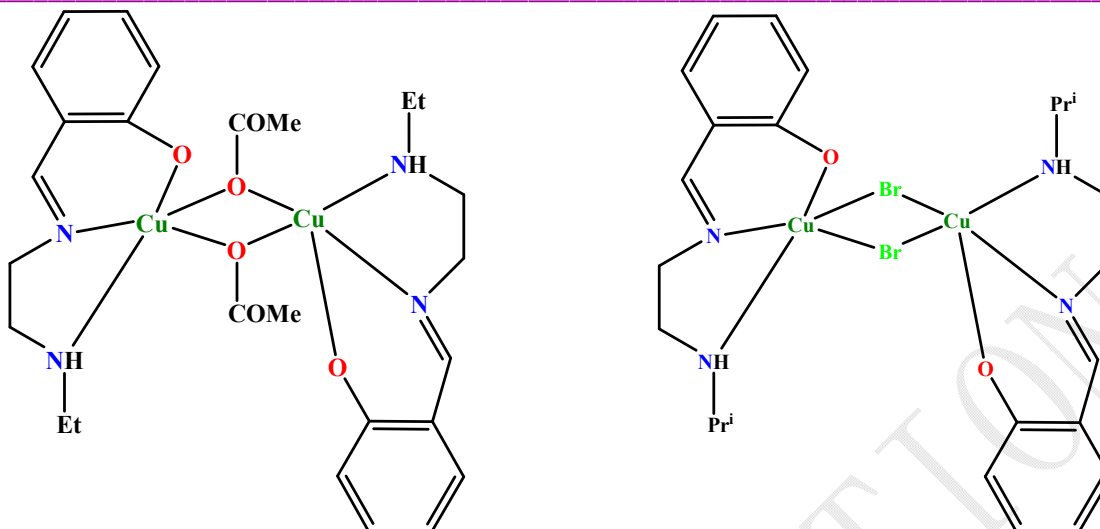
A novel centrosymmetric dinuclear, [Cu₂(NCS)₂(C₁₀H₁₃N₂O)₂] complex has been synthesized and characterized by Y.-J. Wei *et al* [27]. A novel acetate bridged polynuclear Schiff base Cu(II) complex, [CuL^D(CH₃COO)]_n (L^D = 4-nitro-2-[(2-methylaminoethylimino)methyl]phenol), has been synthesized and characterized by elemental analysis, IR spectrum and single crystal X-ray diffraction by C. Wang. Each Cu atom in the complex is five coordinate in a trigonal bipyramidal configuration, with one imine N atom and two acetate O atoms defining the basal plane, and one phenolate O and one amine N atoms occupying the axial positions [Scheme 4] [28].

The [4-nitro-2-[(2-methylaminoethylimino)methyl]phenolato]copper(II) units are linked by the bridging acetate ligands, forming polymeric chains.



Scheme 4. Schematic view of a novel acetate bridged polynuclear Schiff base Cu(II) complex of N₂O donor Schiff base ligand.

A pair of novel dinuclear Schiff base copper(II) complexes, [Cu₂(L^E)₂(MeCOO)₂].2H₂O and [CuBr₂(L^F)₂].2MeOH where HL^E = 2-[(2-ethylaminoethylimino)methyl]phenol and HL^F = 2-[(2-isopropylaminoethylimino)methyl]phenol, have been synthesized from an identical synthetic procedure and similar Schiff base ligands but with different Cu(II) salts by R.-H. Hui *et al.* [Scheme 5] [29]. Each copper (II) centre in the complexes is in a square pyramidal coordination. There exists crystallographic imposed centre of inversion in each of the complexes.

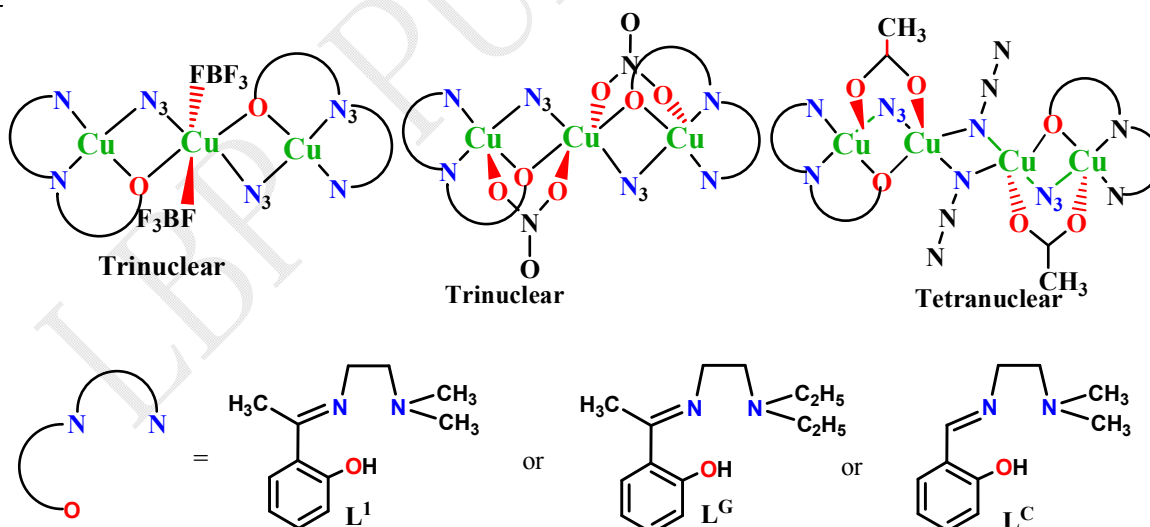


Scheme 5. Schematic view of a pair of novel dinuclear N₂O donor Schiff base Cu(II) complexes.

A large number similar type of dinuclear and trinuclear Cu(II) complexes have been synthesized by changing the substituents in the amine or aldehyde part of the tridentate Schiff base ligands or by changing the coligand by different research groups [30].

Trinuclear, tetranuclear and polynuclear complexes

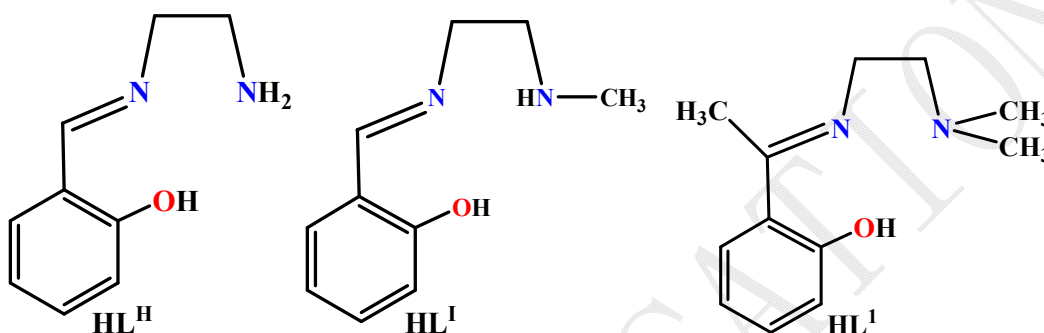
Ghosh *et al.* reported three novel mixed bridged trinuclear and one tetranuclear Cu(II) complexes of tridentate N₂O donor Schiff base ligands [Cu₃(L¹)₂(μ_{1,1}-N₃)₂(CH₃OH)₂(BF₄)₂], [Cu₃(L¹)₂(μ_{1,1}-N₃)₂(NO₃)₂], [Cu₃(L^G)₂(μ_{1,1}-N₃)₂(NO₃)₂] and [Cu₄(L^C)₂(μ_{1,1}-N₃)₄(CH₃COO)₂] [scheme 6] [31]. Also, a theoretical study using density functional theory methods has been performed to explain the magnetic properties of the complexes. First three complexes are the first examples of linear mixed (azido/phenoxo/nitrato) bridged trinuclear Cu(II) complexes, which have been characterized structurally and magnetically.



Scheme 6. Schematic view of three novel mixed bridged trinuclear and one tetranuclear copper(II) complexes of tridentate N₂O donor Schiff base ligands.

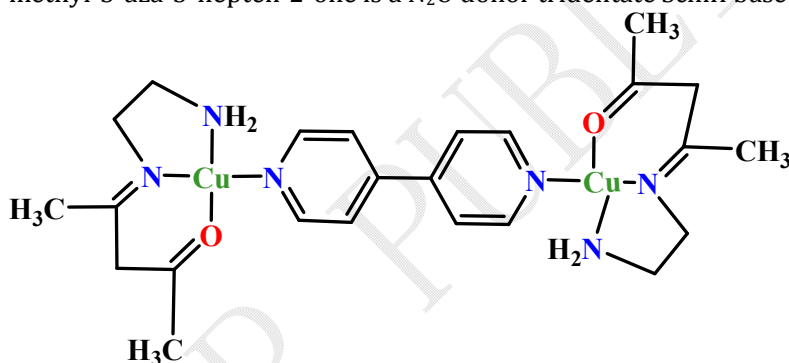
The same group report the synthesis, crystal structure, and magnetic properties of three new cyclic trinuclear μ₃-hydroxo bridged Cu(II) complexes of general formula, [(CuL)₃(μ₃-OH)]²⁺ [32] in

which L is a Schiff base derived from three different diamines (ethylenediamine, N-methylethylenediamine and N,N-dimethylethylenediamine) and salicylaldehyde or *o*-hydroxyacetophenone (ligands HL^H, HL^I, and HL^I in (Scheme 7). Complexes are [(CuL^H)₃(μ₃-OH)](ClO₄)₂·3.75H₂O, [(CuL^I)₃(μ₃-OH)](ClO₄)₂ and [(CuL^I)₃(μ₃-OH)](BF₄)₂·0.5CH₃CN where first two exhibit weak antiferromagnetic exchange interactions whereas third one shows ferromagnetic interaction, all of them confirmed by fitting procedures using empirical magnetic exchange Hamiltonians. Magnetostructural correlations for these complexes have been proposed by means of calculations based on Density Functional Theory (DFT).



Scheme 7. ligands HL^H, HL^I, and HL^I.

C. Biswas *et al.* synthesized and characterize a novel dinuclear Cu(II) complex using HL^I and 4,4'-bipyridine having the molecular formula [(Cu(L))₂(4,4'-bipy)](BF₄)₂ where HL^I = 7-amino-4-methyl-5-aza-3-hepten-2-one is a N₂O donor tridentate Schiff base ligand [Scheme 8] [33].

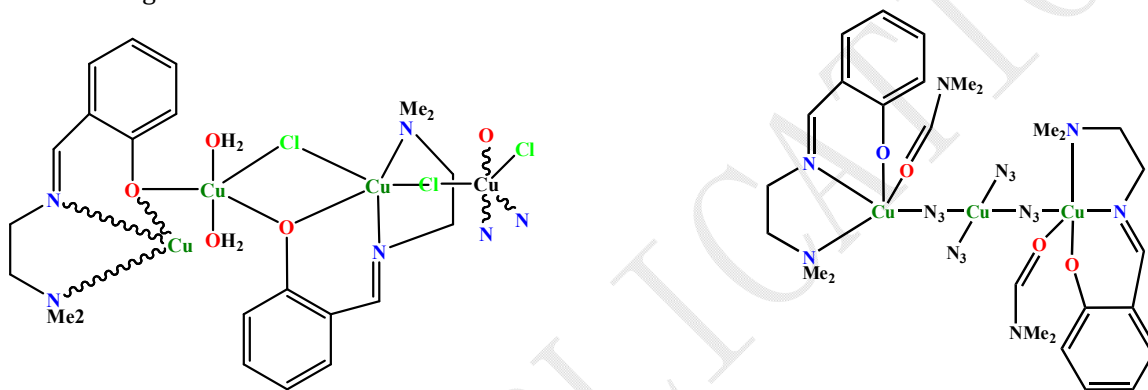


Scheme 8. Schematic view of a novel 4,4'-bipyridine bridged dinuclear Schiff base Cu(II) complex of N₂O donor Schiff base ligand.

Naiya *et al.* report the synthesis, structural characterization, and variable-temperature magnetic behavior of four new complexes: a mononuclear [CuL^I(N₃)], a single μ_{1,1}-azido bridged dinuclear [CuL^I(N₃)₂], a 2D coordination polymer with μ_{1,1}- and μ_{1,1,3}-azide bridges [Cu₇L^I(N₃)₁₂]_n, and a μ_{1,3}- and μ_{1,1}- azido bridged double stranded helix, [Cu₂L^I(dmen)(N₃)₃]_n. All four complexes have been prepared by using the same N₂O donor Schiff base ligand (2-[1-(2-dimethylaminoethylimino)ethyl]phenol) (HL^I), the condensation product of N,N-dimethyl-1,2-diaminoethane (dmen) and *o*-hydroxyacetophenone by varying the reaction conditions. The factors that allow the conversion of one complex into another have also been explored in detail [34]. The four complexes have been characterized by X-ray structural analyses and variable temperature magnetic susceptibility measurements. The magnetic properties of first two complexes show the presence of very weak antiferromagnetic exchange interactions mediated by a ligand π overlap and by an asymmetric 1,1-N₃ bridge respectively. Third complex is a decorated

chain structure with both ferro- and antiferromagnetic interactions. Fourth Complex is an alternating helicoidal chain with two weak antiferromagnetic exchange interactions.

Four new Cu(II) complexes of formulae $[\text{Cu}_3\text{L}^c_2(\text{H}_2\text{O})_2\text{Cl}_3]\text{Cl}\cdot 6\text{H}_2\text{O}$, $[\text{Cu}_3\text{L}^c_2(\text{N}_3)_4(\text{DMF})_2]$, $[\text{CuL}^c(\text{N}_3)\cdot\text{DMF}]$ and $[\text{CuL}^c(\text{N}_3)\text{-CuL}^c(\text{N}_3)]$ with tridentate Schiff base ligands HL^c, derived from condensation of N,N-dimethylethylenediamine with salicylaldehyde, have been synthesized and characterized by IR spectroscopy, X-ray structural analyses and variable temperature magnetic susceptibility measurements by Y.-B. Jiang *et al.* First complex has an alternating phenoxy- and chlorobridged 1D chain structure. Second complex possesses trinuclear units in which the Cu(II) ions are bridged by single *end-on* ($\mu_{1,1}$) azido ligand. Uniformly spaced 1D chains with *end-to end* ($\mu_{1,3}$) azido bridges are found in other two complexes [Scheme 9] [35]. As expected, first two complexes exhibit antiferromagnetic and ferromagnetic exchange, respectively. Other two complexes show unusual ferromagnetic coupling though the 1,3-azido pathways. Theoretical simulations have been carried out in order to evaluate the magnitude of the magnetic exchange and the magnetostructural correlation has been investigated.



Scheme 9. Schematic view of a novel alternating phenoxy- and chlorobridged 1D chain (left) and a trinuclear complex bridged by single *end-on* ($\mu_{1,1}$) azido group of N₂O donor Schiff base ligand (right).

CONCLUSION

The structural, magnetic and catalytic properties of polynuclear Cu(II) complexes of N₂O donor Schiff base ligands with polyatomic anions have been developed to a considerable extent because of their interesting applications in the different fields of chemistry. Among them N₂O donor dinuclear Cu(II) species are of continuing interest mostly because of the occurrence of a pair of copper(II) centers in the active site of the enzyme catechol oxidase. However, recent interest in the magnetic study and weak forces of polynuclear Cu(II) complexes with various bridging groups emerges from their potential use as functional materials. The selection of both N₂O donor and bridging ligands is crucial for the formation of the desired polynuclear complexes with specific bridging modes of the ligands. In particular, the tridentate N₂O donor Schiff base ligands, along with some additional bridging coligands *e.g.* azide, thiocyanate, benzoate, carboxylates, 4,4'-bipyridine, dicyanamide, hydroxido, oxido, nitrite, nitrate *etc.*, favours the formation of multinuclear Cu(II) complexes showing different kind of non-covalent interactions (hydrogen bonding, C-H/ π , cation- π , anion- π and $\pi\cdots\pi$ stacking) and interesting magnetic exchange interactions. Among the several transition metal centers, the exploration of structure, catalytic activity, non-covalent interactions and magnetic properties of polynuclear Cu(II) complexes N₂O donor ligands and polyatomic anions still remain relatively limited. Therefore the synthesis of the new polynuclear Cu(II) complexes with various bridging groups are expected to help in better understanding of the factors that govern the catalytic properties and coupling between the metal centers, which in turn would help to design and produce functional materials.

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