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SYNTHESIS, AND SPECTRAL ANALYSIS OF ZINC (II) MACROCYCLIC COMPLEXES WITH MOLECULAR DOCKING STUDIES

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ABSTRACT

Ten complexes of the general formula $[ZnX^2L^{1-5}]$ have been produced and thoroughly analyzed through elemental profiling, molar electrical conductivity measurements, IR spectroscopy, and Electron spectroscopy for chemical analysis (ESCA) data. In these complexes, X represents Cl or Br, while L corresponds to newly designed macrocyclic Schiff base ligands, derived from the condensation of benzil (2 mmol) and aliphatic diamines, i.e., Ethylenediamine, Propane-1,3-diamine, 1,4-Diaminobutane, Pentane-1,5-diamine, 1,6-Diaminohexane (2 mmol). All of these prepared $[ZnX_2L^{1-5}]$ complexes have been suggested to have an octahedral geometry. To determine the optimal ligand-protein interactions, molecular docking was performed using a single protein and four distinct ligands, such as L^1, L^2, L^3, and L^4 receptors on a molecule.

Keywords: Macrocyclic Schiff Base, Zinc (II) Metal Complexes; Molar Conductivity, IR, And XPS.

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INTRODUCTION

The macrocyclic chemistry of metals, nonmetals, actinides, and lanthanides has grown rapidly during the last three decades as a result of the applications and utility of metal complexes in coordination and bioinorganic chemistry.[1-5] Various macrocyclic ligands, including Robson-type tetraiminodiphenol[6-7], N_4S_2 donor macrocyclic[8-9], polyazamacrocyclic[10-11], saturated and unsaturated polyamines[12-13], porphyrins, and crown ethers, have been discovered in recent decades.[14] In recent decades, several studies have explored macrocyclic Schiff base ligands and their corresponding metal complexes.[15-17] Macrocyclic metal complexes have demonstrated a range of functionality such as RNA cleavage catalysts[18], NMR shift agents[19] and relaxation agents, in magnetic resonance imaging (MRI)[20], environmental importance[21], antitumor[22], anticancerous[23], toxicity against bacterial fungal growth[24], versatile coordination behavior[25-26], photosensitizer[27-28], in photosynthesis and dioxygen transport[29], catalyst[30-31], and pharmacological agent.[32-33] Radiotherapy[34], cancer diagnosis[35]; metal ion separation[36], and tumour lesion detection.[37] The discovery of diverse macrocyclic ligands and metal complexes has significantly contributed to the development of supramolecular chemistry.[38] Six extensive reviews of macrocyclic ligands, their uses, and utility were also published in 2010.[39] Limited research has been carried out on the preparation and comprehensive analysis of zinc (II) metal macrocyclic complexes, including Zn (II) mixed macrocyclic O-N donor complexes and zinc halide compounds of the forms $[ZnLX_2]$ and $[ZnLX]_2$, where L denotes the 1,12,15-triaza-3,4,19,10-dibenzo-5,8-dioxacyclo-heptadecane ligand, according to a review of the literature conducted over the previous three decades on the subject. [Zn (1,4,7,10-tetra-azacyclododecane) $NO_3)ClO_4$, $[Zn_2$(1,4,7,10-tetra-azacyclododecane$)_2$-m-nic] $(ClO_4)_3$, and Zn_2(1,4,7,10-tetra-azacyclododecane$)_2$(m-pic)] are the type Zn(II) macrocyclic complexes[40-41], $[ZnLCl]^{2+}$ complexes where (L= macrocyclic ligand produced by anthracene fragment added through a methylene group to the scorpionands ethylenediamine side chain)[41-42], The $[ZnLCl_2]$ complex consists of a 12-membered N_2O_2 macrocyclic ligand (L), which is synthesized through a 1+1 condensation reaction between various α-diketones, such as benzyl ketone, 1-phenylpropane-1,2-dione, 2,3-butanedione, Pentane-2,3-dione, 2,3-hexanedione, Hexane-3,4-dione with 1,8-diamino-3,6-

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dioxaoctane,[43] Zn (II) macrocyclic complexes with 14,21-membered tetraazamacrocycles,[44] Zn (II) macrocyclic complexes with piperazine moiety[45], $[ZnLX_2]$ Complexes where L is derived from the condensation of benzyl with malonyldihydrazine[46], $[Zn_4(L)Cl_4]$ tetranuclear Zinc macrocyclic complexes[47], Zn(II) complexes of 18-membered tetraimine macrocyclic ligands with chloro, methyl and nitro functional groups[48], $[ZnLX_2]$ (Where L is the tetradentate macrocyclic ligand that is produced by reacting diaminoaleonitrile with doacetyl using a template reaction),[49] $[ZnLX_2]$ (where L=5,5,7,12,12,14-hexa-methyl-1,4,8,11-tetraazacyclotetradeca-7,14-dienium macrocyclic ligand and X = Cl or Br)[50], $[Zn(L)(H_2O)_2].2NO_2$, $[Zn(L)(NO_3)_2]$ (where L= 3,14-dimethyl 2,6,13,17-tetraazatricyclo [14,4, $O^{1.18}$, $O^{7.12}$] decasane macrocyclic ligands)[51] and $[ZnLX_2]$ (where L= macrocyclic ligands generated from 1,4 dihydroaqinoxaline 2,3-dione and various diamines)[52] have been described. This study reports the synthesis of $[ZnLX_2]$ complexes, where L represents macrocyclic ligands derived from benzyl and various aliphatic diamines, and X denotes Cl or Br. The characterization of these complexes has been obtained through elemental profiling, molar electrical conductivity measurements, IR spectroscopy, and Electron spectroscopy for chemical analysis (ESCA) data.

EXPERIMENTAL

Material and Methods

Prior to use, benzil, along with aliphatic diamines such as ethylenediamine, propane-1,3-diamine, 1,4-Diaminobutane, pentane-1,5-diamine, 1,6-Diaminohexanewere purified through distillation, filtration, and drying with suitable reagents. Methanol, ethanol, and ether were also processed similarly before use, employing appropriate agents.[53-54]

General Procedure

It was refluxed for two hours with benzil (2 mmol) in methanol and aliphatic diamines (2 mmol), such as ethylenediamine, propane-1,3-diamine, 1,4-Diaminobutane, pentane-1,5-diamine, and 1,6-Diaminohexane in methanol. The mixture was again refluxed for the same hours by dropwise addition of a solution of ZnX_2 (where X represents Cl or Br) in methanol (1mmol). After filtering the product, it was dried and recrystallized using a mixture of ethanol and ether (9:1). Figure-1 displays the macrocyclic Schiff base ligands that were synthesized along with their zinc (II) metal complexes.

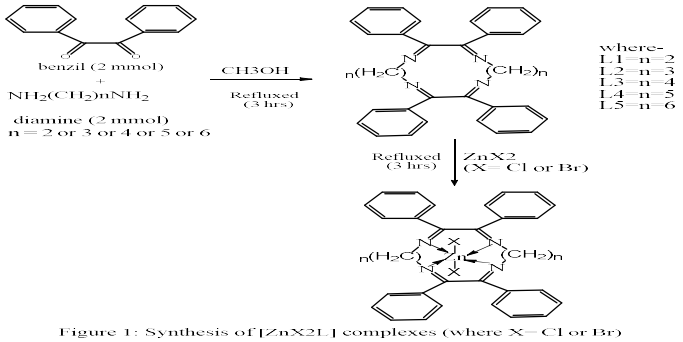

Figure 1: Synthesis of [ZnX2L] complexes (where X=Cl or Br)

Fig.-1: Synthesis of ZnX_2 complexes (where X= Cl or Br)

Detection Method

The melting points of products were determined using a capillary melting point apparatus. The C, H, and N analyses of the products were performed at CDRI in Lucknow. The infrared spectra using CsI/KBr pellets were acquired on a Perkin-Elmer 1000 FT-IR spectrophotometer. Molar conductance was measured in DMF solution at normal temperature using a Digi Sun Digital Conductivity Meter model DL-909. The ESCA data was obtained using the electron spectrometer type of VG Scientific ESCA-3MK II, using Al Kα (1486.6 eV) for the photoexcitation of the product. ESCA Apesoftware[55-56] was used to fit all of the XPS peaks with a Shirley backdrop and a combination of Gaussian and Lorentzian line shapes.

RESULTS AND DISCUSSION

All the synthesized ligands and their zinc metal complexes were white solid. The elemental composition of C, H, and N was determined within 0.5%. The conductivity measurements of these zinc complexes ranged

from 24 to 35 ohm^{-1} cm^{-2} mol^{-1} in a 10^{-3} M DMF solution at room temperature, suggesting their non-electrolytic nature.[57] The IR band for the –CH=N– group has been observed at 1580 cm^{-1} in each of the ligands, L^1, L^2, L^3, L^4, and L^5. However, this infrared band has been moved to a lower wave-number between 1540 and 1560 cm^{-1}, indicating that azomethine coordination is complete. In each metal complex [ZnX_2L^{1-5}], two new IR bands developed at 480-520 cm^{-1} and 420-440 cm^{-1}, associated with γ_{M-N} and $\gamma_{M-X,}$ respectively.[58-59] The binding energy values (eV) for $Zn3p_{1/2}$, N1s, and Xnp (X = Cl or Br) are summarized in Table 1 (Figure-2 to 4). The $Zn3p_{1/2}$ binding energies in the precursor ZnX_2 (X = Cl or Br) were found to be higher compared to those in the synthesized complexes [ZnX_2L^{1-5}], suggesting an increased electron density around the zinc metal ions upon coordination[55-56] (Table-1 and Figure-2). The N1s photoelectron peak in the synthesized complexes [ZnX_2L^{1-5}] exhibited a single symmetric peak shifted towards a higher binding energy relative to the free ligands, suggesting all four nitrogen atoms of each prepared ligand are coordinated with the zinc metal ion, as shown in Table-1, Fig.-4. Further, X2p (where X= Cl or Br) photoelectron peak in these prepared metal complexes [ZnX_2L^{1-5}] have shown higher binding energies than the starting zinc salts i.e., ZnX_2, suggesting halide atoms i.e., Cl or Br, are coordinated to the metal ion in the inner coordinated sphere (Table-1 and Figure-3).[55-56]

Table-1: Binding Energies (eV) of $Zn3p_{1/2}$, Xnp, and N1s in Ligands and [ZnX_2L] Complexes

S. No.	Ligands and Complexes	$Zn3p_{1/12}$	Xnp		N1s	
			Cl2p	$Br3p_{1/12}$	Uncoordinated	coordinated
1.	Ligand L^1	–	–	–	400.2	–
2.	[$ZnCl_2L^1$]	87.4	202.4	–	–	402.4
3.	[$ZnBr_2L^1$]	87.2	–	190.8	–	402.4
4.	Ligand L^2	–	–	–	400.2	–
5.	[$ZnCl_2L^2$]	87.4	202.4	–	–	402.4
6.	[$ZnBr_2L^2$]	87.2	–	190.8	–	402.4
7.	Ligand L^3	–	–	–	400.2	–
8.	[$ZnCl_2L^3$]	87.4	202.4	–	–	402.4
9.	[$ZnBr_2L^3$]	87.2	–	190.8	–	402.4
10.	Ligand L^4	–	–	–	400.2	–
11.	[$ZnCl_2L^4$]	87.4	202.4	–	–	402.4
12.	[$ZnBr_2L^4$]	87.2	–	190.8	–	402.4
13.	Ligand L^5	–	–	–	400.2	–
14.	[$ZnCl_2L^5$]	87.4	202.2	–	–	402.4
15.	[$ZnBr_2L^5$]	87.2	–	190.8	–	402.4
16.	[$ZnCl_2$]	88.2	200.8	–	–	–
17.	[$ZnBr_2$]	88.0	–	189.8	–	–

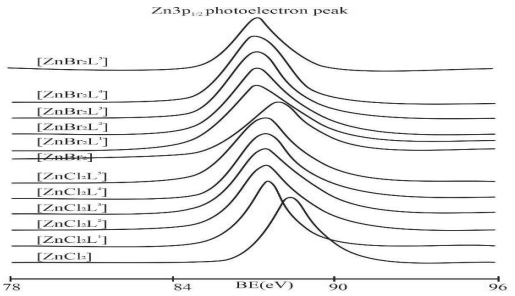

Fig.-2: $Zn3p_{1/2}$ Binding Energies ($eV_)$ in ZnX_2 and ZnX_2L complexes

Molecular Docking

Molecular docking is a computer approach used extensively in structural molecular biology and drug creation. Its major purpose is to forecast the preferred orientation of a ligand when it interacts with a target protein, leading to the formation of a stable molecular complex. The technique is critical for understanding the interactions of physiologically important molecules such as proteins, peptides, and nucleic acids, which

are required for a variety of biochemical processes.[60-62] The selected protein, 1M17, is obtained from the protein data bank's website and used for molecular docking with the ligands (L^{1-4}) using the software Chimaera 1.14 and AutoDock-Vina.

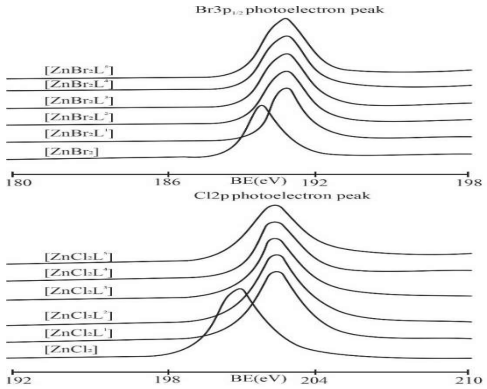

Fig.-3: Xnp Binding Energies (ev) in ZnX_2 and (ZnX_2L) Complexes

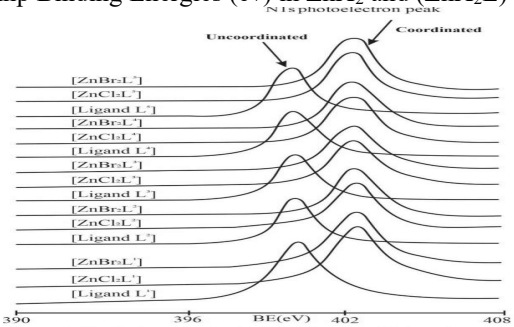

Fig.-4: N1s Binding Energies (eV) in Lignads and ($ZnLX_2$) Complexes

Table-2: Molecular Docking of Ligands (L^{1-4}) and their Hydrogen bonding with Protein 1M17

S. No.	Ligand	Residue Count	Bond Length (Å)	Inhibition Value (μM)	Interaction Energy (kcal/mol)	RMSD Reference
1	L^1	3	2.444	0.21001	-9.1	8.898
2	L^2	3	2.474	0.03876	-10.1	9.794
3	L^3	3	2.024	46.8239	-5.9	7.667
4	L^4	3	2.602	3199.01	-3.4	6.277

The reduced binding energies of the Protein-Ligands interaction demonstrate the ligands' excellent bioactivity. Figure-5 and Table-2 demonstrate the docking results of the ligands (L^{1-4}) with the protein 1M17, as well as the binding energy, bond distance, and inhibition constant. The docking data show that the ligands L^1 and L^2 have a higher binding affinity. L^2 has the highest binding energy of all ligands, at -10.1 kcal/mol.[62]

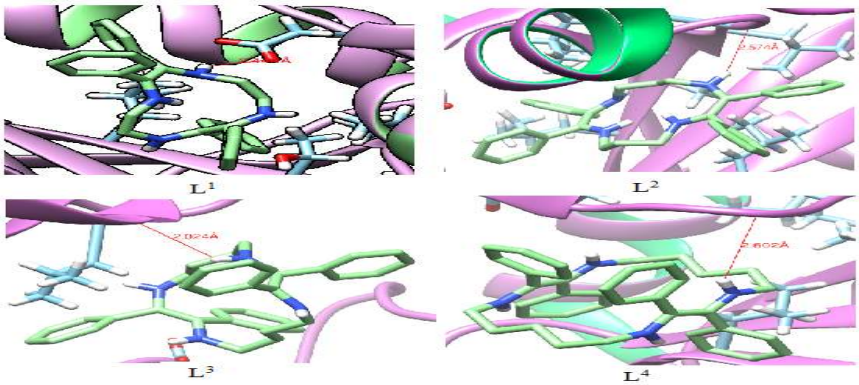

Fig.-5: The Molecular Docking Interaction of Ligand (L^{1-4}) with Protein 1M17

CONCLUSION

On the basis of the observed physicochemical data of $[ZnX_2L^{1-5}]$ complexes, the structure of $[ZnX_2L^{1-5}]$ complexes as shown in figure-1 has been suggested and an octahedral geometry may be determined. Protein 1M17 is linked to anti-cancer agents. Compared to the published data, the ligands L^1 and L^2 may demonstrate anti-cancer action. Molecular nontoxicity was established, and the drug's safety and potency were depicted. The aforementioned research offers a comprehensive foundation for the synthesis of a range of novel bioactive nitrogen-containing derivatives, which play a pivotal role in the pharmaceutical industry.

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CONFLICT OF INTERESTS

The authors declare that there are no conflicts of interest.

AUTHORS CONTRIBUTION

The present work is related to SDG-3: *Good Health and Well-being*. All the authors contributed significantly to this manuscript, participated in reviewing/editing, and approved the final draft for publication. The research profile of the authors can be verified from their ORCID ids, given below:

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