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[Cu(Ibu)$_2$(MEA)$_2$] KOORDINATSION BIRIKMASINING ELEKTRON TUZILISHINI KVANT-KIMYOVIY TADQIQ ETISH QUANTUM-CHEMICAL STUDY OF THE ELECTRONIC STRUCTURE OF THE [Cu(Ibu)$_2$(MEA)$_2$] COORDINATION COMPLEX

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Annotatsiya. Ushbu maqolada [Cu(Ibu)$_2$(MEA)$_2$] tarkibli koordinatsion birikmaning elektron tuzilishi zichlik funksionali nazariyasi (DFT) asosidagi kvant-kimyoviy hisoblash usullari yordamida tadqiq etildi. Sintez qilingan kompleks birikmaning barqarorligi, koordinatsion xususiyatlari va elektron tuzilishini baholash maqsadida molekulyar geometriyasi optimallashtirildi, HOMO–LUMO molekulyar orbitallari, Mulliken bo‘yicha atom zaryadlarining taqsimlanishi hamda kimyoviy bog‘larning geometrik parametrlari tahlil qilindi.

Kalit so‘zlar: Cu(II) kompleksi, ibuprofen, monoetanolamin, DFT hisoblashlari, HOMO–LUMO, elektron tuzilish, koordinatsion birikma.

Аннотация. В данной статье исследована электронная структура координационного соединения состава [Cu(Ibu)$_2$(MEA)$_2$] с использованием квантово-химических расчётов на основе теории функционала плотности (DFT). С целью оценки устойчивости, координационных особенностей и электронных свойств синтезированного комплексного соединения проведены оптимизация молекулярной геометрии, анализ молекулярных орбиталей HOMO–LUMO, распределения атомных зарядов по Малликену, а также геометрических параметров химических связей.

Ключевые слова: комплекс Cu(II), ибупрофен, моноэтаноламин, DFT-расчёты, HOMO–LUMO, электронная структура, координационное соединение.

Abstract. The electronic structure of the [Cu(Ibu)$_2$(MEA)$_2$] coordination complex was investigated using DFT-based quantum-chemical calculations. Geometry optimization, HOMO–LUMO analysis, Mulliken charge distribution, and bond parameters were studied to

evaluate the stability, coordination behavior, and electronic properties of the synthesized complex compound.

Keywords: Cu(II) complex, ibuprofen, monoethanolamine, DFT calculations, HOMO–LUMO, electronic structure, coordination compound.

INTRODUCTION. Metal complexes based on biologically active ligands have attracted considerable attention in coordination and medicinal chemistry due to their diverse structural features and biological properties. In particular, non-steroidal anti-inflammatory drugs (NSAIDs) such as ibuprofen are widely used as ligands because their carboxylate groups can coordinate to metal ions through different coordination modes (Núñez et al., 2014) [1]. Several studies have reported the synthesis and characterization of ibuprofen-based transition metal complexes. Abu Ali et al. (2016) synthesized Zn(II) ibuprofen complexes with nitrogen-donor ligands and investigated their structural and antimicrobial properties. The authors demonstrated that complex formation significantly influences the coordination behavior and biological activity of the ligand system [2]. Copper(II) ibuprofen complexes containing N-donor ligands were also investigated by Kumar et al. (2017). Their study included FT-IR, UV–Vis, EPR, and single-crystal X-ray analyses, revealing monomeric coordination structures and enhanced cytotoxic activity compared to the free ligand [3]. In addition, Bamigboye et al. (2021) reported mixed-ligand metal complexes of ibuprofen and salicylic acid, showing improved antimicrobial activity after coordination with transition metal ions [4]. Despite numerous studies on ibuprofen-based metal complexes, the electronic structure and quantum-chemical properties of mixed-ligand Cu(II) complexes containing monoethanolamine remain insufficiently investigated. Therefore, the quantum-chemical study of the $[Cu(Ibu)_2(MEA)_2]$ coordination complex is of significant scientific interest.

Literature Review. In recent years, considerable attention has been devoted to the synthesis of transition metal complexes derived from biologically active organic compounds, the investigation of their molecular and electronic structures, and the evaluation of their physicochemical properties using quantum-chemical methods. In particular, the coordination behavior of ibuprofen, a nonsteroidal anti-inflammatory drug (NSAID), toward metal ions, as well as the relationships among molecular geometry, electron density distribution, and biological activity, has become an important research area in coordination chemistry. The carboxyl group of ibuprofen can undergo deprotonation to form the ibuprofenate anion, which coordinates to metal ions through oxygen donor atoms, resulting in coordination compounds with different compositions and geometrical arrangements. The electronic configuration of the central metal ion, the donor properties of auxiliary ligands, and the resulting coordination environment significantly influence the structural characteristics and stability of these complexes [5-8].

Kumar et al. synthesized and characterized four mononuclear copper(II) complexes containing ibuprofenate anions and nitrogen-donor organic ligands, investigating their crystal structures, spectroscopic properties, and cytotoxic activities [5]. The study employed β-picoline, γ-picoline, pyrrolidine, and unsymmetrical dimethylethylenediamine as auxiliary

ligands. The synthesized compounds were characterized using elemental analysis, Fourier-transform infrared (FTIR) spectroscopy, UV-Vis spectroscopy, electron paramagnetic resonance (EPR) spectroscopy, thermogravimetric analysis, and single-crystal X-ray diffraction. The crystallographic results demonstrated that the incorporation of additional nitrogen-donor ligands could promote the formation of mononuclear complexes instead of the binuclear structures characteristic of certain copper(II) ibuprofenate systems. These findings emphasize the important role of auxiliary N-donor ligands in modifying the coordination geometry of the Cu(II) center. Consequently, this study provides a useful structural foundation for understanding metal-ligand interactions and coordination behavior in the $[Cu(Ibu)_2(MEA)_2]$ complex. Malis et al. investigated copper(II) coordination compounds containing nonsteroidal anti-inflammatory drugs, focusing on their structural characterization and in vitro and in silico biological profiles [6]. Their research highlighted the importance of considering not only the pharmacological properties of the organic ligands but also the coordination environment and molecular architecture of the corresponding metal complexes when evaluating their biological properties. Metal-ligand interactions are essential factors influencing molecular geometry and electronic characteristics. Therefore, the theoretical investigation of the mixed-ligand copper(II) complex containing ibuprofenate and monoethanolamine is relevant for understanding how coordination affects the electronic structure of the resulting compound.

In another study, the coordination behavior of ibuprofen toward Zn(II) and Cu(II) ions was investigated through the synthesis and characterization of metal–ibuprofenate complexes in both the solid state and solution [7]. Crystallographic methods, potentiometric measurements, and nuclear magnetic resonance (NMR) spectroscopy were employed to examine their structural and physicochemical characteristics. The findings indicated that ibuprofenate ligands may form coordination compounds with different structural arrangements depending on the nature of the metal center. In particular, the coordination preferences of the metal ion and the binding modes of the carboxylate group play significant roles in determining the molecular or polymeric structures of the resulting complexes. These observations provide a basis for comparing the optimized geometry of $[Cu(Ibu)_2(MEA)_2]$, particularly the Cu-O bond lengths and the coordination characteristics of the ibuprofenate fragment, with those reported for related metal–ibuprofenate complexes.

Abuhijleh conducted one of the earlier investigations of mononuclear and binuclear copper(II) complexes derived from ibuprofen, examining their spectroscopic characteristics and catalytic properties [8]. Mononuclear complexes containing imidazole and 2-methylimidazole were reported to exhibit CuO_2N_2 coordination environments, whereas other complexes involving metronidazole and caffeine displayed binuclear structures. These findings demonstrated that the coordination behavior of the ibuprofenate ligand is closely associated with the nature of the additional nitrogen-donor ligands. Such structural diversity provides further justification for investigating copper(II) complexes containing ligands with both nitrogen and oxygen donor atoms, including monoethanolamine, particularly with respect to their coordination geometry and electronic properties.

Density functional theory (DFT) represents an important theoretical approach for the detailed investigation of the electronic structures of coordination compounds. Tabrizi et al. performed experimental and quantum-chemical investigations of a copper(II) complex containing lidocaine- and ibuprofen amide–phenanthroline-based ligands [9]. Molecular geometry, atomic charge distribution, electrostatic potential, and electronic structure parameters were evaluated using DFT calculations. In particular, molecular electrostatic potential maps and atomic charge analysis provided information about electron-rich regions associated with nitrogen and oxygen atoms, contributing to a better understanding of their roles in coordination interactions. This computational approach provides a methodological basis for evaluating the electronic characteristics of Cu–O and Cu–N bonds, electron density redistribution, and donor–acceptor interactions in the $[Cu(Ibu)_2(MEA)_2]$ coordination complex. Liu and Gao investigated the molecular geometry and vibrational spectra of ibuprofen using several DFT functionals and basis sets [10]. The authors employed the mPW1PW91, B3PW91, B3LYP, HCTH, and LSDA functionals and compared the calculated results with experimental data, demonstrating the importance of selecting an appropriate computational method for the accurate description of molecular geometry and vibrational frequencies. Particular attention was paid to bond lengths, bond angles, and theoretical infrared spectra of the ibuprofen molecule. These results are relevant for comparing the structural characteristics of free ibuprofen with those of the ibuprofenate fragment incorporated into a coordination compound. Furthermore, the comparison of theoretical and experimental spectroscopic parameters may provide additional evidence supporting the proposed molecular structure of the $[Cu(Ibu)_2(MEA)_2]$ complex.

Tasić et al. investigated the corrosion-inhibiting properties of ibuprofen on copper surfaces using experimental and theoretical approaches [11]. Their study employed electronic structure parameters, including molecular orbital energies, to explain the interactions between ibuprofen molecules and the metal surface. The energies of the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO), the corresponding energy gap, and related electronic descriptors are useful for evaluating the electron-donating and electron-accepting characteristics of molecular systems. These parameters may also be applied to investigate the frontier molecular orbitals, spatial electron density distribution, and potential electronic transitions of the $[Cu(Ibu)_2(MEA)_2]$ coordination complex. However, since the cited study primarily addressed adsorption processes on a metallic surface, its results should not be directly equated with the thermodynamic stability of discrete molecular coordination compounds. Another investigation focused on metal complexes derived from an ibuprofen-based ligand and combined physicochemical characterization with DFT modeling, molecular docking, in vitro anti-inflammatory evaluation, and DNA interaction studies [12]. Such an integrated experimental and theoretical approach provides opportunities to examine possible relationships between the electronic structure of coordination compounds and their physicochemical and biological properties. In particular, the combined interpretation of optimized molecular geometries, electronic descriptors, and experimental observations can

contribute to a more comprehensive understanding of structure–property relationships in metal coordination compounds.

The coordination behavior of monoethanolamine in metal complexes is also of considerable scientific interest. Petrov et al. synthesized water-soluble copper(II) coordination compounds containing fumarate and succinate anions together with monoethanolamine and investigated their crystal structures [13]. The authors reported complexes with the compositions $[Cu(Suc)\cdot 2MEA]\cdot H_2O$ and $[Cu(Fum)\cdot 2MEA]\cdot H_2O$. Single-crystal X-ray diffraction analysis revealed polymeric structures consisting of zigzag chains. These findings highlight the ability of monoethanolamine to participate in the formation of copper(II) coordination compounds. The presence of amino and hydroxyl functional groups in monoethanolamine provides potential nitrogen and oxygen donor sites, allowing different coordination modes depending on the chemical environment. Accordingly, investigation of the $[Cu(Ibu)_2(MEA)_2]$ complex requires particular attention to the donor atoms of the MEA ligands, the geometrical parameters of Cu-N and Cu-O bonds, and the redistribution of electron density upon coordination. Kamariah et al. investigated the solvation and coordination behavior of Cu(II), Zn(II), and Pb(II) ions in monoethanolamine-containing solutions [14]. Their findings provided insight into the possible participation of amino and hydroxyl functional groups in metal–ligand interactions. The study considered theoretical modeling together with solution-phase chemical processes to evaluate the coordination environment of metal–monoethanolamine systems. This approach is relevant for understanding the electronic characteristics of ligand donor atoms, the coordination preferences of metal centers, and the influence of solvation on metal–ligand interactions. Nevertheless, differences between solution-phase coordination environments and the structures of isolated molecular complexes should be considered when interpreting quantum-chemical results. Overall, the reviewed literature demonstrates that ibuprofenate anions can form structurally diverse copper(II) coordination compounds and that the incorporation of additional N-donor or N,O-donor ligands may substantially influence the coordination geometry and electronic characteristics of the metal center [5-14]. Furthermore, DFT-based calculations provide an effective theoretical framework for describing molecular geometries, electronic distributions, frontier molecular orbitals, and metal–ligand bonding characteristics. Nevertheless, the reviewed studies do not provide a comprehensive quantum-chemical characterization specifically focused on the electronic structure, HOMO–LUMO properties, and atomic charge distribution of the mixed-ligand $[Cu(Ibu)_2(MEA)_2]$ coordination complex. This observation indicates the relevance of conducting a dedicated theoretical investigation of this compound.

Therefore, the primary objective of the present study is to investigate the molecular and electronic structures of the $[Cu(Ibu)_2(MEA)_2]$ coordination complex using DFT-based quantum-chemical calculations. Particular attention is devoted to the optimization of molecular geometry, analysis of HOMO and LUMO orbital energies, evaluation of Mulliken atomic charge distribution, and determination of the geometrical parameters of Cu-O and Cu-N coordination bonds. The calculated results are expected to provide insights into the

electronic characteristics of the complex, metal-ligand interactions, and the distinctive features of its coordination environment.

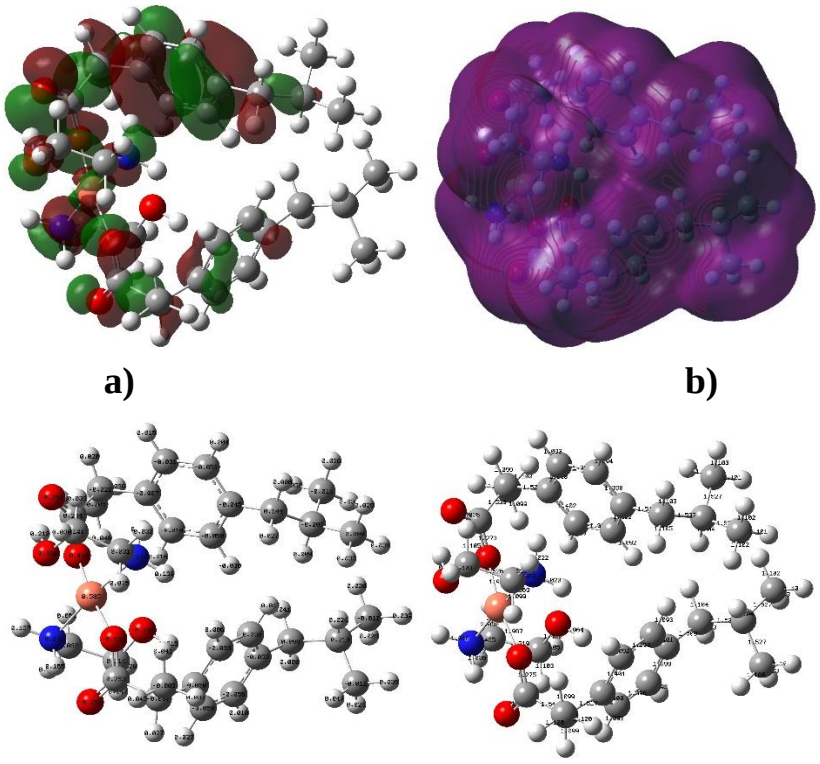
Research methodology. The electronic structure and quantum-chemical properties of the $[Cu(Ibu)_2(MEA)_2]$ coordination complex, where Ibu represents the ibuprofenate ligand and MEA denotes monoethanolamine, were investigated using density functional theory (DFT). All quantum-chemical calculations were performed using the Gaussian 16 software package. The molecular geometry of the complex was fully optimized in the gas phase without imposing symmetry constraints, employing the Coulomb-attenuating method based on the range-separated hybrid exchange–correlation functional CAM-B3LYP. This functional incorporates a distance-dependent treatment of Hartree–Fock exchange and provides an improved description of long-range exchange interactions, making it suitable for investigating the electronic characteristics of molecular systems.

Geometry optimization was performed to determine the equilibrium molecular configuration of the $[Cu(Ibu)_2(MEA)_2]$ complex and to evaluate the structural parameters associated with the coordination environment of the Cu(II) center. Particular attention was paid to the Cu–O and Cu–N coordination bonds, as well as the bond lengths and bond angles of the ibuprofenate and monoethanolamine ligands. Vibrational frequency calculations were performed on the optimized molecular structure to characterize the corresponding stationary point on the potential energy surface. The absence of imaginary vibrational frequencies was used as the criterion for confirming that the optimized geometry represented a local energy minimum. Natural Bond Orbital (NBO) analysis was carried out using the NBO 7 program interfaced with Gaussian 16 to investigate the electronic structure and bonding characteristics of the coordination complex. Natural atomic charges and orbital hybridization parameters were evaluated to characterize the electron density distribution and the electronic environment of the central Cu(II) ion and coordinating donor atoms. Donor–acceptor interactions were examined using second-order perturbation theory to assess electron delocalization and orbital interactions associated with metal–ligand coordination bonding. Particular attention was given to the electronic interactions involving the oxygen atoms of the ibuprofenate ligands and the donor atoms of monoethanolamine.

Frontier molecular orbital analysis was performed to investigate the electronic characteristics of the $[Cu(Ibu)_2(MEA)_2]$ complex. The energies of the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) were extracted from the converged DFT wavefunction, and the corresponding frontier orbital energy gap (ΔE) was calculated as the difference between the LUMO and HOMO energies. These parameters were considered in evaluating the frontier electronic structure, electron-donating and electron-accepting tendencies, and potential chemical reactivity of the coordination compound.The calculated geometrical parameters, vibrational characteristics, natural atomic charges, orbital hybridization features, donor–acceptor interactions, and frontier molecular orbital energies were analyzed collectively to provide a comprehensive theoretical description of the electronic structure and coordination bonding in the $[Cu(Ibu)_2(MEA)_2]$ complex. The results were interpreted in relation to the electronic interactions between the

Cu(II) center and the ibuprofenate and monoethanolamine ligands, with particular emphasis on the redistribution of electron density accompanying coordination.

Analysis and results. All quantum chemical calculations were carried out using the Gaussian 16 software package. The molecular geometry was fully optimized in the gas phase without symmetry constraints using the range-separated hybrid functional CAM-B3LYP, which incorporates the Coulomb-attenuating method to improve the description of long-range exchange interactions. Frequency calculations were performed on the optimized structure to confirm that the geometry corresponds to a true energy minimum on the potential energy surface (absence of imaginary frequencies). Natural bond orbital (NBO) analysis was performed using the NBO 7 program interfaced with Gaussian 16 to obtain natural atomic charges, orbital hybridizations, and donor-acceptor interactions via second-order perturbation theory. Frontier molecular orbitals (HOMO, LUMO) and their energy eigenvalues were extracted from the converged DFT wavefunction.



a) **b)**

c) **d)**

a — HOMO – LUMO orbitals of the $[Cu(Ibu)_2(MEA)_2]$ complex;
b — electron density distribution of the $[Cu(Ibu)_2(MEA)_2]$ complex;
c — Mulliken charge distribution;
d — bond lengths.

Frequency calculations were performed for the optimized structures to confirm that the geometries correspond to true minima on the potential energy surface, indicated by the absence of imaginary frequencies. Quantum-chemical analysis was carried out for the two synthesized complex compounds, and the obtained results are presented in Table 1.

Table 1

Results of quantum-chemical calculations of copper complex compounds

Coordination compound		Quantum-chemical parameters	Values of Quantum-

			Chemical Parameters
[Cu(Ibu)₂(H₂O)₂]	1	EHOMO, a.u	−0,31427 eV
	2	ELUMO, a.u	−0,04897 eV
	3	|ΔE|= EHOMO - ELUMO, (a.u)	0,26530 eV
	4	Ionization potential, I = -EHOMO, (a.u)	0,31427eV
	5	Electron affinity, A = - ELUMO, (a.u)	0,04897 eV
	6	Electronegativity □ = (I + A)/2 (a.u)	0,18162 eV
	7	Dipole moment, μ (Debay)	3,819548
[Cu(Ibu)₂(MEA)₂]	1	EHOMO, a.u	-0,30724 eV
	2	ELUMO, a.u	-0,01430 eV
	3	|ΔE|= EHOMO - ELUMO, (a.u)	0,29294 eV
	4	Ionization potential, I = -EHOMO, (a.u)	0,30724 eV
	5	Electron affinity, A = - ELUMO, (a.u)	0,01430 eV
	6	Electronegativity □ = (I + A)/2 (a.u)	0.16077 eV
	7	Dipole moment, μ (Debay)	4,824952

Conclusion. The electronic structure of the [Cu(Ibu)₂(MEA)₂] coordination complex was investigated using quantum-chemical calculations. The obtained computational results confirmed that the optimized molecular structure is consistent with the experimentally synthesized complex and its proposed composition. The frontier molecular orbital analysis revealed that the HOMO and LUMO energy values are −0.30724 and −0.01430, respectively, indicating the electronic stability and reactivity of the complex. In addition, the calculated dipole moment was found to be 4.825 D, reflecting the polar nature of the coordination compound.

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