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EFFECT OF Cu(II) COORDINATION ON THE REDOX BEHAVIOUR OF CURCUMINOIDS: A CYCLIC VOLTAMMETRIC INVESTIGATION

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ABSTRACT
The electrochemical behaviour of a series of curcuminoids and their Cu(II) complexes was investigated using cyclic voltammetry to evaluate the influence of metal coordination and substituent effects on electron-transfer characteristics. Cyclic voltammetric measurements were carried out in a methanolic medium containing tetrabutylammonium iodide as the supporting electrolyte using a glassy carbon electrode at different scan rates. The free curcuminoid ligands exhibited a single, well-defined reversible redox couple in the positive potential region with ΔE_p values ranging from 0.05–0.07 V and I_{pa}/I_{pc} ratios close to unity, indicating diffusion-controlled reversible electron transfer. Upon coordination with Cu(II), noticeable changes in the redox potentials were observed, indicating modification of the electronic environment through metal–ligand charge delocalisation. In addition to the ligand-centered process, the Cu(II) complexes displayed an extra redox couple assigned to the Cu(II)/Cu(I) transition, characterized by quasi-reversible behaviour with ΔE_p values of 0.27–0.34 V and I_{pa}/I_{pc} ratios greater than unity. The results further indicate that electron-donating substituents such as hydroxyl and methoxy groups influence the electrochemical response of the curcuminoid framework. The study provides important insight into the role of metal coordination in tuning the redox properties of curcuminoids, which is relevant to their biological and catalytic applications. The study also supports Sustainable Development Goal 3 (Good Health and Well-being) through the development of biologically relevant redox-active coordination compounds.
Keywords: Cu(II) Complexes, Curcuminoids, Cyclic Voltammetry, Electrochemical Reversibility, Redox Behaviour.

INTRODUCTION

Curcuminoids (1,7-diarylhepta-1,6-diene-3,5-diones) are naturally occurring polyphenolic compounds isolated from turmeric (*Curcuma longa*) and are widely recognized for their antioxidant, anti-inflammatory, antimicrobial, and anticancer properties.[1-3] These biologically active compounds possess a conjugated β-diketone framework capable of keto–enol tautomerism, which plays a significant role in their chemical reactivity and electron-transfer behaviour.[4] Owing to the presence of phenolic and β-dicarbonyl donor sites, curcuminoids readily coordinate with transition-metal ions to form stable chelate complexes with modified physicochemical and biological properties.[2-6] Among transition-metal complexes, Cu(II) complexes of curcuminoids have attracted considerable attention because copper is a biologically relevant redox-active metal ion capable of participating in electron-transfer reactions under physiological conditions.[2-5] Coordination of Cu(II) with curcuminoid ligands often enhances antioxidant efficiency, improves chemical stability, and alters the electronic distribution within the ligand framework. These changes significantly influence the redox characteristics of the complexes, which are associated with their biological and catalytic activities. In this context, understanding the electrochemical behaviour of curcuminoids and their metal complexes is essential for correlating structural features with functional properties. Cyclic voltammetry (CV) is one of the most widely used electroanalytical techniques for investigating the redox behaviour of organic molecules and coordination compounds.[7-10] The technique provides valuable information regarding electron-transfer processes, electrochemical reversibility,

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diffusion control, and kinetic parameters through the analysis of anodic and cathodic peak characteristics.[9,11] Recent electrochemical studies on biologically active metal complexes have demonstrated that substituent effects and metal coordination strongly influence oxidation–reduction potentials and charge-transfer properties.[7,8,12] However, despite extensive reports on the synthesis and biological evaluation of curcuminoid metal complexes, detailed electrochemical investigations concerning the influence of Cu(II) coordination on the redox behaviour of curcuminoids remain limited.[5,7,8,12] The present study aims to investigate the cyclic voltammetric behaviour of selected curcuminoids and their Cu(II) complexes at different scan rates in methanolic medium using tetrabutylammonium iodide as the supporting electrolyte. Particular emphasis is given to evaluating the effect of metal coordination and aromatic substituents on electrochemical reversibility and redox potential shifts. The study aims to provide electrochemical characterisation of curcuminoid–Cu(II) systems and their electron-transfer properties. Since redox-active Cu(II) complexes of curcuminoids are associated with antioxidant and therapeutic applications, the work also contributes to Sustainable Development Goal-3 (Good Health and Well-being) by supporting the development of biologically relevant coordination compounds with potential medicinal importance.

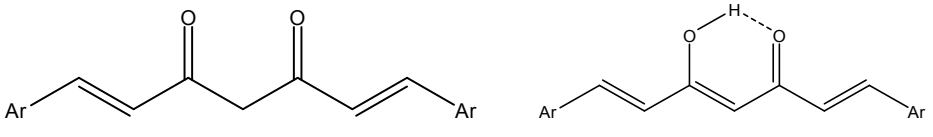
EXPERIMENTAL

Materials and Instrumentation

All chemicals and solvents used in the present study were of analytical reagent grade and used without further purification. Cyclic voltammetric measurements were carried out using a Biologic SP-50 electrochemical analyser equipped with a conventional three-electrode system consisting of a glassy carbon working electrode, a platinum wire auxiliary electrode, and an Ag/AgCl reference electrode. Methanol containing 0.1 M tetrabutylammonium iodide (TBAI) was employed as the supporting electrolyte. Prior to each experiment, the working electrode was polished carefully to obtain a clean and reproducible surface. The electrochemical measurements were performed at room temperature under a nitrogen atmosphere to minimize interference from dissolved oxygen. The potential was scanned in the range from –3.0 to +3.0 V at scan rates of 20, 50, and 100 mV s^{-1}.

Synthesis of Curcuminoids and their Cu(II) Complexes

The curcuminoid ligands (HL^1–HL^4) and their Cu(II) complexes were synthesized according to reported methods.[2-6] The ligands were prepared by condensation of substituted aromatic aldehydes such as benzaldehyde, 3,4-dihydroxybenzaldehyde, 3,4-dimethoxybenzaldehyde, and vanillin with the acetylacetone–boron complex in the presence of n-butylamine and tri(sec-butyl)borate, followed by acid hydrolysis. The corresponding Cu(II) complexes were synthesized by refluxing the ligands with Copper(II) acetate in ethanol. The obtained coloured complexes were filtered, washed thoroughly with ethanol, and dried under vacuum before use. The structures of the synthesized curcuminoids and their Cu(II) complexes are presented in Figs.-1 and 2, respectively.



Keto form (Enol form)

Fig.-1: Structure of Curcuminoids

Ar = Phenyl (HL^1); 3,4-dihydroxyphenyl (HL^2); 3,4-dimethoxyphenyl (HL^3); 4-hydroxy-3-methoxyphenyl (HL^4)

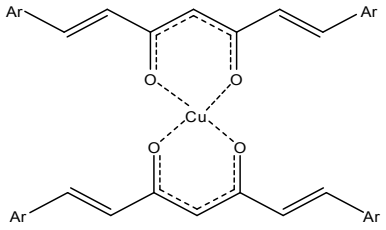


Fig.-2: Structure of Cu(II) Complexes of Curcuminoids [CuL_2]

Cyclic Voltammetric Measurements

For electrochemical analysis, 0.01 M solutions of each ligand and Cu(II) complex were prepared in methanol containing 0.1 M TBAI as the supporting electrolyte. The solutions were purged with nitrogen gas for several minutes before measurements to remove dissolved oxygen. Cyclic voltammograms were recorded at scan rates of 20, 50, and 100 mV s^{-1} to evaluate the effect of scan rate on the redox behaviour of the compounds. The electrochemical parameters including anodic peak potential (E_{pa}), cathodic peak potential (E_{pc}), peak separation (ΔE_p), anodic peak current (I_{pa}), cathodic peak current (I_{pc}), and current ratio (I_{pa}/I_{pc}) were determined directly from the recorded voltammograms. To avoid redundancy and improve clarity, representative cyclic voltammograms are presented in the figures, while the detailed electrochemical parameters are summarized separately in the tables.

RESULTS AND DISCUSSION

The cyclic voltammograms of the curcuminoid ligands (HL^1–HL^4) were recorded at scan rates of 20, 50, and 100 mV s^{-1} in methanolic solution containing 0.1 M TBAI as the supporting electrolyte. Representative voltammograms are shown in Figs.-3 to 6, while the corresponding electrochemical parameters are summarised in Table-1. The voltammetric profiles of all four curcuminoids exhibited a single, well-defined redox couple in the positive potential region. The anodic and cathodic peak currents are nearly symmetrical, and the peak separations (ΔE_p = 0.05–0.07 V) remain relatively small, indicating facile electron transfer at the electrode surface.[9,11] The I_{pa}/I_{pc} ratios are close to unity (0.88–0.97), which is characteristic of a reversible one-electron electrochemical process controlled predominantly by diffusion.[9,11] These observations are consistent with previously reported electrochemical behaviour of conjugated diketone systems and biologically active polyphenolic compounds.[7,8,12] The electrochemical responses remained nearly unchanged with increasing scan rate, suggesting that the redox processes are electrochemically stable and not significantly affected by kinetic complications.[11,12] The reversible behaviour observed for the ligands may be attributed to the delocalized π-electron system present in the curcuminoid framework, which facilitates efficient charge transfer during oxidation and reduction processes.[4] The keto–enol tautomeric equilibrium of curcuminoids also contributes to stabilisation of the intermediate oxidised species through conjugation and intramolecular hydrogen bonding.[1,4] Substituent effects on the aromatic ring considerably influenced the redox potentials of the ligands. The oxidation potentials followed the order $HL^2 > HL^4 > HL^3 > HL^1$, indicating that hydroxyl and methoxy substituents alter the electron density distribution within the conjugated system. Electron-donating substituents influence the electron density distribution through resonance interactions, thereby affecting the redox potentials.[2,5] Similar substituent-dependent electrochemical effects have been reported for other biologically active aromatic systems containing phenolic donor groups.[7,8] The cyclic voltammograms of the Cu(II) complexes of curcuminoids are presented in Figs.-7 to-0, and the electrochemical parameters are summarized in Tables-2 and 3. Upon coordination with Cu(II), significant changes in the electrochemical behaviour of the ligands were observed. The voltammograms exhibited two distinct redox couples, corresponding to ligand-centred and metal-centred electron-transfer processes. The ligand-centred couple appeared in the positive potential region (1.83–2.13 V) with ΔE_p values of 0.04–0.07 V. Although these values are comparable to those of the free ligands, the I_{pa}/I_{pc} ratios decreased slightly (0.35–0.45), indicating reversible to quasi-reversible behaviour due to the influence of metal coordination on electron-transfer kinetics.[9,11] Coordination with Cu(II) produced noticeable changes in the ligand-centred redox potentials, indicating modification of the electronic environment through metal–ligand charge delocalization.[2-5] Coordination of the Cu(II) ion withdraws electron density from the ligand framework and modifies the electronic environment of the conjugated β-diketone system, thereby influencing the oxidation and reduction characteristics.[5] Such changes in electrochemical behaviour upon metal coordination are commonly observed in transition-metal complexes possessing redox-active ligands.[7,8] An additional redox couple appeared at lower potentials in all Cu(II) complexes, which was assigned to the Cu(II)/Cu(I) redox system. The anodic peak potentials were observed in the range 1.05–1.15 V, while the cathodic peak potentials appeared between 0.76–0.84 V. The corresponding ΔE_p values (0.27–0.34 V) were considerably larger than those of the ligand-centred processes, and the I_{pa}/I_{pc} ratios were greater than unity (1.37–1.44), indicating quasi-reversible electron-transfer behaviour.[9,11] The larger

peak separation suggests slower electron-transfer kinetics and partial structural reorganization during the Cu(II)/Cu(I) conversion process.[8,12] Similar quasi-reversible Cu(II)/Cu(I) transitions have been reported for copper complexes containing oxygen donor ligands and conjugated coordination environments.[7,8] The scan-rate-dependent voltammetric behaviour further supports diffusion-controlled electron transfer in both ligand-centred and metal-centred redox processes.

Table-1: Cyclic Voltammetric Data of Curcuminoid Ligands

HL	Scan rate	E_{pc}	E_{pa}	ΔE_p	I_{pc}	I_{pa}	I_{pa}/I_{pc}
HL^1	20	1.83	1.90	0.07	0.41	0.38	0.92
	50	1.87	1.92	0.05	0.43	0.41	0.95
	100	1.89	1.94	0.05	0.45	0.44	0.97
HL^2	20	1.99	2.06	0.07	0.31	0.28	0.89
	50	2.01	2.07	0.06	0.32	0.29	0.90
	100	2.03	2.09	0.06	0.34	0.31	0.91
HL^3	20	1.87	1.94	0.07	0.38	0.35	0.92
	50	1.88	1.94	0.06	0.40	0.37	0.92
	100	1.92	1.98	0.06	0.42	0.39	0.92
HL^4	20	1.93	1.97	0.06	0.42	0.39	0.92
	50	1.96	2.02	0.06	0.45	0.40	0.88
	100	1.98	2.03	0.05	0.46	0.41	0.91

Table-2: Cyclic Voltammetric Data for the Ligand-Centered Reversible Redox Process of Cu(II) Complexes of Curcuminoids

Complex	Scan rate	E_{pc}	E_{pa}	ΔE_p	I_{pc}	I_{pa}	I_{pa}/I_{pc}
$[Cu(L^1)_2]$	20	1.77	1.83	0.06	0.62	0.23	0.37
	50	1.83	1.87	0.04	0.67	0.25	0.37
	100	1.91	1.97	0.06	0.71	0.29	0.41
$[Cu(L^2)_2]$	20	1.98	2.03	0.05	0.42	0.17	0.40
	50	2.05	2.09	0.04	0.45	0.19	0.42
	100	2.08	2.13	0.05	0.49	0.22	0.44
$[Cu(L^3)_2]$	20	1.95	2.01	0.06	0.42	0.15	0.35
	50	1.97	2.02	0.05	0.45	0.17	0.37
	100	2.00	2.05	0.05	0.47	0.19	0.40
$[Cu(L^4)_2]$	20	1.99	2.06	0.07	0.52	0.21	0.40
	50	2.01	2.07	0.06	0.53	0.23	0.43
	100	2.03	2.09	0.06	0.55	0.25	0.45

Table-3: Cyclic voltammetric Data for the Cu(II)/Cu(I) Quasi-Reversible Redox Process of Cu(II) Complexes of Curcuminoids

Complex	Scan rate	E_{pc}	E_{pa}	ΔE_p	I_{pc}	I_{pa}	I_{pa}/I_{pc}
$[Cu(L^1)_2]$	20	0.78	1.05	0.27	0.52	0.73	1.37
	50	0.81	1.09	0.28	0.51	0.72	1.38
	100	0.84	1.12	0.28	0.50	0.71	1.42
$[Cu(L^2)_2]$	20	0.76	1.08	0.32	0.53	0.73	1.37
	50	0.79	1.12	0.33	0.52	0.74	1.42
	100	0.82	1.15	0.33	0.52	0.75	1.44
$[Cu(L^3)_2]$	20	0.77	1.09	0.32	0.51	0.72	1.41
	50	0.81	1.11	0.30	0.50	0.72	1.44
	100	0.84	1.13	0.29	0.51	0.73	1.43
$[Cu(L^4)_2]$	20	0.77	1.09	0.32	0.52	0.75	1.44
	50	0.78	1.12	0.34	0.54	0.78	1.44
	100	0.81	1.14	0.33	0.54	0.79	1.44

The gradual increase in peak currents with increasing scan rate confirms the electroactive nature of the complexes and the stability of the redox species under the experimental conditions.[9,11] The combined electrochemical observations clearly demonstrate that both substituent effects and Cu(II) coordination play decisive roles in controlling the redox characteristics of curcuminoids.

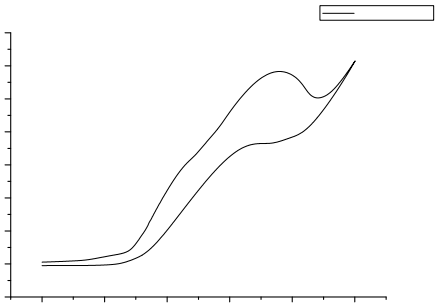


Fig.-3: Cyclic Voltammograms of HL^1

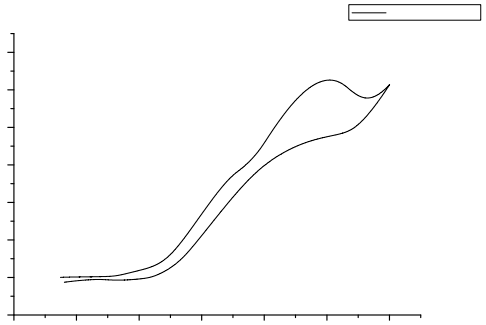


Fig.-4: Cyclic Voltammograms of HL^2

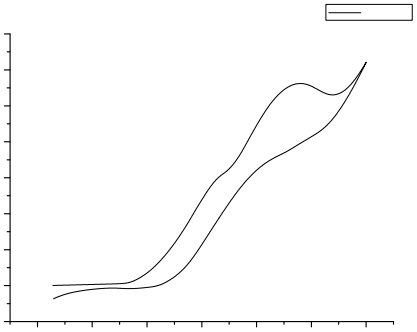


Fig.-5: Cyclic Voltammograms of HL^3

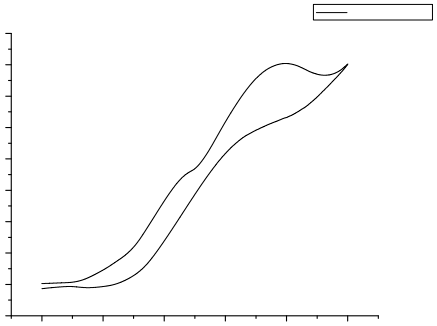


Fig.-6: Cyclic Voltammograms of HL^4

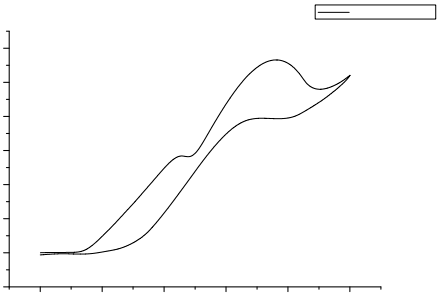


Fig.-7: Cyclic Voltammograms of $[Cu(L^1)_2]$

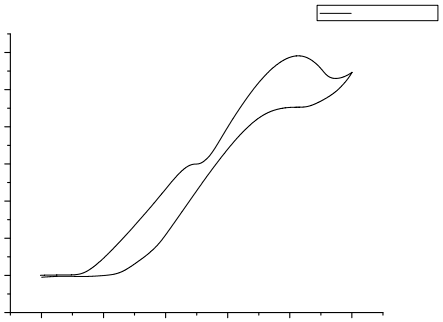


Fig. 8: Cyclic Voltammograms of $[Cu(L^2)_2]$

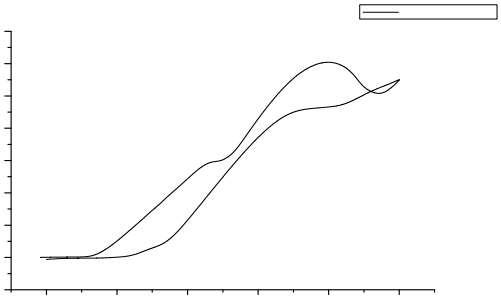


Fig. 9: Cyclic Voltammograms of $[Cu(L^3)_2]$

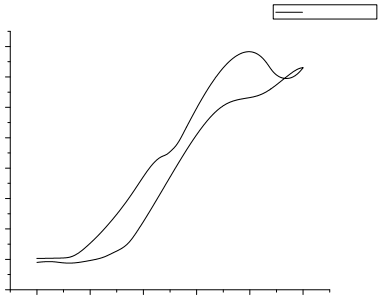


Fig.-10: Cyclic Voltammograms of $[Cu(L^4)_2]$

The electrochemical findings obtained in the present study are also relevant from a biological perspective. Curcuminoid metal complexes possessing reversible or quasi-reversible electron-transfer properties are often associated with enhanced antioxidant activity and redox-mediated therapeutic behaviour.[1-5] Since oxidative stress is associated with several pathological disorders, the ability of these complexes to undergo controlled electron transfer may contribute to their medicinal potential. Thus, the present work supports Sustainable Development Goal-3 (Good Health and Well-being) by contributing to the understanding and development of biologically significant redox-active coordination compounds.

CONCLUSION

The present cyclic voltammetric investigation demonstrates that curcuminoid ligands (HL^1–HL^4) undergo well-defined reversible redox processes in a methanolic medium containing tetrabutylammonium iodide as the supporting electrolyte. The small ΔE_p values (0.05–0.07 V) and I_{pa}/I_{pc} ratios close to unity indicate diffusion-controlled reversible electron transfer for the free ligands, while the nature of the aromatic substituents significantly influences the redox potentials through electronic effects. Upon coordination with Cu(II), the electrochemical behaviour changes considerably, with the appearance of an additional Cu(II)/Cu(I) redox couple exhibiting quasi-reversible behaviour along with noticeable changes in ligand-centred redox potentials due to metal–ligand charge delocalisation. The results clearly establish that both substituent effects and Cu(II) coordination play crucial roles in modulating the electron-transfer characteristics of curcuminoids. The study contributes to understanding the electrochemical properties of biologically active curcuminoid metal complexes and their potential redox-dependent applications. Since such redox-active Cu(II) complexes are associated with antioxidant and therapeutic properties, the present work also supports Sustainable Development Goal 3 (Good Health and Well-being). Further investigations involving other transition-metal complexes and biological correlation studies may provide additional scope for developing pharmacologically important coordination compounds.

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CONFLICT OF INTERESTS

Both authors declare that there is no conflict of interest in connection with the publication of this article.

AUTHOR CONTRIBUTIONS

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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