RASĀYAN *J. Chem.*
Vol. 19 | No. 3 | 1507-1514 | July-September | 2026
ISSN: 0974-1496 | e-ISSN: 0976-0083 | CODEN: RJCABP
http://www.rasayanjournal.com
http://www.rasayanjournal.co.in

DESIGN, SYNTHESIS, CHARACTERIZATION AND ANTIMICROBIAL ACTIVITY OF METAL (II)-AZO COMPLEXES

H. R. Mallikarjuna✉, K. S. Manjunatha, B. M. Prasanna, J. K. Prasannakumar, P. S. Bhumika, K. S. Pavithra, and M. G. Thriveni
Department of Chemistry, Bapuji Institute of Engineering and Technology, Davangere-577004, An Autonomous Institute Affiliated to VTU, Karnataka, India
✉Corresponding author: mallikarjunahr@bietdvg.edu

ABSTRACT
Azo dyes and their derivatives with respective metal complexes have emerged as promising drug candidates due to their broad-spectrum physiological properties. The presently used medicines in the market are facing challenges like antimicrobial resistance; it would insist to innovate therapeutic remedies through different strategies like synthetic metalation. Among them, metal (II)-azo derivatives, especially the Cu, Co, Ni and Zn complexes, by modifications, can rationalize their efficacy against microorganisms, making them promising lead molecules for treating various infectious agents by the chelation process. Chelation will increase lipophilicity and reduce polarity; due to this, the molecules will easily enter the microbial cell membrane and disrupt the microbial activity. The present study highlights the synthesis of novel azo-metal complexes, which were characterized based on their UV-VIS, FT IR, NMR, XRD, physical, and thermal properties. The synthesized compounds were allowed for antimicrobial screening against various bacterial and fungal strains; the results showed that the compounds showed significant to moderate activity, and they were considered for promising lead molecules for future research work.
Key words: Dyes, Azo Metal Complexes, Chelation, TGA-DTA, Antimicrobial Activity.
RASĀYAN *J. Chem.*, Vol. 19, No.3, 2026

INTRODUCTION

In the present scenario, azo metal (II)complexes played a very important role because of their broad-spectrum applications like chemical, thermal, optical and biological properties. They had good organic solubility in different solvents for digital optical applications in versatile recording media.[1-8]

The azo metal (II) complexes with a 500 *nm* wavelength having high absorption capacity, and they were short effort associations among the modification (λ_{max}) of maximum from azo metal (II) complex compounds with their donors and the structures of the respective azo metal complexes. The blue-violet light has a shorter wavelength, and it appears in the range 405 nm,[9] giving the impression of being a hopeful organic material for recording to the near future with maximum density disc-recordable (HDDVD-R) and digital versatile systems that are used for a high wavelength numerical aperture in the range of 0.85 of wavelength 405 nm.

Recently, the azo metal complexes have been reported isoxazole with β-diketone derivatives have absorption in the region of blue violet light wavelength poses virtuous optical characteristic features with high refractive index and low extension coefficient value between 0.2 to 2.08 at the wavelength 405 nm,[10] though, limited research have been described on azo metal (II) complex syntheses and their thermal properties of, and little homework has done on sandwiched between the difference (λ_{max}) in absorption maximum of azo metal (II) complexes and ligands with the structures of the complex compounds.

Based on attention of the above-mentioned requirements; the present work is mainly focused on the syntheses of the azo dye and its complex structures having different central metal atoms (Cu^{2+}, Co^{2+}, Zn^{2+} and Ni^{2+}) to ligands and their structures of the complexes were addressed meanwhile, based on the literature we were allowed the synthesized complex molecules for biological activity to address the sustainable development goal good health and well-being.

EXPERIMENTAL

Material and Methods

IR spectra of azo dye compounds were verified in KBr pellets by FT-IR 8400s SHIMADZU spectrometer in the region of 4000 cm^{-1}- 400 cm^{-1}. Proton NMR spectra were recorded using an AMX400 FT-NMR spectrometer in DMSO-d_6 at 400 MHz, and tetramethyl silane was used as an internal standard. An LC-MSD-trap-XCT plus mass spectrometer was used to record the mass spectra of the synthesized compounds. The instrument SHIMADZU UV-Visible 1650 spectrometer was employed to measure the UV-Visible spectra in different solvents, and the absorption wavelength ranged from 400 to 800 nm. Elemental breakdown recorded on Thermo Finnigan FLASH EA 1112 CHN analyzer. THE SHIMADZU TA-60WS thermal analyzer was used to study the thermal behavior of the structures in open air at the rate of 5 ^oC min^{-1}. The XRD was of the metal complexes were recorded by a Philips XRD 'X' PERT PRO diffractometer.

Synthesis of Ligands (^{1}L-^{6}L)

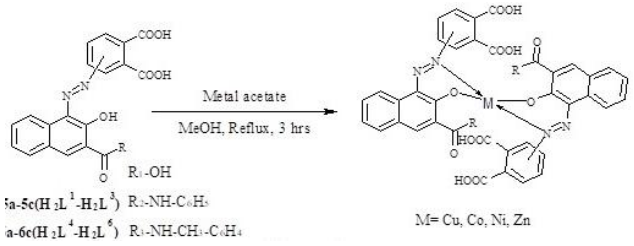
Synthesis of Dyes

The ligands are synthesized by a known procedure with standard chemicals.[11]

Synthesis of Azo Metal Complex Dyes

Synthesis of [CuL^1] Complexes

A hot solution of ligand H_2L^1 (5a-5c & 6a-6c) (0.85 g, 0.2 mol) in MeOH was added to a solution of Cu $(OAc)_2$ (0.18 g, 0.1 mol) in methanol with stirring. The mixture was stirred for about 3 hours. The solid separated was filtered, washed and finally dried in a hot air oven at 80 ºC. Similarly, other metal complexes of Co, Zn, and Ni were prepared by using the corresponding metal acetates as given in Scheme-1.



Scheme-1

RESULTS AND DISCUSSION

The metal complex was found by the reaction of one mole of metal with two moles of the ligand. The product is found in good yield. The molecular formula, molecular weight, solubility and yield were reported in Table-1. As can be seen in this table, the synthesized complexes are colored and are stable at room temperature and soluble in DMSO, DMF, and NaOH.

Spectral Studies

The infrared spectra of the synthesized complexes were recorded with KBr pellets on a SHIMADZU-8400 FTIR spectrometer. The spectra of the complexes with various ligands exhibited an identical pattern. The infrared spectrum of the azo dyes showed absorption in the region of 1600-1610 cm^{-1}, which is for (-N=N-) of the azo group. This band gradually shifts to the region of 10-15 cm^{-1} due to chelation of the ligand with the central metal ion. The N-H absorption peak was expected to appear in the region of 3300-3400 nm, and the acid containing a carboxylic group appeared in the region of 1700-1725 nm. The ^{1}H NMR spectrum of compounds shows aromatic protons in the expected range between δ: 6.5-8.7 ppm. The peak at δ: 16 ppm for -OH protons disappeared in the metal complex.

Absorption Spectra

Solvent effect: DMSO and 1N NaOH are used to record the absorption spectra of the complexes in a concentration range of 10^{-4} to 10^{-5} molar solution. A typical spectrum of Co^3L, Cu^3L, Ni^3L and 5c in DMSO is shown in Fig.-1. The visible absorption spectra of the complexes were found in the range of

380 to 502 nm, which shows variation with the polarity of the solvents. λ_{max} of the dyes and metal complexes in 1N NaOH solution, in case of free ligands, due to the formation of phenoxide ion, formation undergone blue shift is observed, but in case of complexes, both in DMSO and NaOH, the absorption appears at showed marginal variation (e.g. λ_{max}: 488 nm for 5c, 507nm for ^{3}LCu, 509nm for ^{3}LCo, in NaOH).

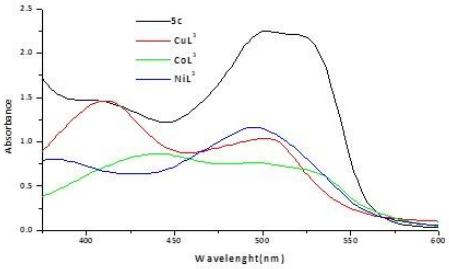


Fig.-1: Visible Spectrum of 5c. $Co^{3}L$, $Cu^{3}L$ and $Ni^{3}L$ in

Powered X-ray Studies

The Powder X-ray diffraction (XRD): Crystallographic data for compounds were recorded by using Cu-Kα (λ=1.5438°A) radiation, and the data are summarized in Table-1. The spectrum gives a continuous baseline, which is due to the nanocrystalline portion, which simply scatters the X-ray beam in the background. The resulting X-ray diffraction investigation patterns of the complexes are shown in Fig.-2.

Table-1: Powdered XRD data of the complex

Complex	2 θ	Lattice spacing d(A^0)	Relative intensity (%)	Crystal size(*nm*)
^{2}L-Cu	10.244 6.7997 9.4357 26.168	8.63459 12.9998 9.37317 3.40547	100 73.7 28.2 25.3	68.12
^{2}L-Ni	32.135 19.052 8.5610 28.063	2.78542 4.65821 10.3288 3.17965	100 76.8 48.7 48.4	61.79
^{3}L-Cu	5.1534 6.5927 18.616 32.452	17.1485 13.4074 4.76631 2.75893	100 68.3 23.8 21.6	29.72
^{3}L-Ni	8.2666 10.163 47.486 36.630	10.6960 8.70346 1.91311 2.45332	100.0 41.5 39.8 36.3	17.02
^{4}L-Cu	8.6625 26.962 11.844 23.018	10.2080 3.30690 7.47197 3.86382	100 64.5 37.5 18.2	79.46
^{4}L-Ni	7.8390 8.6997 11.884 8.4205	11.2784 10.1645 7.44658 10.5008	100 83.7 25.8 23.5	79.32
^{5}L-Cu	6.1787 19.286 25.249 28.463	14.3049 4.60230 3.52723 3.13590	100 75.6 60.3 45.7	52.83
^{5}L-Ni	6.1490	14.3739	100	47.55

	19.260 25.253 28.452	4.60832 3.52669 3.13712	48.4 28.4 24.6	
^{6}L-Cu	7.1074 6.4283 7.6211 26.572	12.4376 13.7500 11.6004 3.35454	100 50.5 39.3 26.3	67.98
^{6}L-Ni	7.7504 8.4758 6.9017 9.3112	11.4071 10.4324 12.8078 9.49829	100 88.0 75.3 36.6	59.51

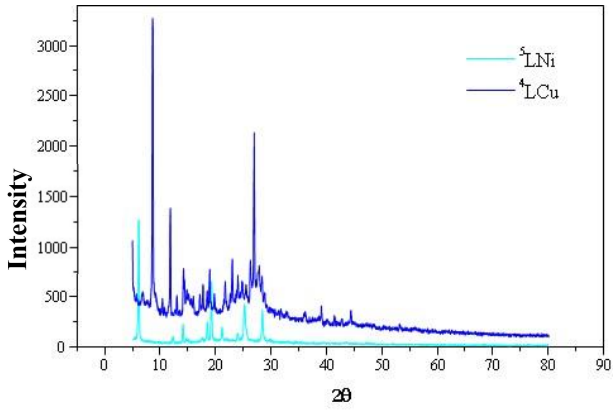


Fig.-2: XRD Spectra of ^{4}LCu, ^{5}LNi

Thermal Studies

Broido's method[12] was adopted to record the kinetic and thermodynamic parameters of the synthesized complexes in air between the temperature range of 29-600 °C. The thermo analytical information of all the complexes is specified in Table-2 and 3. The TG and DTA curves of the complexes are given in Fig.-3 and 4. Thermo gravimetric analytical curves and the corresponding data indicate that the complexes decompose in two stages. The first decomposition corresponds to moisture and water molecules, the second step, at the range between 230-480 °C, corresponds to the degradation of the substituent moiety in the complex. The second step shows that the degradation is very rapid. DTA curves reveal that degradation is exothermic in nature. The activation energy of the TGA graph clearly talks about the decomposition temperatures of the corresponding complexes (Table-5). If the complex with a high activation energy causes rapid degradation, whereas a low activation energy represents the gradual degradation around its decomposition temperatures.

Table-2: Represents Thermo-Analytical Data of the Metal Complexes

Complex	Mass loss temp (°C)	DTA $_{max}$ (°C)	Mass loss (%)		Probable mode of decomposition and fragment loss
			Found	Calc.	
^{1}LCu	60-180	90	9.55	11.62	Loss of moisture
	220-480	338	79.54	78.54	Complex moiety
^{2}LNi	60-150	77	10.50	11.22	Loss of moisture
	315-380	346	80.21	80.62	Complex moiety
^{3}LCu	70-170	111	8.56	10.56	Loss of moisture
	230-350	303	78.63	82.12	Complex moiety
^{4}LCu	50-160	98	9.03	10.23	Loss of moisture
	295-362	309	76.66	78.74	Complex moiety
^{5}LCu	50-150	106	9.23	10.56.	Loss of moisture
	230-400	313	79.00	82.56	Complex moiety

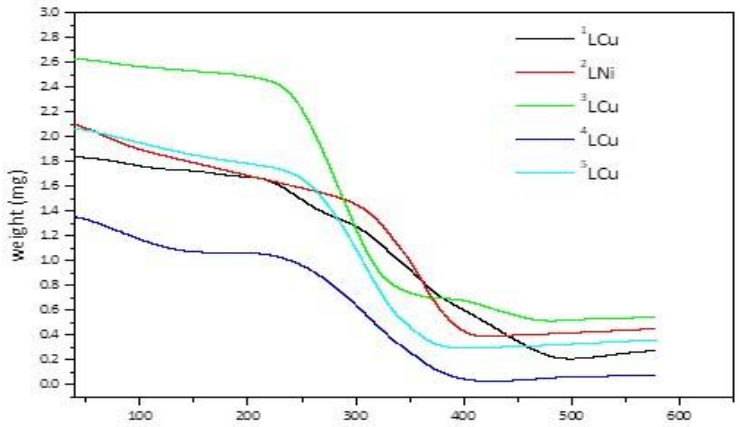


Fig.-3: TGA Curves of Complex

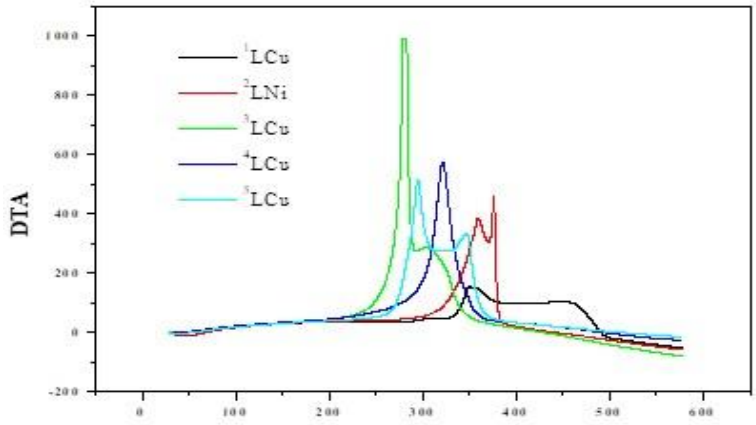


Fig.-4: DTA Curves of Complex

Table-3: Kinetic Parameters of the Metal Complexes

Dye	Decomposition range	E_a (kJ mol^{-1})	Ln A(s^{-1})	ΔH (kJ mol^{-1})	ΔS (kJ mol^{-1})	ΔG (kJ mol^{-1})
[1]LCu	70-180 (^{0}C)	0.920	4.1263	-3.0678	-160.86	55.913
	220-480	14.167	8.8363	9.0880	-147.48	90.122
[2]LNi	60-150	0.8420	3.4023	-2.0678	-152.86	53.501
	315-380	20.810	10.509	15.664	-147.12	91.086
[3]LCu	70-170	0.950	4.2683	-3.512	-163.86	58.025
	230-350	16.086	9.5470	11.297	-147.87	85.187
[4]LCu	50-160	0.8420	3.4023	-2.0678	-152.86	53.501
	295-362	25.692	11.9820	20.853	-147.62	85.937
[5]LCu	50-150	0.7530	2.9023	-1.905	-149.72	51.542
	230-400	39.125	15.192	34.253	-147.03	86.199

Magnetic Properties of the Complex

In the present investigation, the magnetic properties of Cu (II) complex of various azo dye derivatives were discussed in Table-4.

Antimicrobial Activity

The compounds were tested for *in vitro* antimicrobial activity against fungal strains like, *Candida albicans, Aspergillus niger and Aspegillus flavons* and bacterial strains like *Pseudomonas aureginosa*, *Escherichia coli*, and *Bacillus subtilis* for antibacterial activity with fluconazole and Kanamycin as standards, respectively. The compounds were dissolved in DMSO, and the inhibition values of the

compounds were determined by using nutrient agar medium A and Potato dextrose agar for bacteria and fungi, respectively. Table-5 represents the results of the *in vitro* activity determined by the cup plate method.[13]

Table-4: Magnetic Susceptibility Data of Cu (II) Complex

S. No.	Compound	Field strength(K. Gauss)	Magnetic susceptibility $\times 10^{-6}$	μ_{eff} (B.M)
1	^{1}LCu	1.33	2116.15	2.27
		1.75	1951.41	2.18
		2.20	1837.05	2.11
		2.66	1757.80	2.06
		3.10	1731.19	2.05
		3.58	1598.57	1.97
2	^{2}LCu	1.33	2129.13	2.28
		1.75	1964.02	2.21
		2.20	1842.16	2.12
		2.66	1762.58	2.09
		3.10	1734.26	2.06
		3.58	1649.64	1.98
3	^{3}LCu	1.33	1960.16	2.21
		1.75	1846.28	2.16
		2.20	1748.57	2.07
		2.66	1712.29	2.02
		3.10	1642.10	1.96
		3.58	1624.57	1.94
4	^{4}LCu	1.33	1800.32	2.09
		1.75	1660.46	2.01
		2.20	1562.95	1.95
		2.66	1495.68	1.90
		3.10	1472.99	1.89
		3.58	1400.45	1.84
5	^{5}LCu	1.33	1953.77	2.18
		1.75	1930.71	2.16
		2.20	1884.64	2.14
		2.66	1859.88	2.12
		3.10	1864.96	2.13
		3.58	1788.05	2.08
6	^{6}LCu	1.33	2259.93	2.34
		1.75	2084.37	2.25
		2.20	1961.97	2.18
		2.66	1877.52	2.13
		3.10	1725.76	2.05
		3.58	1654.56	2.00

Table-5: Antimicrobial Activity of the Metal Complexes.

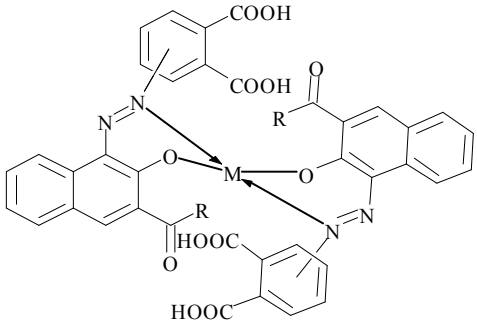
Compound	Zone of inhibition in mm (mean±SD)					
	Bacterial Strains			Fungal Strains		
	E. Coli	*P. aeruginosa*	*B. subtilis*	*A. niger*	*A. flavons*	*C. albicans*
^{2}LCu	9.17±0.28	12.50±0.46	10.67±0.52	18±0.30	12±0.30	18±0.30
^{2}L Ni	7.00±0.38	11.00±0.48	15.17±0.40	16±0.30	08±0.20	18±0.30
^{3}L Cu	13.33±0.41	9.33±0.37	12.00±0.55	08±0.30	08±0.20	13±0.20
^{3}L Co	14.33±0.43	11.00±0.39	13.83±0.54	16±0.20	10±0.20	14±0.40
^{3}L Ni	12.17±0.50	12.67±0.38	11.83±0.48	10±0.20	12±0.30	18±0.40
^{4}L Cu	8.67±0.25	11.67±0.49	15.00±0.50	16±0.30	20±0.30	12±0.10

^{4}L Ni	11.83±0.43	15.67±0.35	11.50±0.31	14±0.20	17±0.40	15±0.20
^{5}L Cu	14.67±0.39	17.67±0.47	12.33±0.62	22±0.20	26±0.20	20±0.20
^{5}L Ni	10.50±0.41	15.17±0.49	9.67±0.18	14±0.20	13±0.20	10±0.20
^{6}L Zn	12.17±0.32	12.67±0.37	15.83±0.21	18±0.20	15±0.20	13±0.20
Kenamycin	18.83±0.21	21.33±0.39	19.33±0.29	--	--	--
Flucanazole	--	--	--	28±0.30	32±0.20	24±0.20

Escherichia coli (E. coli); Pseudomonas aeruginosa (P. aeruginosa); Bacillus subtilis (B. subtilis); Aspergillus niger (A. niger); Aspergillus flavus (A. flavus); Candida albicans (C. albicans). Bore Size: 9 mm, SD: Standard deviation.

In the magnetic susceptibility measurements, the mercury tetrathiocyanatecobaltate(II) was used as a calibrate and it was prepared by the following method is as follows, to the boiling solution of mercuric chloride (5.4 *g*) in 60 ml distilled water was added an aqueous solution of cobalt sulphate heptahydrate (5.6) and ammonium thiocyanate (6.0 *g*) in 10 ml distilled water, continue the reaction for two more minutes with constant stirring and heating, the resulting product was washed, filtered and dried at 120 °C for 30 minutes.

The measured spin values of μ_{eff} for all the Cu (II) complexes are higher than the expected 1.73 BM, and it corresponds to one unpaired electron. These developed values are from the orbital contribution to μ_{eff} as a result of mixing with ground state orbitals $(b_2g)^2$, $(e_g)^3$ and (a_{1g}) with high orbital degeneracy interaction states of $(b_2g)^2$, $(e_g)^3$ and (a_{1g}). The difference in μ_{eff} may be encountered due to intermolecular cooperative interactions with orbital interactions. The square planar structure for the metal complexes is proposed based on ^{1}H NMR, IR and magnetic susceptibility analysis.



CONCLUSION

In a chelation process of chemical reactions, the metal atom binds with the ligands at the centre, forming a stable ring-like structure. This is very important in medical therapy. Chelates will help to remove the toxic metals from the host. Chelates will also extend their biopotency against disease-causing microorganisms by increasing lipophilicity and reducing polarity; due to this, the chelates will easily enter the microbial cell membrane and disrupt the microbial activity. The present study highlights the synthesis of novel azo-metal complexes, which were confirmed from different spectral analyses, mainly absorption spectra, XRD and thermogravimetric analysis. In continuation with our research, adherence of the synthesised compounds was allowed for antimicrobial activity, and the results revealed that the compounds ^{3}LCo, ^{3}LNi and ^{5}LCu showed good activity against bacteria *E. Coli*, *P. aeruginosa* and *B. subtilis*, as well as fungal microorganisms *A. niger*, *A. flavons* and *C. albicans*. The remaining compounds ^{2}LCu, ^{3}LCu, ^{4}LCu, ^{4}LNi, ^{5}LNi and ^{6}LZn showed moderate activity as compared with kanamycin and fluconazole standards. Overall, the compound ^{5}LCu exhibits potent antimicrobial activity. These complexes are promising lead molecules for future drug candidates, and this will support the Sustainable Development Goals in the category of good health and well-being.

ACKNOWLEDGMENTS

The authors express their heartfelt thanks and gratitude towards our institute, Bapuji Institute of Engineering and Technology, and also our management, Bapuji Education Association, for the completion of this research work.

CONFLICT OF INTERESTS

The authors have no conflicts of interest to declare. All co-authors have seen and agree with the contents of the manuscript, and there is no financial interest to report. We certify that the submission is original work and is not under review at any other publication.

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:

H. R. Mallikarjuna http://orchid.org/0000-0002-3353-8611
K. S. Manjunatha https://orcid.org/0000-0003-0796-2002
B. M. Prasanna http://orchid.org/0000-0002-8972-5360
J. K. Prasannakumar http://orchid.org/0000-0002-9249-5359
P. S. Bhumika http://orchid.org/0009-0004-5176-6767
K. S. Pavithra https://orcid.org/0000-0002-8458-6891
M. G. Thriveni https://orcid.org/0009-0007-2378-7095

Cite this article as: H. R. Mallikarjuna *et al.*, *Rasayan J. Chem.*, 19(3), 1507-1514(2026), https://doi.org/10.31788/RJC.2026.1939739

REFERENCES

1. V. Nguyen, A. T. Ngoc Vu, L. V. Bazan, R. T. Galeev, A. N. Utenyshev, E. B. Markova, V. Thuan Le and O. V. Kovalchukova, *RSC Advances*, **12**, 888(2022), https:// doi.org/10.1039/D1RA07254D
2. D. P. Mishra, P. K. Sahu, B. Acharya, S. P. Mishra, S. Bhati, *Results in Chemistry*, **10**, 101712 (2024), https://doi.org/10.1016/j.rechem.2024.101712
3. S. F. Kayed, *Inorganic Chemistry Communications*, **142**, 109634(2022), https://doi.org/10.1016/j.inoche.2022.109634
4. H. A. El-Ghamry, M. Gaber, M. S. Napolion, A. Atta, T. M. Mohamed and N. A. El-Wakiel, *Scientific Reports*, **15**, 20213 (2025), https://doi.org/10.1038/s41598-025-06518-4.
5. S. M. Obaid, A. E. Abd-Almonuim, H. A. S. Al -Naymi, A. J. Jarad, M. M. Saleh, *Heliyon*, **10(18)**, e37849(2024), https://doi.org/10.1016/j.heliyon.2024.e37849
6. F. Eltaboni, N. Bader, R. El-Kailany, N. Elsharif, A. Ahmida, *Journal of Chemical Reviews,* **4(4)**, 313(2022), https://doi.org/10.22034/jcr.2022.349827.1177
7. B. Kumari, K. Ahmad, *Applied Organometallic Chemistry*, **38(7)**, e7525(2024), https://doi.org/10.1002/aoc.7525
8. H. M. Abd El-Lateef**,** K. Shalabi, A. M. Arab and Y. M. Abdallah, *ACS Omega*, **7(27)**, 23380(2022), https:// doi.org/10.1021/acsomega.2c01629
9. H. Park, E. Kim, D., Kim, H. Lee, Bulletin *of the Chemical Society of Japan*, **75**, 2067 (2002)
10. F. Huang, Y. Wu, D. Gu, F. Gan, *Chinese Physics Letters*, **20(12)**, 2259 (2003)
11. H. R. Mallikarjuna, K. S. Manjunatha, B. M. Prasanna, Vinay Parol, *International Journal of Science and Technology,* **16(1)**, 2229(2025), https:// doi.org/10.71097/IJSAT.v16.i1.1791
12. A. Broido, *Journal of Polymer Science Part A-2*, **7(10)**, 1761(1969), https://doi.org/10.1002/pol.1969.160071012
13. K. S. Manjunataha, N. D. Satyanarayan, S. Harishkumar, *International Journal of Pharmacy and Pharmaceutical Sciences,* **8(10)**, 251(2016), http://dx.doi.org/10.22159/ijpps.2016v8i10.13957

[RJC-9739/2026]