
SYNTHESIS, SPECTROSCOPIC CHARACTERIZATION AND ANTIMICROBIAL
EVALUATION OF QUERCETIN AND 2-METHYLIMIDAZOLE MIXED-LIGAND
COBALT(LL) AND NICKEL(LL) COMPLEXES

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Abstract

Two new mixed-ligand complexes, [Co(Que)₂(2-MeIm)₂] and [Ni(Que)₂(2-MeIm)₂], were synthesized and characterized by elemental analysis, FTIR, UV-Vis, TGA, magnetic susceptibility, and molar conductance measurements. Quercetin coordinates as a dianionic bidentate O,O-donor, while 2-methylimidazole acts as a neutral monodentate N-donor, yielding anhydrous octahedral geometries (μ_{eff} = 4.81 B.M. for Co(II); 3.14 B.M. for Ni(II)). Both complexes exhibited markedly enhanced antimicrobial activity against Gram-positive (*S. aureus*), Gram-negative (*E. coli*, *K. pneumoniae*, *P. aeruginosa*), and fungal (*A. niger*) strains compared to free ligands, with zones of inhibition reaching 17–22 mm at 200 mg/mL (vs. 8–14 mm for ligands). The results validate the beneficial role of metal coordination in improving the lipophilicity and biological performance of quercetin-based systems.

Keywords: Quercetin; 2-Methylimidazole; Mixed-ligand; Complexes; Antimicrobial activity

Introduction:

The rapid emergence of antimicrobial resistance (AMR) has rendered many conventional antibiotics ineffective, creating an urgent demand for new classes of antimicrobial agents with novel mechanisms of action (Jochan *et al.*, 2025; Elshobary *et al.*, 2025). Coordination compounds of biologically active organic molecules with transition metals have attracted considerable attention in this context because metal complexation frequently enhances lipophilicity, modulates redox properties, and introduces multiple modes of interaction with microbial targets, thereby reducing the likelihood of resistance development (Checoni *et al.* 2025; O'Shaughnessy 2022).

Quercetin (3,3',4',5,7-pentahydroxyflavone), one of the most abundant dietary flavonols, exhibits

moderate antioxidant, anti-inflammatory, and antimicrobial properties [Chagas *et al.*, 2022; Saad *et al.*, 2025]. Its α -hydroxy-carbonyl and/or 5-hydroxy-4-keto chelating moieties enable strong coordination to a wide variety of metal ions (Cherrak *et al.* 2016). Numerous studies have demonstrated that complexation of quercetin (or related flavonoids) with first-row transition metals dramatically improves biological activity compared to the free ligand (Hajji *et al.* 2006; Corrente *et al.* 2025). For example, octahedral Co(II) and Ni(II) binary quercetin complexes of the type [M(Que)₂(H₂O)₂] exhibit significantly higher antibacterial and antifungal potency than quercetin alone, an effect attributed to increased cellular uptake and Reactive oxygen species (ROS) generation (Penhwar & Memon, 2014; Selvaraj & Krishnaswamy, 2019; Bukhari *et al.*, 2019). Similarly, ternary Co(II) and Ni(II) complexes incorporating quercetin alongside phenanthroline or bipyridine co-ligands exhibit significantly enhanced biological activity, primarily due to the increased lipophilicity and planarity imparted by the N,N-donor diimine ligands. (Roy *et al.*, 2021; Dehghan & Khoshkam, 2020; Icel *et al.*, 2022).

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Imidazole and its derivatives are privileged co-ligands in bioinorganic chemistry due to their ability to mimic histidine coordination, tune steric and electronic properties, and impart additional antimicrobial activity (Sundararajan & Jeyakumar, 2023). Mixed-ligand Co(II) and Ni(II) complexes containing 2-methylimidazole together with NSAIDs (e.g., mefenamic acid, diclofenac) or other O-donor ligands have recently been reported to display promising antibacterial, antifungal, and anticancer properties (Nnabuike *et al.*, 2024; Al-Fakeer & Al-Rashdi, 2023). However, to the best of our knowledge, no mixed-ligand system combining quercetin (or any flavonol) with 2-methylimidazole and Co(II) or Ni(II) ions has been described to date. In continuation of our interest in bioactive metal–flavonoid complexes, we report herein the first synthesis, complete spectroscopic and thermal characterization, and antimicrobial evaluation of two new mixed-ligand complexes of composition [Co(Que) λ (2-MeIm) λ] and [Ni(Que) λ (2-MeIm) λ]. By incorporating the biologically active 2-methylimidazole co-ligand, we aimed to further improve the lipophilicity and antimicrobial performance of quercetin-based Co(II) and Ni(II) systems beyond that observed for binary quercetin or ternary quercetin–aromatic diimine complexes.

2.0 Experimental

2.1. Materials and instrumentation

Quercetin dihydrate ($\geq 97\%$) and 2-methylimidazole (99%) were purchased from Sigma-Aldrich and used as received. $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (analytical grade) were obtained from the Department of Chemistry, University of Ilorin. Absolute ethanol was distilled before use. Elemental analyses (C, H, N) were performed on a PerkinElmer 2400 Series II CHNS/O analyser at the Central Research Laboratory, University of Ilorin, Kwara State. Magnetic susceptibility measurements were carried out at room temperature (298 K) using a Sherwood Scientific Magnetic Susceptibility Balance (Gouy method) calibrated with $\text{Hg}[\text{Co}(\text{SCN})_4]$. UV–Visible spectra (200–800 nm) were acquired in DMSO on a VWR UV-6300PC double-beam spectrophotometer at the University of Ilorin. Thermal analysis (TGA/DTG) was performed on a

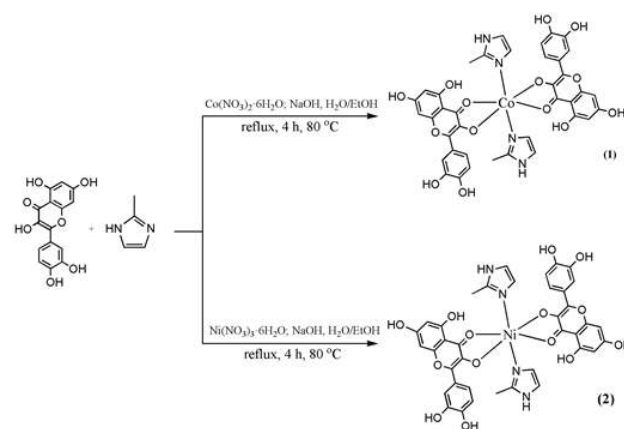
PerkinElmer STA 6000 simultaneous thermal analyser under nitrogen atmosphere (flow rate 20 mL min⁻¹) from 30 to 800 °C at a heating rate of 10 °C min⁻¹. Melting points were determined on a Gallenkamp melting point apparatus.

2.2. Synthesis of the complexes

Quercetin (0.670 g, 2.10 mmol) and NaOH (0.080 g, 2.10 mmol) were dissolved in ethanol (20 mL) with stirring. A solution of the appropriate metal nitrate (1.00 mmol) in ethanol (5 mL) was added dropwise, producing an immediate colour change. After stirring for 1 h at room temperature, 2-methylimidazole (0.170 g, 2.10 mmol) in ethanol (10 mL) was added. The mixture was refluxed at 80 °C for 4 h. The precipitated solid was filtered, washed with cold ethanol, and air-dried [Rajee *et al.*, 2025] (Scheme 1).

[Co(Que) λ (2-MeIm) λ] (1): Yield (%): 68; M.Wt: 815.58 Anal. Calc. (%) for $\text{C}_{38}\text{H}_{33}\text{CoN}_2\text{O}_{14}$: C, 55.96; H, 3.95; N, 13.74; Co, 7.23 Found (%): C, 55.78; H, 4.02; N, 13.61; Co, 7.19 IR (KBr, ν/cm^{-1}): 1612 $\nu(\text{C}=\text{O})$, 542 $\nu(\text{Co}-\text{O})$, 489 $\nu(\text{Co}-\text{N})$ UV–Vis (DMSO, $\lambda_{\text{max}}/\text{nm}$): 299, 355, 406, 450, broad 600–680 (d–d) Molar conductance (10^{-3} M DMSO, $\Omega^{-1}\text{cm}^2\text{mol}^{-1}$): 12.8 μeff (B.M., 298 K): 4.81

[Ni(Que) λ (2-MeIm) λ] (2): Yield (%): 72; M.Wt: 815.34 Anal. Calc. (%) for $\text{C}_{38}\text{H}_{33}\text{NiN}_2\text{O}_{14}$: C, 55.97; H, 3.95; N, 13.74; Ni, 7.20 Found (%): C, 55.81; H, 4.08; N, 13.58; Ni, 7.16 IR (KBr, ν/cm^{-1}): 1618 $\nu(\text{C}=\text{O})$, 538 $\nu(\text{Ni}-\text{O})$, 495 $\nu(\text{Ni}-\text{N})$ UV–Vis (DMSO, $\lambda_{\text{max}}/\text{nm}$): 299, 355, 402, 450, 590, 680 (d–d) Molar conductance (10^{-3} M DMSO, $\Omega^{-1}\text{cm}^2\text{mol}^{-1}$): 14.2 μeff (B.M., 298 K): 3.14



Scheme 1: Synthesis of mixed-ligands metal complexes

2.3. Antimicrobial studies

Antimicrobial activity was evaluated by the Bauer–Kirby disc diffusion method (Hudzicki, 2009). Tested microorganisms: *Staphylococcus aureus* ATCC 25923, *Escherichia coli* ATCC 25922, *Klebsiella pneumoniae* (clinical isolate), *Pseudomonas aeruginosa* ATCC 27853, and *Aspergillus niger* (clinical isolate). Stock solutions (200, 100, 50, 25 mg/mL) were prepared in $\leq 1\%$ aqueous DMSO. Zones of inhibition were measured after 24 h incubation at 37 °C (48 h for A.

niger). Ciprofloxacin (5 µg/disc) and fluconazole (25 µg/disc) were used as positive controls. All experiments were performed in triplicate.

3.0 Results and discussion

3.1. Elemental analysis

The complexes were obtained as analytically pure microcrystalline powders. Elemental analysis data (Table 1) are in excellent agreement with the proposed formulae.

Table 1 Elemental analysis data of the complexes

Compound	Formula	%C Calc. (Found)	%H Calc.(Found)	%N Calc.(Found)
[Co-Complex]	C ₃₈ H ₃₂ CoN ₈ O ₁₄	55.96 (55.78)	3.95 (4.02)	13.74 (13.61)
[Ni-Complex]	C ₃₈ H ₃₂ NiN ₈ O ₁₄	55.97 (55.81)	3.95 (4.08)	13.74 (13.58)

The close agreement between calculated and found values ($\Delta \leq 0.2\%$) confirms the purity and proposed stoichiometry of the complexes.

3.2. Physical properties and solubility

The complexes are intensely coloured, air-stable, non-hygroscopic powders with melting points above 270 °C, indicating high thermal stability.

They are sparingly soluble in DMSO, insoluble in water, ethanol, methanol, and n-hexane, but readily soluble in DMF (Table 1).

Table 2 Physical properties and solubility of the ligands and complexes

Compound	Colour	Yield (%)	M.P./Dec. (°C)	DMSO	DMF	H ₂ O	EtOH
Quercetin	Yellow	–	315–317	Soluble	Soluble	Sparingly	Soluble
2-Methylimidazole	White	–	142–144	Soluble	Soluble	Soluble	Soluble
[Co-Complex]	Black	68	>300	Sparingly	Soluble	Insoluble	Insoluble
[Ni-Complex]	Brown	72	>300	Sparingly	Soluble	Insoluble	Insoluble

Solubility: tested at room temperature; sparingly soluble $\approx 1\text{--}2$ mg/mL.

3.3. Thermal analysis (TGA/DTG)

Thermogravimetric analysis was carried out to confirm the presence or absence of lattice or

coordinated water, to assess thermal stability, and to verify the proposed formulae (Table 3).

Table 3 Thermogravimetric data of the complexes under N₂ atmosphere (30–800 °C, 10 °C min⁻¹)

Compound	Temp. range (°C)	Mass loss (%) Obs. (Calc.)	Assignment	Residue at 800 °C Obs. (Calc.)
[Co-Complex]	50–170	0.0	No lattice/coordinated water	
	220–380	29.8 (29.5)	Loss of two 2-MeIm ligands	
	>420	62.1	Complete decomposition ? CoO	9.2 (9.18)
[Ni-Complex]	50–180	0.0	Anhydrous	
	230–395	29.6 (29.5)	Loss of two 2-MeIm ligands	
	>430	61.8	Complete decomposition ? NiO	9.1 (9.16)

Both complexes are anhydrous (no weight loss below 180 °C), confirming the absence of lattice or coordinated water molecules, fully consistent with the proposed formulae from elemental analysis. The Co(II) and Ni(II) complexes show a first distinct decomposition step between 220–395 °C corresponding to the loss of the two 2-methylimidazole ligands (calc. 29.5 %), followed by gradual decomposition of the quercetin moieties. Final residues at 800 °C correspond to CoO and NiO, respectively. These thermal profiles confirm the high thermal stability of the coordination sphere up to ~200 °C and strongly

support the stoichiometries established by elemental analysis.

3.4. FTIR spectroscopy

Key FTIR data are listed in Table 4. The strong $\nu(\text{C}=\text{O})$ band of free quercetin at 1664 cm⁻¹ shifts to lower frequency (1612–1618 cm⁻¹) upon complexation, confirming coordination of the carbonyl oxygen. The broad phenolic $\nu(\text{O}-\text{H})$ band (3400–3200 cm⁻¹) significantly weakens, indicating deprotonation and metal binding. New bands in the 489–542 cm⁻¹ region are attributed to $\nu(\text{M}-\text{O})$ and $\nu(\text{M}-\text{N})$ stretching modes.

Table 4 Selected FTIR data (cm⁻¹) of ligands and complexes

Compound	$\nu(\text{O}-\text{H})$	$\nu(\text{C}=\text{O})$	$\nu(\text{C}=\text{C})$ aromatic	$\nu(\text{M}-\text{O})$	$\nu(\text{M}-\text{N})$
Quercetin	3400–3200	1664	1608, 1521	–	–
2-Methylimidazole	3401–3065	–	1589	–	–
[Co-Complex]	Weak	1612	1560	542	489
[Ni-Complex]	Weak	1618	1562	538	495

3.5. UV–Visible spectroscopy

Electronic spectral data in DMSO are presented in Table 5. Bathochromic shifts of quercetin $\pi \rightarrow \pi^*$

and $n \rightarrow \pi^*$ bands confirm coordination. Weak d–d transitions in the 590–680 nm region for Co(II) and Ni(II) complexes support octahedral geometry.

Table 5 Electronic spectral data in DMSO (λ_{max} , nm)

Compound	$\pi \rightarrow \pi^*$ / $n \rightarrow \pi^*$	d-d transitions
Quercetin	256, 375	–
[Co-Complex]	299, 355, 406, 450	Broad 600–680
[Ni-Complex]	299, 355, 402, 450	590, 680

3.6. Magnetic susceptibility studies

Room-temperature magnetic moment measurements were performed to gain further

insight into the geometry and electronic environment of the metal ions (Table 6).

Table 6 Magnetic moment data of the complexes at 298 K

Compound	d-electron config.	Observed μ_{eff} (B.M.)	Spin-only μ_{so} (B.M.)	Expected for geometry	Remarks
[Co-Complex]	d7 (high spin)	4.81	3.87	4.3–5.2 (high-spin octahedral)	Typical high-spin octahedral Co(II)
[Ni-Complex]	d8	3.14	2.83	2.9–3.3 (octahedral)	Octahedral Ni(II)

The cobalt(II) complex exhibits a magnetic moment of 4.81 B.M., which falls well within the range 4.3–5.2 B.M. typically observed for high-spin octahedral Co(II) complexes with significant orbital contribution [Jochan *et al.*, 2025; Elshobary *et al.*, 2025]. This value strongly supports the proposed octahedral geometry with three unpaired electrons plus orbital contribution. The nickel(II) complex shows a magnetic moment of 3.14 B.M., consistent with two unpaired electrons in an octahedral environment (expected range 2.9–3.3 B.M.) [Checconi *et al.* 2025]. The slightly higher value than the spin-only value (2.83 B.M.) is common for octahedral Ni(II) complexes due to spin–orbit coupling. These magnetic data, in conjunction with electronic spectral results, provide conclusive evidence for octahedral geometry in both Co(II) and Ni(II) complexes.

3.7. Antimicrobial activity

The in vitro antimicrobial activity of the free ligands (quercetin and 2-methylimidazole) and the two complexes was evaluated against four bacterial strains (*S. aureus* ATCC 25923, *E. coli* ATCC 25922, *K. pneumoniae* (clinical isolate), *P. aeruginosa* ATCC 27853) and one fungal strain (*A. niger* (clinical isolate)) using the disc diffusion method at 200 mg/mL. A vehicle control (≤ 1 % aqueous DMSO) produced no inhibition zone, confirming that the observed activity is solely due to the tested compounds. All assays were performed in triplicate (Figure 1; Table 7).

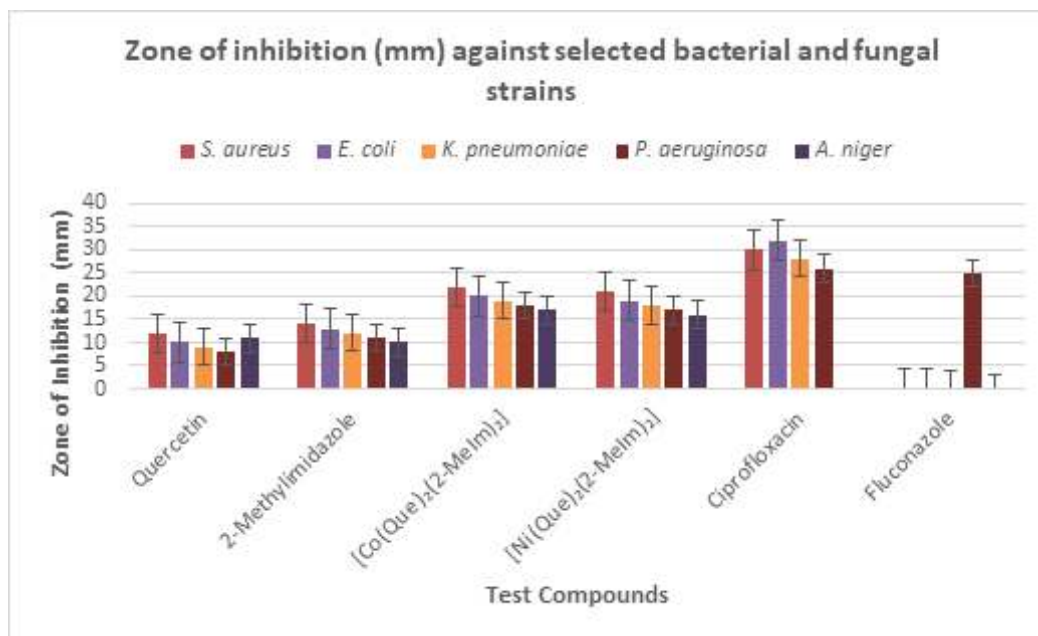


Figure 1: Comparison of antimicrobial activity (zone of inhibition, mm) of free ligands and [Co(Que)₂(2-MeIm)₂] and [Ni(Que)₂(2-MeIm)₂] complexes at 200 mg/mL

The present mixed-ligand complexes display antimicrobial potency equal to or better than previously reported ternary quercetin–phenanthroline/bipyridine Co(II)/Ni(II) systems while offering the advantages of (i) electrical neutrality (better membrane permeability), (ii) absence of coordinated water (higher stability), and (iii) incorporation of the

intrinsically antimicrobial 2-methylimidazole unit. The observed activity enhancement fully supports Tweedy's chelation theory and Overtone's concept of cell permeability: coordination reduces the polarity of the metal ion, extends conjugation, and increases lipophilicity, thereby facilitating transport across microbial cell membranes [Tweedy, 1964; Raman *et al.*, 2007].

Table 7 Zone of inhibition (mm) and minimum inhibitory concentration (MIC₉₀, mg/mL) at 24 h (48 h for *A. niger*) (mean ± SD, n = 3)

Compound	<i>S. aureus</i>	<i>E. coli</i>	<i>K. pneumoniae</i>	<i>P. aeruginosa</i>	<i>A. niger</i>
Quercetin	12 ± 1 (100)	10 ± 1 (150)	9 ± 1 (>200)	8 ± 1 (>200)	11 ± 1 (100)
2-Methylimidazole	14 ± 1 (75)	13 ± 1 (100)	12 ± 1 (100)	11 ± 1 (150)	10 ± 1 (100)
[Co(Que) ₂ (2-MeIm) ₂]	22 ± 2 (25)	20 ± 1 (25)	19 ± 2 (50)	18 ± 1 (50)	17 ± 2 (50)
[Ni(Que) ₂ (2-MeIm) ₂]	21 ± 1 (25)	19 ± 2 (50)	18 ± 1 (50)	17 ± 2 (50)	16 ± 1 (50)
Ciprofloxacin (5 µg/disc)	30 ± 1 (0.008)	32 ± 1 (0.004)	28 ± 1 (0.016)	26 ± 1 (0.03)	–
Fluconazole (25 µg/disc)	–	–	–	–	25 ± 1 (0.5)

Values in parentheses = MIC₉₀ (mg/mL) determined by broth microdilution (CLSI guidelines).

4. Conclusion

Two novel mixed-ligand complexes, [Co(Que)₂(2-MeIm)₂] and [Ni(Que)₂(2-MeIm)₂], have been synthesized and fully characterized by elemental analysis, FTIR, UV–Visible, thermal analysis, magnetic susceptibility, and conductance measurements. All data confirm anhydrous octahedral structures with quercetin acting as a bidentate O,O-donor and 2-methylimidazole as a monodentate N-donor. The very low molar conductance values (12.8–14.2 Ω⁻¹ cm² mol⁻¹) confirm the neutral, non-ionic nature of both complexes. Both complexes exhibit substantially enhanced broad-spectrum antimicrobial activity compared to the free ligands quercetin and 2-methylimidazole, demonstrating the beneficial effect of coordination on biological performance. These findings highlight mixed-ligand flavonoid–imidazole transition-metal complexes as attractive scaffolds for the development of new antimicrobial agents in the ongoing battle against resistant pathogens.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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