

Review Article

A Review of Azo–Metal Complexes: Coordination Modes, Structure–Property Relationships, and Emerging Applications

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Abstract

Azo ligands are versatile π -conjugated donors whose coordination to metal ions can substantially alter molecular geometry, charge distribution, redox behavior, optical response, thermal stability, and biological properties. This review examines azo–metal complexes from a coordination-chemistry perspective, with emphasis on donor-atom identity, denticity, coordination geometry, metal-ion effects, and the resulting structure–property relationships. Monodentate, bidentate, tridentate, and tetradentate systems are discussed using representative N-, N,O-, N,N-, and higher-denticity coordination modes. Particular attention is given to the fact that azo groups can participate in metal coordination and, in selected systems, display redox non-innocent behavior, allowing metal–ligand cooperativity and ligand-centered electron transfer. The review then evaluates biological, optical, catalytic, sensing, and energy-related applications. In biological systems, increased activity of some metal complexes relative to their parent ligands has been associated with changes in lipophilicity, membrane transport, metal-dependent reactivity, reactive oxygen species generation, enzyme interactions, and DNA binding; however, these mechanisms are system-dependent and should not be generalized from in vitro observations alone. Recent developments in photoresponsive and electroactive azo–metal systems further demonstrate the importance of integrating experimental spectroscopy with computational approaches such as density functional theory and time-dependent DFT. Finally, current limitations—including aqueous stability, metal-associated toxicity, mechanistic uncertainty, reproducibility, scalability, and the need for in vivo or device-level validation—are identified to guide future design of multifunctional azo–metal complexes.

1. Introduction

Azo compounds contain the characteristic --N=N-- linkage connecting two carbon- or heteroatom-containing fragments and constitute one of the most important families of synthetic chromophores. Aromatic azo compounds are commonly prepared through diazotization of a primary aromatic amine followed by electrophilic azo coupling with an activated aromatic or heteroaromatic partner. The resulting extended

π -conjugated systems display intense visible absorption and can be modified readily through changes in substitution, conjugation length, donor/acceptor strength, and heterocyclic incorporation [1, 2].

The coordination of azo ligands to metal ions introduces an additional level of structural and electronic control. Depending on ligand architecture, protonation state, metal identity, oxidation state, and the presence of competing donor atoms or auxiliary ligands, azo systems may coordinate through one azo nitrogen, through an azo nitrogen together with an adjacent O/N/S donor, or through multiple donor atoms distributed over a chelating framework. Consequently, the term ‘azo ligand’ encompasses a broad family of coordination environments rather than a single coordination motif [2, 3].

Metal binding may modify the absorption spectrum, charge distribution, redox potentials, photochemical behavior, thermal stability, solubility, and intermolecular interactions of the parent ligand. Importantly, the azo group can behave as a redox-active or redox-non-innocent component in selected complexes, accepting electron density and participating in metal–ligand cooperativity. These properties have stimulated research in catalysis, molecular materials, sensors, biological chemistry, and photoresponsive systems [3–7].

Although numerous individual azo–metal complexes have been reported, much of the literature is organized around individual syntheses and biological assays. A useful review therefore requires more than listing complexes: it should connect donor-atom arrangement and denticity with coordination geometry, electronic structure, stability, and function. The present review adopts this structure–property–function approach and emphasizes both the opportunities and the limitations of azo–metal systems.

2. Coordination Chemistry and Classification of Azo Ligands

Classification according to denticity is useful because it provides a first approximation of the number of metal–donor interactions available to a ligand. However, denticity alone does not determine the final geometry. Chelate-ring size, ligand preorganization, donor-atom basicity, metal-ion preferences, steric effects, protonation state, counterions, and solvent can all alter the observed structure. Accordingly, the following classification should be regarded as a structural framework rather than a rigid prediction of complex geometry as shown in Table 1.

Table 1: Classification of azo ligands based on denticity, donor atom sets, and coordination behavior

Class	Typical donor set	Representative coordination behavior	Examples in this review
Monodentate	N (often azo or heterocyclic N)	One localized donor atom; often supported by additional ligands/counterions	Ag(I)–2-(phenylazo)imidazole [8]
Bidentate	N,O; N,N; N,S	Five- or six-membered chelate rings are common	Phenolic and hydroxyquinoline-derived systems [9, 10]
Tridentate	N,N,O; N,O,O; N, N,N	Three donor atoms cooperate around one metal center	Naphthaldehyde and nitrothiazole azo ligands [11, 12]
Tetradentate	N,N,O,O and related sets	Multiple chelate rings; geometry strongly depends on metal and ligand preorganization	Oxydianiline-derived azo ligands [13]

2.1. Monodentate Azo Ligands

Monodentate coordination is less frequently encountered than chelating coordination because many azo ligands contain additional donor groups positioned to favor intramolecular metal binding. Nevertheless, monodentate behavior is well documented when the ligand architecture, steric environment, or electronic properties favor a single donor site. A representative example is 2-(phenylazo)imidazole, which coordinates Ag(I) through the imidazole nitrogen (Figure 1 [8]). The reported complex forms a discrete linear Ag(I) environment with a 1:2 metal-to-ligand stoichiometry [8].

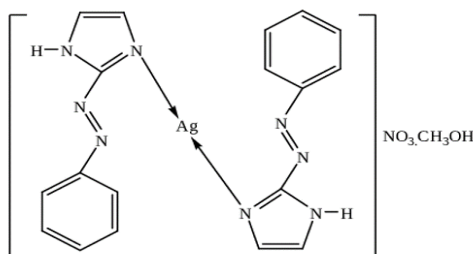


Figure 1: Representative Ag(I) complex containing a monodentate azo-derived ligand

2.2. Bidentate Azo Ligands

Bidentate azo ligands constitute one of the most widely represented classes. A common motif is N,O coordination in which an azo nitrogen and an adjacent phenolic oxygen form a chelate ring. Deprotonation of an ortho-hydroxyl group can strengthen metal binding and increase ligand-field control. In other systems, two nitrogen donors—for example an azo nitrogen and a heterocyclic nitrogen—can generate N,N coordination. The resulting geometry is determined by the metal ion and the remaining coordination sites.

Abdallah reported Ni(II) and Cu(II) complexes of substituted azo ligands derived from 4-acetamidophenol, illustrating N,O coordination by the azo nitrogen and phenolic oxygen. The complexes were assigned octahedral environments containing additional chloride and/or aqua ligands [9]. Similarly, azo-functionalized 8-hydroxyquinoline derivatives have been investigated with Zn(II), Cd(II), and Hg(II), demonstrating that the condensed heterocyclic donor framework can provide a different bidentate environment and influence the resulting stoichiometry and geometry [10]. Representative bidentate azo ligands and their corresponding transition-metal complexes are illustrated in Figures 2, 3, and 4 [9, 10].

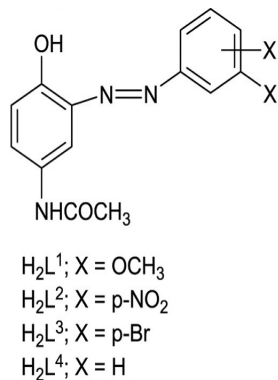


Figure 2: Representative phenolic bidentate azo ligand. Adapted from Ref. 4.

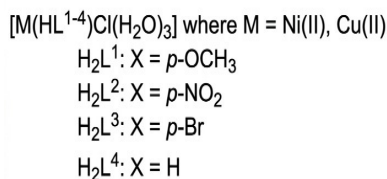
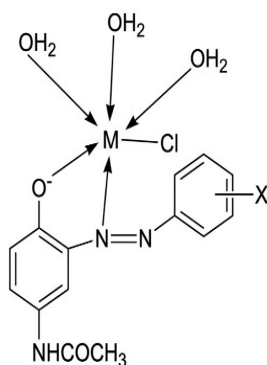


Figure 3: Representative N,O-coordinated bidentate azo complex. Adapted from Ref. 4.

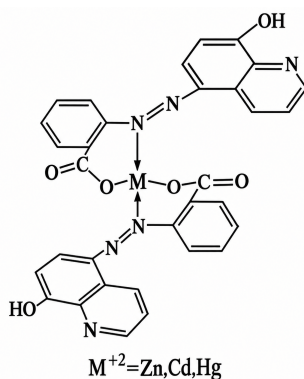


Figure 4: Representative complexes of an azo-functionalized 8-hydroxyquinoline ligand with Zn(II), Cd(II), and Hg(II). Adapted from Ref. 5.

2.3. Tridentate Azo Ligands

Tridentate azo ligands contain three donor atoms arranged so that a single metal center can interact with a compact multidentate framework. Common donor combinations include N,N,O, N,O,O, and N,N,N. Such ligands can impose a more defined coordination environment than bidentate analogues, although the final geometry still depends on metal coordination number and auxiliary ligands.

Ahmed and Yousif reported a tridentate azo ligand derived from 4-nitroaniline and 2-hydroxy-1-naphthaldehyde and investigated its Cr(III), Mn(II), Co(II), Ni(II), and Cu(II) complexes. Spectroscopic, analytical, and magnetic data were used to support the proposed coordination structures [11]. Al-Khateeb and co-workers similarly investigated a heterocyclic [N,N,O] azo ligand derived from 2-amino-5-nitrothiazole and 3-methyl-4-nitrophenol with several metal ions [12]. These examples demonstrate how incorporation of heterocyclic nitrogen and phenolic oxygen can expand coordination options beyond simple azo-N binding. Representative tridentate azo ligands and their corresponding heterocyclic transition-metal complexes are illustrated in Figures 5 and 6 [11, 12].

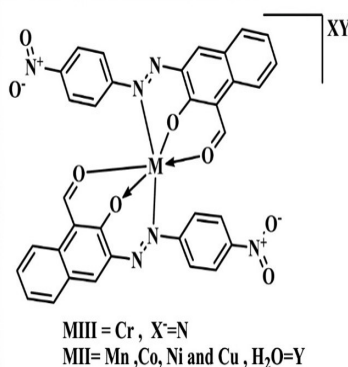


Figure 5: Representative tridentate azo-metal complex. Adapted from Ref. 6.

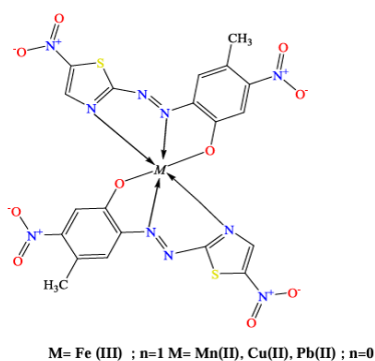


Figure 6: Representative heterocyclic tridentate azo-metal complex. Adapted from Ref. 7.

2.4. Tetradentate Azo Ligands

Tetradentate azo ligands provide four donor atoms to a metal center and can generate multiple chelate rings. Their enhanced preorganization may increase complex stability, but tetradentate coordination should not be equated automatically with macrocyclic coordination: a tetradentate ligand may be acyclic or macrocyclic. The stability of a tetradentate complex depends on donor-atom arrangement, chelate-ring size, ligand rigidity, metal–ligand matching, and the presence of competing solvent or anionic ligands.

Deghadi and co-workers reported tetradentate azo-dye complexes derived from 4,4'-oxydianiline and 2-amino-4-chlorophenol with Cr(III), Mn(II), Fe(III), Co(II), Ni(II), Cu(II), and Cd(II). The ligand was assigned an N,N,O,O donor set, and multiple analytical and spectroscopic methods were used to investigate the resulting complexes. Biological testing was also reported, illustrating the multifunctional character frequently sought in azo-metal systems [13].

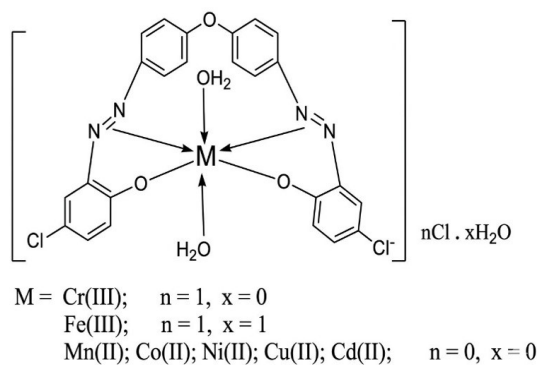


Figure 7: Representative tetradentate azo-metal complex. Adapted from Ref. 8.

3. Factors Governing Coordination Behavior and Structure–Property Relationships

The coordination behavior of azo ligands is controlled by a combination of ligand-centered and metal-centered factors. A useful interpretation therefore considers donor atoms, protonation state, electronic substituent effects, steric constraints, metal oxidation state, coordination preferences, and the solution environment together as shown in Table 2.

Table 2: Structural and chemical factors governing coordination behavior and structure property relationships of azo ligands

Structural/chemical factor	Expected influence	Important caveat
Denticity	Controls number of direct donor interactions and potential chelate rings	Higher denticity does not guarantee a single geometry or higher activity
Donor-atom identity	N, O and S differ in basicity, hardness/softness and bonding preferences	Actual donor strength depends on protonation and substituent effects
Ortho-OH / carboxylate	Can enable N,O or O,O chelation after deprotonation	pH and competing ligands can change the dominant species
Heterocyclic N	Provides additional directional coordination sites	Steric and electronic effects can favor one N over another
Metal identity/oxidation state	Controls coordination number, geometry, ligand-field effects and redox behavior	Complex stability and biological behavior may change substantially between metals
Substituent electronics	Modifies azo π -system, donor strength and optical/redox properties	Effects are not always monotonic across different metals
Solvent/counterion/pH	Can alter protonation, aggregation and speciation	Solid-state structures may not represent the dominant solution species

One important feature that is sometimes overlooked is the redox non-innocence of coordinated azo groups. Selected azo ligands can accept electron density from a metal center or from external reducing agents, producing ligand-centered radical or reduced states. This behavior provides an additional mechanism by which metal and ligand properties become coupled and can be exploited in electron-transfer catalysis and molecular materials [3, 4].

Photoisomerization is another important structure–property relationship. Azo groups can undergo E/Z isomerization under appropriate irradiation, and coordination to a metal center may modify the energetics and kinetics of this process. Recent studies of photoresponsive metal complexes emphasize that coordination can be used to tune absorption, emission, photoisomerization rates, and redox response, supporting applications in molecular switches and optoelectronic materials [5, 6].

4. Biological and Pharmaceutical Applications

Azo-metal complexes have attracted extensive interest as experimental antimicrobial, antifungal, antioxidant, and anticancer systems. However, biological activity should be interpreted at the level of the individual complex rather than assumed to be an intrinsic consequence of metal coordination. Activity depends on complex charge, stability, speciation in biological media, metal-ion lability, lipophilicity, ligand structure, redox properties, and the biological target [2, 14–16].

Al-Gaber and co-workers evaluated a series of Cr(III), Mn(II), Fe(III), Co(II), Ni(II), Cu(II), Zn(II), and Cd(II) complexes derived from a heterocyclic azo ligand. The study reported antimicrobial testing against Gram-positive and Gram-negative bacteria and fungi and found that several metal complexes showed greater inhibition than the free ligand under the experimental conditions used [14]. Such comparisons are informative, but they should be reported together with assay conditions, concentrations, controls, and where available MIC or IC50 values.

Similarly, Mohammed et al. reported Mn(II) and Zn(II) complexes of a benzimidazole/indole-containing azo ligand and evaluated their antifungal properties [15]. Gajiwala et al. reported pyrazolone-derived transition-metal complexes and observed antifungal activity, with Cu(II) and Co(II) complexes showing notable activity against *Candida albicans* in their experimental system [16]. These studies illustrate the recurring observation that complexation can alter biological performance, but they do not establish a universal superiority of metal complexes over their parent ligands. A concise summary of the reported in vitro biological evaluations, including antimicrobial, antifungal, and antiproliferative activities of the representative azo–metal complexes, is presented in Table 3.

Table 3: Selected biological studies discussed in this review. The table summarizes reported observations rather than ranking the complexes; direct comparison is limited by differences in assay conditions and endpoints.

Study	Metal/complex	Biological model	Main observation	Ref.
Al-Gaber et al. (2023)	Cr, Mn, Fe, Co, Ni, Cu, Zn, Cd complexes	Bacteria and fungi	Several complexes showed greater inhibition than the free ligand under the reported assay conditions.	[14]
Mohammed et al. (2022)	Mn(II), Zn(II) complexes	Aspergillus niger	Both free ligand and complexes were investigated; complexation altered antifungal performance.	[15]
Gajiwala et al. (2026)	Cu(II), Co(II), Ni(II), Cr(II), Fe(II) complexes	Candida albicans; Aspergillus niger	Cu(II) and Co(II) complexes showed notable antifungal activity in the reported experiments.	[16]
Mishra et al. (2025)	Cu(II), Co(II), Ni(II), Zn(II) complexes	MCF-7 and related assays	A selected Ni(II) complex showed notable in-vitro antiproliferative activity; docking was used as a mechanistic hypothesis.	[17]

4.1. Possible Mechanisms of Biological Action

Several mechanisms have been proposed for azo-metal complexes, including membrane interaction, enzyme inhibition, DNA association, reactive oxygen species generation, and metal-dependent disruption of cellular redox processes. A recent review specifically focused on azo dyes and metal complexes summarizes DNA interaction, enzyme inhibition, membrane disruption, and reactive oxygen species generation as possible antimicrobial mechanisms [2]. These mechanisms should be regarded as complementary hypotheses whose importance varies with complex structure.

Tweedy's chelation theory provides one classical explanation for increased activity after complexation: coordination can redistribute electron density and reduce the effective polarity of the metal center, potentially increasing the lipophilic character of the complex and facilitating membrane partitioning. Nevertheless, this model should not be used as the sole explanation for biological activity. Metal-ion release, ligand exchange, redox chemistry, protein binding, ROS production, and specific target interactions can also contribute [2, 7].

DNA binding is another frequently investigated property. Planar aromatic ligands may interact with DNA through intercalation or groove association, while some metal complexes can interact more directly with nucleobase donor atoms. Because these possibilities cannot be distinguished from docking or UV–Vis changes alone, mechanistic claims should ideally be supported by complementary experiments such as viscosity measurements, fluorescence displacement, circular dichroism, electrophoretic mobility assays, DNA cleavage experiments, or direct structural methods.

For anticancer studies, statements about therapeutic efficacy should remain proportional to the evidence. In vitro cytotoxicity, even when accompanied by molecular docking or mechanistic assays, does not by itself demonstrate selectivity, pharmacokinetic suitability, safety, or clinical potential. Future studies should therefore prioritize normal-cell controls, mechanistic biomarkers, stability in physiological media, serum-protein interactions, metal-release behavior, and in vivo validation.

5. Optical, Photoresponsive, and Optoelectronic Applications

The extended π -conjugation of azo ligands makes them attractive for optical materials, while metal coordination provides an additional means of tuning electronic transitions. Coordination can modify ligand-centered $\pi \rightarrow \pi^*$ and $n \rightarrow \pi^*$ transitions and may introduce metal-to-ligand or ligand-to-metal charge-transfer contributions. These changes can affect absorption wavelength, extinction coefficient, emission behavior, redox response, and photoisomerization.

Early studies investigated metal–azo complexes as recording materials for optical storage, including DVD-R thin films [18, 19]. These studies are historically important, but modern research has moved toward broader photoresponsive and optoelectronic applications, including molecular switches, sensors, electrochromic systems, and light-responsive functional materials [5, 6, 20]. Recent reviews emphasize the relationship between ligand substitution, conjugation, metal coordination, charge-transfer pathways, and photoisomerization behavior [6, 20].

Azo–metal systems are particularly attractive for stimuli-responsive materials because their optical response can be influenced by light, redox conditions, pH, and coordination environment. A 2026 review highlights emerging applications in biosensing, bioimaging, drug delivery, and optoelectronic devices, while also identifying stability, similarity between systems, and scalability as practical challenges [6].

6. Catalysis and Green Chemistry

The catalytic chemistry of azo–metal complexes deserves separate treatment from the use of metal-containing catalysts in azo-dye synthesis. In selected coordination compounds, the azo ligand itself can behave as a redox-active component and participate in electron transfer. Recent work has shown that redox-non-innocent azo ligands can act as electron reservoirs, enabling metal–ligand cooperativity in catalytic oxidation and related transformations [3, 4].

Swatiputra and co-workers reviewed first-row transition-metal complexes containing redox-active azo functions and described their use in electron-transfer catalysis. The proposed catalytic pathways include ligand-centered electron uptake followed by proton-coupled electron-transfer or hydrogen-atom-transfer steps, demonstrating that the azo group can contribute directly to catalytic reactivity rather than merely serving as a spectator ligand [4].

Green chemistry claims should nevertheless be supported by quantitative metrics such as catalyst loading, turnover number, turnover frequency, yield, selectivity, energy demand, solvent burden, and recyclability. The use of a metal complex alone does not make a reaction 'green'; sustainability depends on the complete process.

7. Sensing, Analytical, and Energy-Related Applications

Azo chromophores have long been used as colorimetric and spectrophotometric reagents, and metal coordination can amplify or shift their optical responses. This makes azo–metal systems relevant to ion sensing and chemically responsive materials. Recent work increasingly combines coordination chemistry with nanomaterials, gels, and other matrices to improve stability, signal transduction, and practical deployment [6].

Energy-related applications are also emerging, particularly where azo photoisomerization, charge transfer, and redox activity can be coupled to device function. However, examples involving generic copper or cobalt coordination complexes should not automatically be classified as azo–metal applications unless the azo ligand is an integral part of the reported material. For a review specifically focused on azo–metal complexes, this distinction is essential for maintaining a clear scope.

8. Computational and Spectroscopic Tools for Azo–Metal Complexes

Modern characterization increasingly combines experimental spectroscopy with computational chemistry. FT-IR can provide evidence for changes in azo and auxiliary donor vibrations, while UV–Vis spectroscopy can reveal ligand-centered and charge-transfer transitions. $^1\text{H}/^{13}\text{C}$ NMR is informative for diamagnetic complexes, whereas magnetic susceptibility and EPR can provide important information for paramagnetic systems. Mass spectrometry and elemental analysis support molecular composition, while single-crystal X-ray diffraction remains the most direct method for establishing coordination geometry when suitable crystals are available.

DFT calculations can complement experimental observations by examining optimized geometries, charge distribution, frontier molecular orbitals, spin states, and relative stability. TD-DFT can be particularly useful for interpreting electronic transitions and photoresponsive behavior. For biological studies, molecular docking can generate hypotheses about target interactions, but docking scores alone should not be treated as experimental binding constants or proof of mechanism. Where feasible, computational predictions should be linked to experimental assays and, for dynamic systems, molecular dynamics or free-energy approaches.

9. Challenges and Future Perspectives

Despite the broad literature on azo–metal complexes, several challenges limit comparison between studies. First, structural assignment is sometimes based primarily on spectroscopic shifts without crystallographic confirmation. Second, biological studies often use different concentrations, organisms, endpoints, and controls, making quantitative comparison difficult. Third, the stability and speciation of complexes in aqueous or physiological media are not always established. Fourth, claims of enhanced activity are sometimes attributed to chelation without experimentally distinguishing membrane permeability, ROS generation, metal release, enzyme inhibition, or DNA interactions.

Future research should therefore move toward mechanism-guided design. Ligand denticity, donor-atom arrangement, metal identity, oxidation state, and substituent electronics should be varied systematically rather than independently. Biological investigations should include free-ligand and metal-salt controls, normal-cell toxicity, stability measurements, and mechanistic assays. For materials applications, standardized photophysical, electrochemical, thermal, and cycling measurements are needed to permit meaningful comparison between systems.

Another important direction is the integration of computational and experimental workflows. DFT/TD-DFT can be used to predict electronic structure and optical transitions; docking can suggest possible biological targets; and higher-level simulations can test stability and dynamic behavior. Such approaches are most valuable when used to formulate experimentally testable hypotheses rather than as substitutes for experimental validation.

Finally, the field would benefit from greater attention to scalable synthesis, solvent selection, metal sustainability, toxicity, and end-of-life considerations. The most useful next-generation azo–metal complexes will likely be those in which coordination chemistry, photophysics, biological function, and practical processability are optimized together.

10. Conclusions

Azo–metal complexes form a chemically diverse family in which the azo chromophore and the metal center can mutually modify structure and electronic behavior. Monodentate, bidentate, tridentate, and tetradentate systems demonstrate that donor-atom arrangement and ligand preorganization are central determinants of coordination geometry. Metal identity, oxidation state, protonation, counterions, and solvent further influence the final structure and properties.

The functional behavior of these complexes extends from biological activity to optical and photoresponsive materials, catalysis, sensing, and emerging energy-related applications. However, enhanced activity or performance should not be assumed solely from metal coordination. Mechanistic interpretation requires consideration of speciation, stability, redox chemistry, membrane transport, target interactions, and experimental context.

The future development of azo–metal chemistry therefore depends on stronger structure–property–function correlations, standardized biological and materials testing, and closer integration of spectroscopy, crystallography, electrochemistry, and computational chemistry. These approaches can move the field from the preparation of individual complexes toward rationally designed systems with reproducible and application-relevant performance.

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