

Review

Auxiliary Ligand Effects on Metal Coordination Polymer Properties

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Abstract: Coordination polymers have emerged as versatile materials with tunable structural and functional properties that can be systematically modified through rational design strategies. The incorporation of auxiliary ligands alongside primary bridging ligands represents a powerful synthetic approach for controlling the dimensionality, topology, and physicochemical characteristics of these materials. This review examines how auxiliary ligands influence the structural assembly and functional properties of metal coordination polymers, with particular emphasis on multicarboxylate-based systems. The discussion encompasses synthetic methodologies, structural diversity, and property modulation including magnetic behavior, luminescence, catalytic activity, and biological applications. Through analysis of recent developments in mixed-ligand coordination polymer design, this work demonstrates how secondary ligands containing nitrogen-donor atoms, particularly imidazole derivatives, can direct framework formation and enhance material performance. The interplay between metal centers, primary carboxylate ligands, and auxiliary nitrogen-containing ligands creates opportunities for developing coordination polymers with predetermined structures and optimized properties for applications in energy storage, catalysis, sensing, and biomedicine.

Keywords: coordination polymers; auxiliary ligands; multicarboxylate ligands; mixed-ligand synthesis; metal-organic frameworks; structure-property relationships

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1. Introduction

Coordination polymers represent an important class of crystalline materials formed through the self-assembly of metal ions or clusters with organic bridging ligands, creating extended network structures with diverse dimensionalities. While their fundamental chemistry has been widely studied, a key area of current research focuses on applying these versatile materials in electrocatalysis. Their highly tunable porosity, vast internal surface areas, and well-defined active sites make them exceptional platforms for designing efficient electrocatalysts for reactions such as oxygen evolution, hydrogen evolution, and the tandem electrocatalytic reduction of CO₂ to high-value C₂⁺ products [1]. These materials have attracted substantial research attention due to their structural versatility and potential applications across multiple domains including gas storage, separation, catalysis, sensing, and drug delivery. The rational design of coordination polymers requires careful consideration of multiple factors including metal ion selection, ligand geometry and functionality, reaction conditions, and synthetic methodology [2, 3].

The mixed-ligand synthetic strategy has proven particularly effective for controlling coordination polymer architecture and properties through the deliberate combination of

primary bridging ligands with secondary auxiliary ligands [4]. Multicarboxylate ligands serve as excellent primary building blocks due to their diverse coordination modes, strong metal-binding affinity, and ability to bridge multiple metal centers, while auxiliary ligands containing nitrogen-donor groups provide additional structural direction and functional enhancement [5]. The systematic investigation of auxiliary ligand effects enables researchers to establish structure-property relationships and develop predictive design principles for targeted material synthesis.

Recent advances in coordination polymer chemistry have demonstrated that auxiliary ligands exert profound influence on framework topology, dimensionality, pore characteristics, and physical properties through multiple mechanisms including coordination site occupation, hydrogen bonding interactions, and electronic effects [6, 7]. Understanding these influences is essential for advancing coordination polymer design from empirical observation toward rational engineering of materials with predetermined structures and optimized performance characteristics. This review examines current knowledge regarding auxiliary ligand effects on coordination polymer properties, with emphasis on structural modulation, property tuning, and emerging applications.

2. Synthetic Strategies and Design Principles

2.1. Mixed-Ligand Synthetic Approach

The mixed-ligand synthetic strategy involves the simultaneous use of two or more different organic ligands during coordination polymer assembly, typically combining a primary multicarboxylate bridging ligand with a secondary nitrogen-containing auxiliary ligand [4]. This approach provides enhanced control over structural outcomes compared to single-ligand synthesis by introducing additional geometric constraints and coordination preferences that influence metal ion coordination environments and network topology. The selection of appropriate ligand combinations requires consideration of coordination preferences, geometric compatibility, and desired functional properties.

Multicarboxylate ligands containing multiple carboxylic acid groups separated by aromatic or aliphatic spacers function as primary structural building blocks that bridge metal centers and direct overall framework dimensionality [8]. These ligands exhibit remarkable coordination versatility through different deprotonation states and diverse binding modes including monodentate, chelating, and bridging configurations. The flexibility or rigidity of the organic backbone connecting carboxylate groups further influences structural outcomes, with semi-rigid ligands offering balanced control between structural predictability and adaptive framework formation. Table 1 summarizes common multicarboxylate ligand characteristics relevant to coordination polymer synthesis.

Table 1. Characteristics of Multicarboxylate Ligands in Coordination Polymer Synthesis.

Ligand Feature	Structural Impact	Property Influence
Carboxylate number	Connectivity and dimensionality	Framework stability and porosity
Backbone flexibility	Adaptive coordination geometry	Structural diversity and guest accommodation
Aromatic spacers	Rigid framework formation	Luminescence and electron transfer
Aliphatic linkers	Framework flexibility	Dynamic behavior and responsiveness
Functional substituents	Additional coordination sites	Enhanced functionality and selectivity

2.2. Auxiliary Ligand Selection and Function

Auxiliary ligands typically contain nitrogen-donor atoms within heterocyclic structures such as imidazole, pyridine, or triazole derivatives that complement the primary carboxylate ligands through distinct coordination preferences and geometric constraints [9,10]. These secondary ligands influence coordination polymer assembly

through multiple mechanisms including terminal coordination site occupation, pillaring between metal-carboxylate layers, and participation in secondary bonding interactions. The geometric characteristics of auxiliary ligands, particularly angular configurations in V-shaped or bent structures, provide directional control over framework extension and pore architecture.

Imidazole-based auxiliary ligands have proven particularly effective due to their moderate basicity, versatile coordination modes, and ability to engage in hydrogen bonding interactions that stabilize framework structures [6,10]. The nitrogen atoms within imidazole rings can coordinate metal centers in terminal or bridging modes, while the NH groups can participate in hydrogen bonding networks that reinforce structural integrity. Variation of substituents on the imidazole core allows systematic tuning of electronic properties and steric effects, providing additional control over coordination polymer characteristics. The selection of auxiliary ligand dimensions and geometry must be matched to the spacing and connectivity patterns established by the primary multicarboxylate ligand to achieve coherent framework assembly.

2.3. Reaction Conditions and Solvent Effects

Solvothermal and hydrothermal synthesis methods predominate in coordination polymer preparation due to their ability to provide elevated temperatures and pressures that facilitate dissolution of reactants, overcome kinetic barriers, and promote crystallization of thermodynamically stable products. The choice of solvent system significantly impacts coordination polymer formation by influencing metal ion solvation, ligand deprotonation equilibria, and framework assembly kinetics [7]. Mixed solvent systems containing water combined with organic solvents such as dimethylformamide, ethanol, or acetonitrile are frequently employed to optimize solubility and reactivity of all components while modulating crystallization rates.

Solvent molecules can participate directly in coordination polymer structures through metal coordination or incorporation within framework voids, where they may influence topology, stability, and properties. Different solvent environments can drive formation of distinct structural polymorphs from identical reactant combinations, demonstrating the critical role of reaction medium in determining structural outcomes [7]. Temperature, pH, metal-to-ligand ratios, and reaction duration represent additional synthetic variables that must be optimized for each system to achieve desired structural and compositional characteristics. The interplay between solvent effects and auxiliary ligand influences adds complexity to coordination polymer synthesis but also provides opportunities for sophisticated structural control through systematic variation of reaction conditions.

3. Structural Modulation by Auxiliary Ligands

3.1. Dimensionality Control and Topology

Auxiliary ligands exert profound influence on coordination polymer dimensionality through their coordination modes, geometric characteristics, and steric requirements. The incorporation of nitrogen-containing auxiliary ligands can extend two-dimensional metal-carboxylate layers into three-dimensional frameworks through pillaring interactions, or alternatively terminate metal coordination sites to limit framework extension and favor formation of lower-dimensional structures [8, 9]. The geometric constraints imposed by auxiliary ligand shape and size direct framework topology by defining metal-metal distances, metal-ligand-metal angles, and overall connectivity patterns.

Studies examining coordination polymers constructed from identical multicarboxylate ligands with different metal centers have revealed how metal coordination preferences interact with auxiliary ligand characteristics to determine structural outcomes [8]. Transition metals with octahedral coordination geometries typically accommodate both carboxylate and nitrogen donor ligands in mixed coordination spheres, while metals preferring tetrahedral or square planar geometries exhibit more selective ligand binding that can drive specific topological outcomes. The

flexibility of multicarboxylate ligands allows adaptive conformational changes that accommodate auxiliary ligand coordination while maintaining efficient space filling and framework stability. Table 2 illustrates relationships between auxiliary ligand characteristics and resulting structural features in representative coordination polymer systems.

Table 2. Auxiliary Ligand Influence on Coordination Polymer Structure.

Auxiliary Ligand Type	Coordination Mode	Resulting Dimensionality	Topological Features
Linear N-donors	Terminal or bridging	1D to 3D	Extended chains or interpenetration
Angular N-donors	Chelating	2D layers	Corrugated or undulated sheets
V-shaped ligands	Bridging bidentate	3D frameworks	Pillared layer structures
Bulky N-heterocycles	Terminal	0D to 2D	Discrete complexes or limited extension
Flexible dipyridyl	Adaptive bridging	2D to 3D	Conformationally diverse networks

3.2. Metal Coordination Environment

The presence of auxiliary ligands directly modifies metal ion coordination environments by occupying coordination sites and influencing the number and arrangement of coordinated carboxylate groups. Mixed coordination spheres containing both carboxylate oxygen donors and nitrogen donors from auxiliary ligands create distinct electronic environments that affect metal center properties including oxidation state stability, magnetic behavior, and reactivity [11,12]. The trans influence and cis effect associated with different donor atoms influence the strength and character of metal-ligand bonds throughout the coordination sphere, with consequences for structural stability and functional properties.

Auxiliary ligands can enforce specific coordination geometries through their denticity, bite angles, and steric profiles, directing metal centers toward octahedral, square pyramidal, trigonal bipyramidal, or tetrahedral configurations [11]. These geometric preferences propagate throughout the coordination polymer structure, influencing framework topology and void space distribution. The ability of auxiliary ligands to bridge multiple metal centers or terminate coordination sites allows control over metal-metal distances and magnetic exchange pathways, which proves particularly important for designing materials with specific magnetic properties. Systematic variation of auxiliary ligand characteristics while maintaining constant primary ligands and metal centers enables isolation of auxiliary ligand effects on coordination environment and structure.

3.3. Framework Interpenetration and Porosity

Coordination polymer frameworks with large void spaces often exhibit interpenetration where multiple independent networks occupy the same volume, reducing porosity but potentially enhancing mechanical stability and enabling selective guest inclusion [13]. Auxiliary ligands influence interpenetration tendency through their size, shape, and coordination preferences, which affect the dimensions of individual framework cavities and the propensity for multiple network formation. Bulky auxiliary ligands can sterically inhibit interpenetration by occupying void spaces, while smaller auxiliary ligands may permit framework interpenetration or even facilitate it through favorable interactions between independent networks.

The judicious selection of auxiliary ligand dimensions matched to multicarboxylate ligand characteristics enables rational control over framework porosity and guest-

accessible volume. Non-interpenetrated frameworks with permanent porosity are desirable for applications in gas storage and separation, while controlled interpenetration may benefit mechanical properties and enable framework flexibility. The presence of auxiliary ligands within framework structures can create functional pore environments through incorporation of basic nitrogen sites, aromatic surfaces for π - π interactions, or hydrogen bonding groups that enhance host-guest interactions and enable selective molecular recognition. Understanding relationships between auxiliary ligand characteristics and interpenetration behavior is essential for designing coordination polymers with optimized porosity for specific applications.

4. Property Modulation and Functional Enhancement

4.1. Magnetic Properties

Coordination polymers incorporating paramagnetic metal ions such as copper, cobalt, nickel, or gadolinium exhibit diverse magnetic behaviors determined by metal-metal distances, metal-ligand-metal angles, and the magnitude of magnetic exchange interactions mediated through bridging ligands [7,12]. Auxiliary ligands influence magnetic properties through multiple pathways including modification of metal coordination geometries, alteration of metal-metal separations, and electronic effects that modulate orbital overlap and exchange coupling strengths. The incorporation of auxiliary ligands between magnetic metal centers can introduce additional exchange pathways or block direct exchange interactions, leading to antiferromagnetic, ferromagnetic, or more complex magnetic ordering.

Studies of cobalt-based coordination polymers have demonstrated how auxiliary ligands regulate antiferromagnetic coupling between metal centers through their influence on coordination geometry and bridging angles [7]. The semi-rigid nature of certain carboxylate ligands combined with auxiliary ligand constraints can create specific metal-ligand-metal angles that favor particular magnetic exchange mechanisms. Copper coordination polymers with imidazole-based auxiliary ligands exhibit magnetic behaviors ranging from weakly coupled paramagnetic systems to strongly antiferromagnetic chains depending on auxiliary ligand characteristics and resulting structural features [12]. Table 3 summarizes auxiliary ligand effects on magnetic properties in representative coordination polymer systems.

Table 3. Auxiliary Ligand Effects on Magnetic Properties.

Metal Center	Auxiliary Ligand Class	Magnetic Behavior	Exchange Coupling
Cu (II)	Imidazole derivatives	Antiferromagnetic	Moderate to strong
Co (II)	Pyridyl ligands	Antiferromagnetic	Weak to moderate
Ni (II)	Triazole compounds	Paramagnetic	Very weak
Gd (III)	N-donor ligands	Paramagnetic	Negligible
Mixed metal	Bridging N-donors	Complex ordering	Variable

4.2. Luminescence and Sensing Applications

Luminescent coordination polymers combine the structural versatility of metal-organic frameworks with photophysical properties useful for sensing, display technologies, and bioimaging [2, 3]. The incorporation of auxiliary ligands containing aromatic nitrogen heterocycles introduces chromophoric groups that can participate in energy transfer processes with metal centers or primary ligands, enhancing or modifying luminescence characteristics. Zinc and cadmium coordination polymers with multicarboxylate and nitrogen-donor auxiliary ligands exhibit tunable emission colors and quantum yields depending on ligand combinations and structural features [14].

The rigid aromatic structures within auxiliary ligands reduce non-radiative decay pathways and enhance luminescence efficiency through restriction of molecular motions in the solid state. Energy transfer from organic ligands to lanthanide metal centers represents an important mechanism for generating characteristic lanthanide emission,

with auxiliary ligands serving as antenna chromophores that absorb excitation energy and transfer it to metal centers through sensitization processes [14]. Coordination polymers incorporating multiple ligand types offer opportunities for developing white light emission through combination of different emission colors, and for sensing applications where guest molecules selectively quench or enhance luminescence through specific interactions with framework components. The diversity of nitrogen-containing auxiliary ligands enables systematic tuning of photophysical properties for targeted applications [15].

4.3. Catalytic and Biological Activities

The open coordination sites, high surface areas, and compositional tunability of coordination polymers make them attractive platforms for heterogeneous catalysis, with auxiliary ligands providing opportunities to introduce additional catalytic functionality or modify active site environments [1, 5]. Metal centers within coordination polymer frameworks can serve as Lewis acid catalysts for various organic transformations, while auxiliary ligands containing basic nitrogen sites may facilitate bifunctional catalysis through cooperative acid-base mechanisms. The spatial arrangement of metal centers and functional groups within coordination polymer structures enables substrate pre-organization and transition state stabilization that enhance catalytic efficiency and selectivity.

Recent developments have explored coordination polymers as electrocatalysts for carbon dioxide reduction, where dual-metal sites and carefully designed ligand environments promote tandem catalytic mechanisms that enable selective formation of multi-carbon products [1]. The incorporation of auxiliary ligands can tune the electronic properties of metal centers and create microenvironments that stabilize reaction intermediates and direct product selectivity. Coordination polymers have also demonstrated biological activities including enzyme inhibition, with copper-based systems incorporating specific auxiliary ligands showing high efficiency as urease inhibitors [6, 10]. The V-shaped geometry of certain auxiliary ligands appears particularly effective for creating coordination polymer structures with enhanced biological activity, suggesting that geometric factors beyond simple chemical composition influence biomolecular interactions. Table 4 presents examples of auxiliary ligand effects on coordination polymer functionality.

Table 4. Functional Properties Influenced by Auxiliary Ligands.

Application Domain	Property Enhanced	Auxiliary Ligand Role	Representative System
Electrocatalysis	CO ₂ reduction selectivity	Electronic modulation	Dual-metal frameworks
Energy storage	Charge capacity	Structural stability	Zinc-based polymers
Enzyme inhibition	Urease activity suppression	Geometric optimization	Copper-imidazole systems
Luminescence	Quantum yield	Antenna effect	Lanthanide networks
Magnetism	Exchange coupling	Pathway mediation	Transition metal chains

5. Structural Characterization and Analysis Methods

5.1. Crystallographic Techniques

Single crystal X-ray diffraction represents the definitive method for determining coordination polymer structures, providing complete atomic-level information about framework connectivity, metal coordination environments, ligand conformations, and packing arrangements. The ability to obtain high-quality single crystals suitable for diffraction analysis depends on careful optimization of synthetic conditions including solvent selection, temperature programming, and crystallization rates. Powder X-ray diffraction serves as a complementary technique for phase identification, purity

assessment, and structural characterization when single crystals are unavailable, though it provides less detailed structural information than single crystal methods.

Crystallographic analysis reveals how auxiliary ligands influence framework topology through their coordination modes and spatial arrangements, enabling correlation of structural features with synthetic conditions and ligand characteristics [13, 14]. The flexibility of multicarboxylate ligands manifests in their observed conformations within different coordination polymer structures, while auxiliary ligand orientations and coordination modes can be directly determined from crystal structures. Bond distances and angles obtained from crystallographic refinement provide quantitative information about metal-ligand interactions and coordination geometry that correlates with electronic and magnetic properties. Analysis of hydrogen bonding patterns and weak interactions captured in crystal structures illuminates secondary structural influences that contribute to framework stability and properties.

5.2. Spectroscopic Characterization

Infrared spectroscopy provides valuable information about ligand coordination modes through characteristic shifts in carboxylate stretching frequencies and appearance of metal-ligand vibrations not present in free ligands. The difference between asymmetric and symmetric carboxylate stretching frequencies indicates whether carboxylate groups bind metals in monodentate, chelating, or bridging modes, complementing crystallographic coordination mode assignments. The presence of auxiliary ligand characteristic vibrations in coordination polymer spectra confirms their incorporation into framework structures, while shifts in these frequencies reflect electronic perturbations from metal coordination.

Nuclear magnetic resonance spectroscopy of diamagnetic coordination polymers can provide insights into ligand environments and dynamics, particularly for systems with sufficient solubility or through solid-state NMR techniques. Electronic absorption and emission spectroscopy characterize optical properties essential for luminescent coordination polymers, revealing energy transfer processes, excited state lifetimes, and quantum yields. Electron paramagnetic resonance spectroscopy of paramagnetic coordination polymers provides information about metal oxidation states, coordination geometries, and magnetic interactions between metal centers. The complementary application of multiple spectroscopic techniques alongside crystallographic methods enables comprehensive characterization of coordination polymer structures and properties.

5.3. Thermal and Morphological Analysis

Thermogravimetric analysis coupled with differential scanning calorimetry reveals coordination polymer thermal stability, guest molecule content, and decomposition pathways through monitoring of mass loss and thermal events as a function of temperature. The thermal behavior of coordination polymers reflects framework stability and metal-ligand bond strengths, with auxiliary ligands influencing decomposition temperatures through their electronic effects and structural roles. Sequential mass loss steps often correspond to loss of solvent molecules, decomposition of auxiliary ligands, and finally degradation of primary framework components, providing insights into relative bond strengths and structural hierarchy.

Scanning electron microscopy and transmission electron microscopy visualize coordination polymer crystal morphologies, particle sizes, and surface features that influence properties and applications. The crystal habits exhibited by coordination polymers reflect their internal symmetry and growth conditions, with auxiliary ligands potentially influencing preferred growth directions and resulting morphologies. Gas adsorption measurements using nitrogen or other probe molecules quantify permanent porosity, surface areas, and pore size distributions for coordination polymers with guest-accessible void spaces. Table 5 summarizes characterization techniques and the structural or property information they provide for coordination polymer analysis.

Table 5. Characterization Methods for Coordination Polymers.

Technique	Information Obtained	Auxiliary Ligand Insights
Single crystal XRD	Complete structure	Coordination modes and topology
Powder XRD	Phase purity	Structural changes with conditions
Infrared spectroscopy	Functional groups	Coordination mode confirmation
Luminescence spectroscopy	Emission properties	Energy transfer mechanisms
Thermogravimetric analysis	Thermal stability	Decomposition sequence
Electron microscopy	Morphology	Crystal habit and particle size
Gas adsorption	Porosity	Framework interpenetration effects
Magnetic measurements	Exchange coupling	Metal-metal interaction pathways

6. Conclusion

The incorporation of auxiliary ligands in coordination polymer synthesis provides powerful control over structural assembly and functional properties through their influences on metal coordination environments, framework topology, dimensionality, and electronic characteristics. The mixed-ligand synthetic strategy combining multicarboxylate primary ligands with nitrogen-containing auxiliary ligands has proven particularly effective for generating coordination polymers with predetermined structures and optimized properties for diverse applications. Auxiliary ligands function through multiple mechanisms including coordination site occupation, geometric constraint imposition, electronic modulation, and secondary interaction participation that collectively determine structural outcomes and material performance.

Systematic investigation of auxiliary ligand effects has established structure-property relationships that guide rational coordination polymer design, though significant opportunities remain for developing more sophisticated predictive capabilities and exploring new ligand combinations. The sensitivity of coordination polymer structures to subtle variations in auxiliary ligand characteristics underscores both the challenges and opportunities in this field, as minor modifications can produce dramatic structural changes with corresponding property variations. Future developments will likely emphasize computational approaches for predicting structural outcomes, expansion of auxiliary ligand libraries to access new topologies and properties, and development of coordination polymers for emerging applications in energy conversion, environmental remediation, and precision medicine.

The continued advancement of coordination polymer chemistry requires integration of synthetic innovation, advanced characterization methods, and theoretical understanding to establish comprehensive design principles. The versatility of auxiliary ligands as structural and functional modulators ensures their continued importance in coordination polymer development, while emerging areas such as dynamic frameworks, stimulus-responsive materials, and multi-functional systems will benefit from sophisticated auxiliary ligand selection and design. Through systematic exploration of auxiliary ligand effects and their relationships to coordination polymer properties, researchers can develop increasingly sophisticated materials that address important technological and societal challenges.

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