

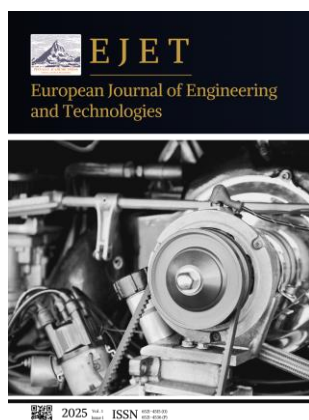
Review **Open Access**

Second Auxiliary Ligand Engineering in the Construction of Functional Copper Coordination Polymers

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Abstract: Copper coordination polymers (Cu-CPs) have garnered significant attention due to their versatile structures and broad applications in catalysis, sensing, and agriculture. Among various design strategies, the engineering of second auxiliary ligands has emerged as a crucial approach to modulate the structural dimensionality, stability, and functional properties of Cu-CPs. By incorporating diverse ligand types such as aromatic carboxylates, bipyridines, and V-shaped heterocyclic compounds, researchers have achieved precise control over framework architectures and enhanced exposure of catalytically active sites. This review comprehensively summarizes recent advances in second auxiliary ligand design, their impact on the structural features of Cu-CPs, and the resulting improvements in performance, particularly in urease inhibition for sustainable agriculture and catalytic applications. Key synthetic and characterization techniques are also discussed. The insights provided herein aim to guide future developments toward multifunctional Cu-CPs with tailored properties for emerging technological needs.

Keywords: copper coordination polymers; second auxiliary ligands; ligand engineering; urease inhibition; catalysis; sustainable agriculture; structural modulation

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1. Introduction

Copper coordination polymers (Cu-CPs) have attracted significant attention in recent years due to their unique structural diversity and versatile functional properties. These materials, formed by the coordination of copper ions with organic ligands, exhibit a wide range of applications spanning catalysis, chemical sensing, environmental remediation, and notably, agricultural practices such as urease inhibition for nitrogen use efficiency improvement [1]. The intrinsic redox activity of copper ions combined with the tunability of coordination environments enables the rational design of Cu-CPs with tailored physical and chemical characteristics. Such properties make them promising candidates for multifunctional applications where performance depends critically on precise control over their structure and stability.

Central to the design of Cu-CPs is the strategic selection and engineering of auxiliary ligands, which play a pivotal role in modulating the coordination geometry, framework topology, and ultimately, the material's functional behavior [2]. Auxiliary ligands can influence factors such as pore size, active site accessibility, and chemical stability, which are essential for enhancing catalytic efficiency or biological activity. Among these, the concept of "second auxiliary ligands" has recently emerged as a powerful approach to fine-tune the architecture and performance of Cu-CPs beyond the capabilities of primary ligands alone [3]. Second auxiliary ligands refer to additional coordinating molecules introduced

alongside the main ligands, often with distinctive geometries or functional groups that direct the formation of novel structural motifs.

The importance of second auxiliary ligand engineering lies in its ability to precisely regulate the formation of copper-based frameworks, enabling the construction of unique two-dimensional (2D) or higher-dimensional networks with improved exposure of active copper sites. This structural control translates directly into enhanced functional properties, such as increased urease inhibition activity for agricultural use or improved catalytic selectivity in chemical reactions. Recent studies have demonstrated the successful application of second auxiliary ligands in designing Cu-CPs with remarkable stability and superior inhibitory performance, highlighting the critical role of ligand engineering in advancing Cu-CPs research.

This review aims to provide a comprehensive overview of recent advances in the field of second auxiliary ligand engineering in Cu-CP construction. It focuses on the synthesis strategies, structural features, and functional applications of these materials, particularly emphasizing their role in urease inhibition and other catalytic processes. Furthermore, the review discusses characterization techniques used to elucidate structural details and evaluates the impact of ligand design on the performance and stability of Cu-CPs. Through this synthesis of current knowledge, we aim to identify key challenges and future opportunities for the rational design of functional Cu-CPs via second auxiliary ligand engineering, ultimately contributing to the development of more efficient and sustainable materials.

2. Fundamentals of Copper Coordination Polymers and Ligand Engineering (Approx. 500 words)

2.1. Overview of Copper Coordination Polymers

Copper coordination polymers (Cu-CPs) are a class of metal-organic frameworks formed by the coordination of Cu (II) ions with organic ligands, resulting in extended polymeric networks. The unique electronic configuration of Cu (II), which has a d9 electron arrangement, imparts variable coordination geometries that are highly sensitive to ligand environment, temperature, solvent, and synthetic conditions.

Common coordination geometries for Cu (II) centers include square planar, square pyramidal, and distorted octahedral configurations. For instance, in square planar geometry, Cu (II) typically coordinates with four donor atoms in a planar arrangement, which often leads to 1D chain or 2D layered structures. The square pyramidal geometry arises when an additional axial ligand weakly coordinates, expanding the dimensionality of the polymer. Octahedral coordination, involving six ligand donor atoms, is often responsible for 3D frameworks with higher connectivity and porosity [4].

These variations in coordination geometry profoundly affect the resulting Cu-CP architectures, leading to structural diversity ranging from one-dimensional (1D) linear chains, two-dimensional (2D) layered sheets, to three-dimensional (3D) frameworks with porous cavities. This structural versatility enables Cu-CPs to exhibit a broad spectrum of functional properties, such as catalytic activity, molecular recognition, gas adsorption, and ion exchange capabilities.

Moreover, the electronic properties of Cu (II) centers, modulated by ligand field strength and geometry, play a critical role in determining the catalytic behavior and stability of Cu-CPs. The interplay between coordination environment and ligand design thus forms the foundation for engineering Cu-CPs with tailored functionalities suitable for applications in catalysis, sensing, and agriculture.

2.2. Primary and Secondary (Second Auxiliary) Ligands

Ligands in Cu-CPs are generally divided into primary and secondary (or second auxiliary) types. Primary ligands form the main coordination framework by directly bonding

to copper centers, constructing the polymer backbone. Secondary ligands coordinate additionally to metal centers or bridging sites, modulating structural features such as dimensionality, porosity, and stability without disrupting the primary framework.

The influence of second auxiliary ligands is significant, as they enable fine-tuning of the metal coordination environment and overall topology, leading to optimized functional properties [5].

Table 1 summarizes representative Cu-CPs constructed with various second auxiliary ligands, showing the diversity in copper coordination geometries and resulting framework dimensionalities. This highlights how second auxiliary ligand engineering is crucial for tailoring Cu-CP structures and their applications.

Table 1. Representative Copper Coordination Polymers with Different Second Auxiliary Ligands.

Compound ID	Second Auxiliary Ligand	Cu (II) Coordination Geometry	Framework Dimensionality	Synthetic Method	Application Example
Cu-CP1	Aromatic Carboxylate	Octahedral	3D	Hydrothermal	Urease Inhibition
Cu-CP2	V-shaped Bipyridine	Square Pyramidal	2D	Solvothermal	Catalysis
Cu-CP3	Amide-based Ligand	Square Planar	1D	Hydrothermal	Environmental Remediation
Cu-CP4	Heterocyclic (Triazole)	Octahedral	2D	Solvothermal	Antimicrobial Applications

As demonstrated in Table 1, the selection of second auxiliary ligands profoundly affects the coordination geometry of Cu (II) centers and the overall framework dimensionality. This ligand-driven modulation directly impacts the physicochemical properties and functional performance of the Cu-CPs.

2.3. Ligand Properties and Synthetic Strategies

The successful design and synthesis of Cu-CPs rely heavily on the choice of ligands, whose physicochemical properties dictate the final polymeric structure and performance [6]. Ligand characteristics such as molecular shape, denticity (number of binding sites), donor atom type, and flexibility are central to guiding the assembly process.

Molecular shape: Rigid, planar ligands tend to enforce ordered frameworks, facilitating the formation of predictable 2D or 3D networks. Examples include aromatic carboxylates and bipyridines, which serve as linear or angular connectors. Conversely, flexible ligands allow for adaptable conformations, potentially yielding interpenetrated or low-dimensional structures that can encapsulate guest molecules or facilitate dynamic behavior.

Donor atom type: Common donor atoms are nitrogen, oxygen, and sometimes sulfur. Nitrogen donors (e.g., pyridyl, imidazole groups) often form strong coordinate bonds with Cu (II), stabilizing the metal center and influencing the electronic structure. Oxygen donors, such as carboxylates and phenolates [7], frequently bridge metal centers, contributing to network extension and dimensionality. Mixed-donor ligands containing both N and O atoms provide additional coordination versatility.

Flexibility and denticity: Multidentate ligands with flexible backbones can wrap around metal centers in various conformations, allowing fine tuning of Cu–ligand bond angles and distances. This flexibility can be harnessed to engineer frameworks with specific pore sizes or surface functionalities.

2.4. Synthetic Strategies

Two main synthetic approaches dominate the preparation of Cu-CPs incorporating second auxiliary ligands: hydrothermal and solvothermal methods.

Hydrothermal synthesis involves crystallization in aqueous media at elevated temperature and pressure within sealed reactors (autoclaves). The high-temperature aqueous environment facilitates ligand deprotonation, metal–ligand coordination, and crystal growth under thermodynamic control. This method often yields high-purity crystalline Cu-CPs with well-defined morphologies, and is well suited for ligands sensitive to organic solvents.

Solvothermal synthesis uses non-aqueous solvents such as ethanol, DMF (dimethylformamide), or DMSO (dimethyl sulfoxide) under similar high-temperature, high-pressure conditions [8]. The choice of solvent modulates ligand solubility, coordination dynamics, and crystal nucleation. Solvothermal routes are particularly valuable for incorporating hydrophobic or poorly water-soluble second auxiliary ligands, enabling fine control over framework dimensionality and topology.

Both synthetic methods allow for systematic variation of reaction parameters such as temperature, reaction time, solvent polarity, ligand-to-metal ratio, and pH, providing a versatile toolbox to tailor Cu-CP structures via second auxiliary ligand incorporation. By carefully adjusting these conditions, researchers can influence the nucleation and growth kinetics, facilitating the formation of targeted architectures with desired physicochemical properties.

In summary, the ligand's structural features combined with the choice of synthetic strategy enable precise engineering of Cu-CP frameworks. The second auxiliary ligands act as crucial modulators in this process, enabling control over the coordination geometry, framework topology, and functional attributes of the resulting polymers [9].

3. Second Auxiliary Ligand Design Strategies

3.1. Types of Second Auxiliary Ligands

Second auxiliary ligands play a pivotal role in the structural engineering of copper coordination polymers (Cu-CPs). These ligands are typically introduced in addition to the primary ligands to fine-tune the dimensionality, connectivity, and functionality of the frameworks. Among the most common types are aromatic carboxylates, bipyridines, amides, V-shaped ligands, and various heterocyclic compounds.

Aromatic carboxylates, such as isophthalic acid and terephthalic acid derivatives, offer multiple coordination sites through their carboxyl groups and exhibit rigid planar structures that favor the formation of extended networks. Bipyridines and related N-donor ligands provide chelating sites with strong binding affinity to Cu (II) centers, which often leads to enhanced framework stability and predictable coordination geometries [10]. Amide-based ligands, characterized by their flexible backbones and hydrogen bonding capabilities, can influence the packing and overall topology of Cu-CPs.

V-shaped ligands, a subclass of auxiliary ligands with an angular molecular geometry, have recently garnered considerable interest due to their ability to direct the formation of two-dimensional (2D) layered structures with exposed active sites [11]. Heterocyclic ligands, including imidazole, pyridine, and triazole derivatives, introduce additional electronic effects and steric hindrance, which can modulate the coordination environment and catalytic properties.

The structural motifs presented by these ligands define their coordination behavior, including bridging modes, denticity, and conformational flexibility. For example, rigid aromatic carboxylates typically act as bridging ligands connecting multiple Cu centers, while flexible amides may coordinate in a chelating fashion or as monodentate ligands depending on the synthetic conditions.

3.2. Structural Effects on Cu-CPs

The incorporation of second auxiliary ligands significantly influences the network dimensionality and topology of Cu-CPs, ranging from one-dimensional (1D) chains to complex three-dimensional (3D) frameworks [12]. These ligands modulate key structural parameters, including pore size, metal coordination environment, and overall framework stability.

Rigid ligands with defined geometric angles favor the formation of extended 2D or 3D structures by linking copper centers in predictable spatial arrangements. For instance, the use of V-shaped auxiliary ligands promotes the assembly of 2D layered architectures, which exposes more catalytically active Cu sites on the surface, facilitating improved interaction with substrates [13]. On the other hand, flexible ligands or those with variable coordination modes can induce structural distortions or interpenetration, often reducing pore volume but enhancing mechanical robustness.

The modulation of pore size by second auxiliary ligands is critical for applications requiring selective molecular adsorption or controlled diffusion, such as catalysis and sensing. Ligand bulkiness and steric hindrance can tune pore aperture and shape, impacting guest molecule accessibility and binding.

The metal coordination environment, including bond lengths and angles around Cu centers, is also affected by auxiliary ligands. By altering electronic density and spatial constraints, these ligands influence catalytic activity and stability under operational conditions.

Examples from recent studies have demonstrated how varied auxiliary ligands induce distinct architectures. For example, the use of V-shaped second auxiliary ligands has led to two-dimensional Cu-CPs with enhanced urease inhibition activity, attributed to improved exposure of active sites and framework stability [5]. Other studies reveal that aromatic bipyridyl ligands can stabilize 3D networks with permanent porosity, which are beneficial for catalysis and adsorption.

3.3. Functional Implications

The strategic design and selection of second auxiliary ligands have profound functional implications for Cu-CPs. One of the key benefits is the enhanced exposure of catalytic sites. By directing framework assembly into low-dimensional or layered structures, auxiliary ligands facilitate substrate access and improve catalytic efficiency.

Moreover, second auxiliary ligands contribute to improved structural stability and robustness. Ligands that provide multidentate coordination or strong chelation enhance framework rigidity and resistance to environmental factors such as moisture and temperature fluctuations [14]. This is essential for practical applications in catalysis, agriculture, and environmental remediation where long-term stability is required.

The physicochemical properties of Cu-CPs can also be tailored for specific applications through ligand engineering. For example, modifying ligand functional groups can adjust hydrophobicity/hydrophilicity, acid-base properties, and electronic characteristics, enabling optimization for urease inhibition in agriculture or selective pollutant adsorption.

It has been shown that Cu-CPs fabricated with carefully chosen second auxiliary ligands exhibit significantly enhanced urease inhibitory performance, prolonged stability, and minimized environmental impact, making them promising candidates for sustainable agricultural applications [9]. These findings underscore the importance of ligand design in the rational development of functional Cu coordination polymers.

4. Case Studies of Functional Cu-CPs with Second Auxiliary Ligand Engineering

4.1. Urease Inhibition and Agricultural Applications

Copper coordination polymers (Cu-CPs) have gained considerable attention as urease inhibitors, a key strategy to enhance nitrogen use efficiency in agriculture by reducing ammonia volatilization [15]. The introduction of second auxiliary ligands enables fine-

tuning of the polymer architecture, which critically influences inhibitory performance. In particular, ligands with distinctive V-shaped geometries promote the formation of two-dimensional layered Cu-CPs, exposing more active copper sites that directly interact with urease enzymes.

This structural optimization leads to enhanced inhibition efficiency and better stability under soil conditions. Additionally, the use of chemical stabilizers alongside Cu-CPs has been shown to prolong their effectiveness in the soil–plant system, mitigating rapid degradation and leaching issues commonly faced by urease inhibitors. Such combined approaches open new avenues for developing more sustainable and long-lasting agro-chemical formulations.

4.2. Catalytic Applications

Beyond agriculture, Cu-CPs with engineered second auxiliary ligands exhibit impressive catalytic capabilities in various chemical transformations [16]. The ligands not only determine the coordination geometry but also affect pore size, surface area, and electronic properties of the copper centers, all of which are essential parameters for catalytic activity.

For instance, Cu-CPs have been successfully applied as catalysts in oxidative coupling reactions, where ligand-induced structural features enhance substrate accessibility and product diffusion. These materials demonstrate high turnover numbers and selectivity, illustrating the pivotal role of ligand design in catalytic performance.

Diverse ligand families—ranging from aromatic carboxylates to heterocyclic compounds—offer multiple coordination modes, thereby enabling tailored catalytic environments. This adaptability makes Cu-CPs versatile platforms for green catalysis with scope for further performance enhancement.

4.3. Emerging Functionalities: Sensing, Antimicrobial, and Environmental Remediation

Second auxiliary ligand engineering has also broadened the functional scope of Cu-CPs into sensing, antimicrobial, and environmental applications. By adjusting ligand types and framework topology, researchers have created materials with selective sensing abilities for detecting pollutants and biomolecules.

In antimicrobial applications, ligand-mediated modulation of copper ion release kinetics can improve efficacy against pathogens while minimizing toxicity. Moreover, Cu-CPs with designed porosity and surface chemistry are effective in adsorbing and degrading environmental contaminants such as dyes and pesticides.

These examples demonstrate how ligand engineering not only tunes structural features but also tailors physicochemical properties for specialized applications, reinforcing the versatility of Cu-CPs across disciplines.

5. Synthetic and Characterization Techniques

The synthesis of copper coordination polymers (Cu-CPs) incorporating second auxiliary ligands requires carefully controlled reaction conditions to ensure the desired structural assembly and functional properties. Among the most commonly employed methods are solvothermal and hydrothermal syntheses, which provide a versatile environment for crystallization under moderate to high temperature and pressure. These approaches facilitate effective coordination between Cu (II) centers and both primary and secondary ligands, allowing precise tuning of dimensionality and framework topology.

Slow diffusion techniques and room-temperature self-assembly are also utilized for specific ligand systems, especially when the goal is to obtain high-quality single crystals for detailed structural analysis. The choice of solvent, temperature, pH, and ligand-to-metal ratio profoundly influences the resultant polymer morphology and stability, underscoring the importance of optimized synthetic protocols.

Comprehensive characterization of the resulting Cu-CPs is essential to confirm their structure and evaluate their potential functionalities. Powder X-ray diffraction (PXRD) provides information on crystallinity and phase purity, while single-crystal X-ray diffraction (SCXRD) offers atomic-level resolution of the coordination environment, including metal–ligand bond lengths and angles. These structural insights are crucial for correlating ligand design with polymer architecture.

Complementary spectroscopic techniques such as Fourier-transform infrared spectroscopy (FTIR) are employed to verify ligand incorporation and identify characteristic functional groups. Morphological features and particle size distribution are investigated using scanning electron microscopy (SEM) and transmission electron microscopy (TEM), which reveal the surface texture and nanoscale framework details. Additionally, Brunauer–Emmett–Teller (BET) surface area analysis quantifies porosity, a key parameter for catalytic and adsorption applications.

Evaluating the functional performance of Cu-CPs involves application-specific tests. For urease inhibition, standardized enzyme assays measure the decrease in urease activity in the presence of the polymers, enabling assessment of inhibitory potency and stability over time. In catalytic studies, reaction kinetics, product yields, and selectivity are analyzed through various chemical assays and chromatographic methods. Such comprehensive testing establishes the practical viability of Cu-CPs and guides further ligand optimization.

6. Conclusions

This review highlights the pivotal role of second auxiliary ligand engineering in advancing the design and functionalization of copper coordination polymers (Cu-CPs). Through careful selection and structural modification of these secondary ligands, researchers have successfully tuned the dimensionality, stability, and porosity of Cu-CP frameworks, enabling the development of materials with tailored physicochemical properties. The incorporation of diverse ligand types—ranging from aromatic carboxylates to V-shaped and heterocyclic motifs—has demonstrated significant influence on network architecture, facilitating enhanced exposure of catalytically active copper centers and improved robustness under operational conditions.

Moreover, the strategic design of second auxiliary ligands has translated into marked improvements in application performance. Notably, in agricultural contexts, Cu-CPs engineered with specific ligands have exhibited superior urease inhibition efficiency, contributing to sustainable nitrogen management practices. In catalysis, ligand-driven modulation of active sites and pore environments has led to increased reaction rates and selectivity. Beyond these, emerging applications in sensing, antimicrobial activity, and environmental remediation underscore the versatility endowed by ligand engineering.

Looking forward, continued exploration of novel ligand scaffolds and synthetic methodologies will be essential to overcome current challenges such as long-term stability, scalability, and environmental compatibility. Integrating computational design with experimental approaches offers promising avenues for predicting and optimizing ligand–metal interactions at the molecular level. Furthermore, expanding the functional scope of Cu-CPs through multifunctional ligand frameworks could unlock new possibilities in smart agrochemicals, green catalysis, and advanced material platforms.

In conclusion, second auxiliary ligand engineering stands as a powerful strategy for fine-tuning copper coordination polymers, bridging fundamental coordination chemistry with practical applications. Its ongoing development is expected to drive innovations that contribute significantly to sustainable technologies and materials science.

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