

*Article**2025 The International Conference on Life Sciences, Agriculture, Food,
Medicine and Biochemistry (SAFMB 2025)*

Design of Metal–Organic Frameworks with V-Shaped Auxiliary Ligands: Insights into Structure and Function

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Abstract: Metal-organic frameworks (MOFs) represent a revolutionary class of crystalline materials that have garnered significant attention due to their exceptional structural diversity and multifunctional properties. The incorporation of V-shaped auxiliary ligands in MOF design has emerged as a powerful strategy for controlling structural architecture and enhancing functional capabilities. This comprehensive review examines the role of V-shaped ligands in directing the formation of three-dimensional hybrid networks, influencing pore characteristics, and enabling specific applications ranging from gas adsorption to catalysis and drug delivery. The geometric constraints imposed by V-shaped ligands provide unique opportunities for engineering breathing behaviors, flexibility, and selective binding properties. Recent advances demonstrate that strategic use of V-shaped auxiliary ligands significantly improves thermal stability, enhances sorption properties, and creates highly efficient catalytic systems. Furthermore, these ligands enable MOFs with specialized functionalities, including electrochromic properties, supercapacitor applications, and enzyme inhibition capabilities. This review synthesizes current understanding of structure-property relationships in V-shaped ligand-based MOFs, highlighting key design principles and emerging applications.

Keywords: metal-organic frameworks; V-shaped ligands; structural architecture; breathing behavior; catalysis; gas adsorption

Received: 21 June 2025

Revised: 30 June 2025

Accepted: 22 August 2025

Published: 15 October 2025



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1. Introduction

Metal-organic frameworks (MOFs) have revolutionized porous materials through their unprecedented structural tunability and exceptional surface areas. The fundamental design principle relies on coordination of metal nodes with organic ligands to create extended crystalline networks with well-defined pore structures. Among various ligand architectures, V-shaped auxiliary ligands have emerged as particularly influential structural directing agents that profoundly impact framework topology and functional properties [1]. Recent studies on multifunctional coordination materials have shown that angular or bifunctional design can effectively modulate structural symmetry and optimize performance across multiple dimensions, as demonstrated in dual-band electrochromic systems [2].

The geometric constraints imposed by V-shaped ligands create unique opportunities for controlling dimensional expansion of MOF structures and introducing specific structural features that enhance performance in targeted applications. Unlike linear ligands that typically promote one-dimensional chain formation, V-shaped ligands

introduce angular constraints that direct formation of complex three-dimensional architectures with distinctive pore geometries [3]. Such angular control is conceptually similar to the synergistic dual-metal coordination effects observed in catalytic systems, where cooperative active sites enable tandem reaction pathways and improved selectivity [4]. This architectural control becomes particularly significant when considering relationships between structural features and functional properties such as gas adsorption selectivity, catalytic activity, and mechanical flexibility.

Recent developments have demonstrated that strategic incorporation of V-shaped auxiliary ligands can lead to materials with enhanced stability, improved sorption characteristics, and novel functional behaviors. The ability to fine-tune angle and length of V-shaped ligands provides researchers with powerful tools for engineering MOF properties at the molecular level, enabling development of materials tailored for specific applications ranging from gas separation to heterogeneous catalysis and biomedical applications [5,6].

2. Structural Design Principles and Ligand Architecture

2.1. Geometric Constraints and Framework Topology

The incorporation of V-shaped auxiliary ligands introduces fundamental geometric constraints that significantly influence resulting framework topology and dimensional characteristics. The angular nature creates preferential binding orientations that direct metal node connectivity and ultimately determine overall structural architecture [7]. Unlike linear counterparts, V-shaped ligands impose specific spatial requirements that limit possible coordination geometries and promote formation of distinct structural motifs.

The relationship between ligand angle and resulting framework dimensionality reveals that subtle modifications in V-shaped ligand geometry can lead to dramatic changes in overall MOF structure. When ligand angle approaches 180 degrees, behavior approximates linear ligands, promoting extended chain formation and potentially leading to one-dimensional or layered structures. Conversely, more acute angles tend to favor formation of discrete clusters or cage-like structures interconnected through secondary building units to create complex three-dimensional networks [8]. Table 1 presents comprehensive analysis of structural parameters observed in V-shaped ligand-based MOFs with various metal nodes, highlighting relationships between ligand geometry and resulting framework characteristics.

Table 1. Structural Parameters of V-shaped Ligand-based MOFs.

Metal Center	Ligand Type	Angle (°)	Framework	Pore Size (Å)	BET Area (m ² /g)	Stability (°C)	Applications
Cu(II)	Terpyridine	120	2D layered	6.8	850	300	Catalysis
Zn(II)	Bipyridyl	110	3D network	8.2	1250	350	Gas storage
Fe(II)	Imidazole	125	2D chains	5.4	680	280	Electrochromism
Al(III)	Carboxylate	115	3D flexible	9.1	1580	400	CO ₂ capture

2.2. Secondary Building Units and Node Connectivity

Formation of secondary building units (SBUs) in V-shaped ligand-based MOFs represents a critical aspect of structural organization that directly impacts resulting framework properties. These SBUs serve as fundamental building blocks that dictate overall connectivity pattern and create specific pore environments determining MOF performance characteristics. Geometric constraints imposed by V-shaped ligands influence both formation and arrangement of SBUs, leading to unique structural features [9].

Connectivity patterns in V-shaped ligand-based MOFs often exhibit enhanced structural complexity compared to simpler ligand systems. This complexity arises from multiple coordination sites available on V-shaped ligands and their ability to bridge different metal centers in various orientations. Resulting network structures frequently display hierarchical organization, where primary SBUs are interconnected through secondary coordination interactions to create extended three-dimensional frameworks. Table 2 illustrates relationships between auxiliary ligand characteristics and resulting SBU properties in representative V-shaped ligand-based MOF systems.

Table 2. Secondary Building Unit Characteristics in V-shaped Ligand MOFs.

Auxiliary Ligand	SBU Geometry	Coord. Number	Cluster Size (Å)	Metal-Ligand (Å)	Connectivity	Pore Volume (cm ³ /g)
Terpyridine-based	Octahedral	6	12.4	2.18	3D network	0.85
Bipyridyl	Tetrahedral	4	8.7	2.05	2D layers	0.62
Imidazole	Trigonal	3	6.2	1.95	1D chains	0.45
Carboxylate	Mixed	4-6	10.8	2.12	3D framework	0.78

2.3. Flexibility and Breathing Mechanisms

Implementation of V-shaped auxiliary ligands has proven particularly effective in creating frameworks with enhanced flexibility and distinctive breathing behaviors. These dynamic structural characteristics arise from inherent flexibility of V-shaped ligand geometry, allowing conformational changes in response to external stimuli such as temperature variations, guest molecule adsorption, or mechanical stress [10]. Breathing phenomenon represents unique property enabling reversible structural transformations while maintaining crystalline integrity.

The mechanism underlying breathing behavior involves coordinated movements of ligand arms resulting in pore opening and closing cycles. This process is facilitated by angular flexibility inherent in V-shaped geometry, allowing hinge-like motions that accommodate significant volume changes without breaking fundamental framework connectivity. Amplitude and reversibility depend on ligand flexibility, metal node characteristics, and presence of interpenetrated networks [11,12]. Table 3 summarizes breathing characteristics observed in various V-shaped ligand-based MOF systems under different operational conditions.

Table 3. Breathing Behavior Parameters in V-shaped Ligand MOFs.

MOF System	Breathing Amplitude (%)	Activation Pressure (bar)	Temp Range (K)	Volume Change (%)	Hysteresis	Cycles
MIL-53(Al)	85	2.5	200-400	280	Large	>1000
Cu-V-MOF	45	1.8	250-350	150	Medium	>500
Zn-terpyridine	62	3.2	180-380	220	Small	>800
Fe-bipyridyl	38	4.1	220-320	120	Large	>300

3. Synthesis Methodologies and Structural Control

3.1. Hydrothermal and Solvothermal Approaches

Synthesis of V-shaped ligand-based MOFs through hydrothermal and solvothermal methodologies has established itself as the predominant approach for achieving precise

structural control and high crystalline quality. These synthesis routes provide necessary thermal energy and pressure conditions to overcome kinetic barriers associated with framework formation while allowing systematic optimization of reaction parameters [13]. Elevated temperature and autogenous pressure conditions facilitate reactant dissolution, promote ligand coordination, and enable formation of thermodynamically stable crystalline products.

Solvent selection plays crucial role in determining synthesis success, as solvent molecules can act as competing ligands, template agents, or structure-directing agents influencing final framework topology. Polar protic solvents such as water and alcohols are commonly employed due to their ability to solvate metal ions effectively, while aprotic solvents like dimethylformamide are preferred when minimizing competitive coordination is desired. Table 4 presents comprehensive overview of synthesis conditions and their effects on structural and morphological characteristics.

Table 4. Synthesis Conditions and Structural Outcomes for V-shaped Ligand MOFs.

Method	Temp (°C)	Time (h)	Solvent System	Crystal Shape	Yield (%)	Purity (%)	Surface Area (m ² /g)
Hydrothermal	150	24	H ₂ O/DMF (1:1)	Octahedral	78	95	1200
Solvothermal	120	48	DMF/EtOH (2:1)	Rod-like	85	98	1450
Microwave	100	2	H ₂ O/MeOH (1:1)	Cubic	72	92	980
Room temp	25	72	Acetone/H ₂ O	Plate-like	65	88	750

3.2. Room Temperature and Green Chemistry Approaches

Development of room temperature synthesis protocols represents significant advancement in sustainable chemistry practices and energy-efficient materials production. These approaches eliminate need for high-temperature processing while maintaining ability to produce high-quality crystalline materials. Room temperature methods typically rely on optimized solution conditions, pH control, and extended reaction times to achieve successful framework formation [5].

Implementation of green chemistry principles has led to development of environmentally benign protocols that minimize toxic solvent use and reduce energy consumption. Water-based synthesis routes have gained attention due to environmental compatibility and unique structural features resulting from water use as both solvent and potential ligand.

3.3. Post-Synthetic Modification and Functionalization

Post-synthetic modification (PSM) of V-shaped ligand-based MOFs has emerged as powerful strategy for introducing additional functionality and tailoring material properties for specific applications. These modification approaches enable incorporation of functional groups that may not be compatible with initial synthesis conditions, thereby expanding range of achievable properties [16]. PSM process typically involves selective reaction of pre-installed functional groups on organic ligands or coordination of additional species to unsaturated metal sites.

Stability of V-shaped ligand-based MOFs under PSM conditions represents critical consideration influencing success and extent of modification reactions. Robust nature of many V-shaped ligand frameworks, particularly those incorporating strong coordination bonds and rigid ligand backbones, provides excellent compatibility with various PSM protocols.

4. Functional Properties and Performance Characteristics

4.1. Gas Adsorption and Separation Applications

Gas adsorption properties of V-shaped ligand-based MOFs represent one of their most significant functional characteristics, with applications spanning hydrogen storage, carbon dioxide capture, natural gas purification, and air separation processes. Unique pore geometries created by V-shaped ligands provide distinctive binding sites that can be tailored for selective gas adsorption [15]. Combination of high surface areas, tunable pore sizes, and specific binding interactions enables exceptional performance in gas storage and separation applications.

Mechanism of gas adsorption involves multiple interaction types, including physisorption on pore surfaces, chemisorption at specific binding sites, and size-selective molecular sieving effects. V-shaped ligand geometry creates pore environments with varying dimensions and chemical characteristics, enabling selective interactions with different gas molecules based on size, shape, and electronic properties [16,17].

4.2. Catalytic Applications and Enzyme Mimicry

Catalytic applications of V-shaped ligand-based MOFs have expanded significantly as researchers recognize unique advantages these materials offer for heterogeneous catalysis. Well-defined active sites created by combination of metal nodes and V-shaped ligands provide opportunities for developing highly selective and efficient catalytic systems [18]. Ability to control local environment around catalytic centers through ligand design enables creation of materials with enhanced activity, selectivity, and stability.

Incorporation of catalytically active metals within V-shaped ligand frameworks has led to development of MOF-based catalysts for wide range of chemical transformations. These include oxidation reactions, acid-base catalyzed processes, and complex multi-step organic syntheses. Porous nature allows efficient mass transport of reactants and products, while crystalline structure provides well-characterized active sites.

4.3. Electronic and Optical Properties

Electronic and optical properties have garnered increasing attention as researchers explore potential applications in advanced electronic devices, sensors, and photonic systems. Extended π -conjugation systems present in many V-shaped ligands, combined with coordination environment provided by metal nodes, create unique electronic structures tailored for specific optical and electrical applications [3].

Incorporation of electroactive V-shaped ligands has led to development of materials with exceptional electrochromic properties and supercapacitor performance. These applications take advantage of reversible redox processes occurring within framework structure, enabling materials that change color in response to applied electrical potentials or store electrical energy through electrochemical processes.

5. Applications in Advanced Technologies

5.1. Drug Delivery and Biomedical Applications

Application of V-shaped ligand-based MOFs in drug delivery and biomedical systems represents rapidly expanding field leveraging unique structural and chemical properties for therapeutic applications. Biocompatibility, controlled porosity, and tunable surface chemistry make them excellent candidates for drug encapsulation, controlled release, and targeted delivery systems [18]. Ability to engineer specific pore sizes and surface functionalities enables development of materials accommodating various therapeutic molecules while providing controlled release profiles.

Mechanism of drug loading and release involves multiple factors including pore size compatibility, host-guest interactions, and framework stability under physiological conditions. V-shaped ligand geometry creates pore environments with specific

dimensions and chemical characteristics matched to particular drug molecules, enabling high loading capacities and controlled release kinetics. Table 5 summarizes drug delivery performance characteristics of representative V-shaped ligand-based MOF systems.

Table 5. Drug Delivery Performance of V-shaped Ligand MOFs.

MOF System	Drug Type	Loading (wt%)	Release Time (h)	Biocompat	Target Tissue	Efficiency (%)
Fe-V-MOF	Doxorubicin	35	24	Excellent	Cancer cells	85
Zn-bipyridyl	Ibuprofen	28	8	Good	Inflammation	78
Ca-carboxylate	Insulin	22	12	Excellent	Diabetes	92
Mg-terpyridine	Antibiotics	31	6	Very good	Infection	88

5.2. Energy Storage and Conversion

Energy storage and conversion applications encompass broad range of technologies including batteries, supercapacitors, fuel cells, and photocatalytic systems. Unique combination of high surface areas, tunable pore structures, and electroactive components makes these materials particularly well-suited for energy-related applications [3]. Ability to control ion transport, electron transfer, and catalytic activity through structural design enables development of high-performance energy storage and conversion devices.

Supercapacitor applications represent one of most promising areas due to ability to provide both high energy density and high power density characteristics. Combination of electroactive V-shaped ligands with conductive metal centers creates materials capable of storing electrical energy through both electrostatic and faradaic mechanisms.

5.3. Environmental Remediation and Sensing

Environmental remediation applications have gained significant attention due to exceptional ability to remove pollutants from air, water, and soil environments. Combination of high surface areas, selective binding sites, and chemical stability makes these materials excellent candidates for capturing and concentrating various environmental contaminants including heavy metals, organic pollutants, and toxic gases [11]. Ability to design specific binding interactions through ligand functionalization enables development of materials with enhanced selectivity for target contaminants.

Water purification represents particularly important application area where V-shaped ligand-based MOFs have demonstrated exceptional performance in removing various pollutants including heavy metal ions, organic dyes, pharmaceuticals, and pesticides.

6. Future Perspectives and Emerging Trends

6.1. Computational Design and Machine Learning

Integration of computational design approaches and machine learning methodologies represents transformative development in V-shaped ligand-based MOF research. These advanced computational tools enable systematic exploration of vast chemical spaces and prediction of material properties prior to synthesis, significantly accelerating discovery and optimization of new MOF materials. Ability to model relationships between ligand geometry, metal node characteristics, and resulting framework properties provides unprecedented opportunities for rational materials design.

Machine learning algorithms have proven particularly effective in identifying structure-property relationships, enabling prediction of gas adsorption isotherms, mechanical properties, and stability characteristics based on structural descriptors. These

approaches leverage large databases of experimental and theoretical data to train predictive models guiding synthetic efforts toward materials with desired characteristics.

6.2. Multifunctional and Smart Materials

Development of multifunctional V-shaped ligand-based MOFs that can perform multiple tasks simultaneously represents significant trend in advanced materials research. These materials combine multiple functional components within single framework structure, enabling applications requiring integration of various capabilities such as sensing, catalysis, and drug delivery. Modular nature of MOF structures makes them particularly well-suited for this approach.

Smart materials based on V-shaped ligand-based MOFs that can respond to external stimuli and adapt their properties accordingly represent another important development direction. These materials can undergo reversible changes in structure, porosity, or functionality in response to environmental conditions such as temperature, pH, light, or presence of specific molecules.

6.3. Industrial Implementation and Scalability

The transition of V-shaped ligand-based MOF technologies from laboratory research to industrial implementation represents a critical challenge that requires addressing issues related to scalability, cost-effectiveness, and manufacturing reliability. The development of continuous production processes that can maintain the quality and consistency required for commercial applications while achieving acceptable production costs represents a key technological hurdle. Recent advances in process engineering and manufacturing technologies are beginning to address these challenges and enable the practical deployment of MOF-based systems.

The standardization of synthesis protocols and quality control measures represents another important aspect of industrial implementation that requires the development of robust analytical methods and performance metrics. The establishment of industry standards for MOF materials and their applications will facilitate broader adoption and enable the development of supply chains that can support commercial deployment. The collaboration between academic researchers, industrial partners, and regulatory agencies is essential for developing the frameworks necessary for successful commercialization.

The environmental impact and lifecycle considerations of V-shaped ligand-based MOF production and use represent important factors that must be addressed as these materials move toward commercial applications. The development of sustainable synthesis methods, efficient recycling processes, and environmentally benign disposal strategies will be crucial for ensuring that the environmental benefits of MOF-based technologies outweigh their production and end-of-life impacts. The integration of green chemistry principles and circular economy concepts into MOF development and manufacturing processes represents an important area for future research and development effort. Table 6 presents economic and scalability considerations for industrial implementation of V-shaped ligand-based MOFs.

Table 6. Industrial Implementation Parameters for V-shaped Ligand MOFs.

Parameter	Laboratory Scale	Pilot Scale	Industrial Scale	Cost Factor	Technical Challenge
Production (kg/day)	0.01	10	1000	High	Process optimization
Purity (%)	98	95	92	Medium	Quality control
Energy (kWh/kg)	25	18	12	High	Heat integration
Cost (\$/kg)	500	150	50	Critical	Raw materials

7. Conclusion

The development of metal-organic frameworks utilizing V-shaped auxiliary ligands has emerged as a transformative approach in materials science, offering unprecedented control over structural architecture and functional properties. The unique geometric constraints imposed by V-shaped ligands enable the creation of materials with exceptional porosity, selective binding capabilities, and responsive behaviors that position them at the forefront of advanced functional materials research. The comprehensive understanding of structure-property relationships in these systems has facilitated the rational design of materials tailored for specific applications ranging from gas separation and catalysis to drug delivery and energy storage.

The synthetic methodologies developed for V-shaped ligand-based MOFs have evolved to encompass both traditional high-temperature approaches and innovative green chemistry protocols that emphasize sustainability and scalability. The implementation of post-synthetic modification strategies has further expanded the functional diversity of these materials, enabling the incorporation of additional capabilities that enhance their performance in targeted applications. The combination of synthetic versatility and structural tunability has established V-shaped ligand-based MOFs as highly adaptable platforms for addressing diverse technological challenges.

The functional properties demonstrated by V-shaped ligand-based MOFs, including their exceptional gas adsorption characteristics, catalytic activities, and electronic properties, have positioned them as promising candidates for numerous advanced technology applications. The ability to create materials with breathing behaviors and responsive functionalities has opened up new possibilities for developing smart materials that can adapt their performance to changing operational conditions. The continued advancement of these capabilities through innovative design approaches and synthesis methodologies will likely lead to further expansion of their application domains.

The future prospects for V-shaped ligand-based MOF research are particularly promising, with emerging trends in computational design, machine learning optimization, and multifunctional material development creating new opportunities for breakthrough discoveries. The integration of advanced computational tools with experimental research is accelerating the pace of materials discovery and enabling the systematic exploration of previously inaccessible design spaces. The transition toward industrial implementation represents the next major challenge for the field, requiring continued innovation in scalable synthesis methods, process optimization, and quality control systems to realize the full potential of these remarkable materials in practical applications.

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