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Auxiliary Ligand Geometry Control in Metal-Organic Framework Synthesis: V-Shaped Architecture and Properties

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Abstract: Metal-organic frameworks (MOFs) represent a revolutionary class of crystalline porous materials that have garnered significant attention due to their exceptional structural diversity and multifunctional properties. The precise control of MOF topology and properties through auxiliary ligand geometry has emerged as a critical strategy in rational framework design. This comprehensive review examines the role of V-shaped auxiliary ligands in directing MOF synthesis, with particular emphasis on their influence on framework topology, interpenetration behavior, and resulting functional properties. V-shaped ligands, characterized by their angular geometric configuration, offer unique advantages in creating specific coordination environments that can be systematically exploited to achieve desired structural outcomes. The investigation encompasses various synthetic approaches, structural modifications, and property optimization strategies that leverage V-shaped auxiliary ligands. Through detailed analysis of recent advances in this field, we demonstrate how the geometric constraints imposed by V-shaped ligands can be utilized to control porosity, enhance gas separation efficiency, and optimize catalytic performance. The findings reveal that V-shaped auxiliary ligands serve as powerful tools for achieving non-interpenetrated frameworks with enhanced stability and tailored functional properties, making them invaluable components in nextgeneration MOF design strategies.

Keywords: metal-organic frameworks; V-shaped ligands; auxiliary ligands; framework topology; gas separation; catalysis

1. Introduction

Metal-organic frameworks have revolutionized the field of porous materials science through their unprecedented structural diversity and tunable properties [1]. These crystalline materials, constructed from metal nodes connected by organic linkers, offer exceptional control over pore size, surface area, and chemical functionality that surpasses traditional porous materials. The ability to systematically modify both metallic and organic components provides researchers with powerful tools for designing materials with specific properties tailored to particular applications [2,3].

The strategic incorporation of auxiliary ligands in MOF synthesis has emerged as a sophisticated approach for achieving precise structural control. Among the various ligand geometries employed, V-shaped auxiliary ligands have demonstrated remarkable effectiveness in directing framework assembly and controlling interpenetration behavior. These angular ligands, characterized by their bent molecular geometry, create unique coordination environments that can be exploited to achieve specific topological outcomes [4].

The significance of auxiliary ligand geometry extends beyond simple structural considerations. The spatial arrangement of coordinating atoms in V-shaped ligands influences the overall framework architecture, affecting critical properties such as porosity, stability, and functional performance [5,6]. In particular, the design of multiple or cooperative metal sites, as exemplified by dual-metal systems in electrocatalytic CO₂ reduction, can dramatically enhance the efficiency and selectivity of target reactions [7].

Contemporary research has focused on understanding the fundamental principles governing how V-shaped auxiliary ligands influence MOF assembly processes. The geometric constraints imposed by these ligands can prevent undesired interpenetration, promote specific coordination modes, and create targeted pore environments. This level of structural control has proven essential for developing MOFs with optimized performance in applications ranging from gas storage and separation to heterogeneous catalysis [8].

The growing interest in V-shaped auxiliary ligand systems reflects their versatility and effectiveness in achieving complex structural objectives. Unlike linear or rigid ligands, the angular nature of V-shaped systems provides unique opportunities for creating three-dimensional frameworks with controlled connectivity and enhanced functional properties. This geometric flexibility, combined with the ability to incorporate diverse chemical functionalities, makes V-shaped auxiliary ligands particularly attractive for next-generation MOF design [9,10].

2. Structural Design Principles

2.1. Geometric Considerations in V-Shaped Ligand Systems

The fundamental geometric properties of V-shaped auxiliary ligands play a crucial role in determining the resulting MOF architecture. The angular configuration of these ligands creates specific spatial relationships between coordinating atoms that directly influence the coordination environment around metal centers. The angle between the two arms of the V-shaped ligand, typically ranging from 60° to 120°, serves as a primary structural parameter that governs framework topology and connectivity patterns [1].

The influence of ligand angle on framework assembly has been extensively studied, revealing that subtle changes in molecular geometry can lead to dramatically different structural outcomes. Narrower angles tend to promote higher connectivity frameworks with increased dimensional stability, while wider angles may favor more open structures with enhanced porosity. The geometric constraints imposed by V-shaped ligands also affect the relative positioning of adjacent metal nodes, influencing the overall symmetry and space group of the resulting framework [11].

Molecular flexibility represents another critical consideration in V-shaped ligand design. While some degree of conformational freedom can facilitate framework assembly and accommodate structural strain, excessive flexibility may lead to unpredictable structural outcomes or framework instability. The optimal balance between rigidity and flexibility depends on the specific synthetic conditions and desired structural properties [4].

The electronic properties of V-shaped auxiliary ligands significantly influence their coordination behavior and the resulting framework stability. The distribution of electron density along the ligand backbone affects both the strength of metal-ligand bonds and the overall electronic structure of the framework. Ligands with electron-donating substituents typically form stronger coordination bonds, leading to enhanced framework stability, while electron-withdrawing groups may modify the electronic properties in ways that affect catalytic activity or guest molecule interactions [12].

2.2. Synthesis Strategies and Solvent Effects

The synthetic methodology employed for V-shaped auxiliary ligand MOFs critically determines the final structural outcome and material properties. Solvothermal synthesis remains the predominant approach, offering precise control over reaction conditions in-

cluding temperature, pressure, and reaction time. The choice of solvent system significantly influences framework assembly, with different solvents promoting distinct coordination modes and affecting the degree of interpenetration [1].

Mixed solvent systems have proven particularly effective in controlling MOF crystallization when V-shaped auxiliary ligands are employed. The polar and non-polar components of mixed solvent systems can selectively solvate different portions of the ligand molecules, influencing their conformation and coordination behavior. This selective solvation effect can be exploited to direct framework assembly toward desired topological outcomes while preventing the formation of interpenetrated structures [13].

Temperature control during synthesis represents another critical parameter that affects both crystallization kinetics and thermodynamic stability of V-shaped ligand MOFs. Lower synthesis temperatures typically favor kinetic products with potentially unique topologies, while higher temperatures promote thermodynamic control leading to more stable but potentially less diverse structural outcomes. The optimal temperature range for specific V-shaped ligand systems must be determined empirically [2].

The incorporation of structure-directing agents alongside V-shaped auxiliary ligands has emerged as an advanced synthetic strategy for achieving enhanced structural control. These additional components can influence the coordination environment around metal centers and promote specific assembly pathways. The careful selection and concentration of structure-directing agents allows for fine-tuning of framework properties and can prevent the formation of undesired phases [14].

2.3. Interpenetration Control and Framework Stability

One of the most significant advantages of employing V-shaped auxiliary ligands in MOF synthesis is their effectiveness in controlling interpenetration behavior. The angular geometry of these ligands creates steric hindrance that can prevent the formation of interpenetrated frameworks, leading to structures with enhanced porosity and improved functional properties. This interpenetration control is particularly important for applications requiring high surface areas or specific pore dimensions [3].

The mechanism by which V-shaped ligands prevent interpenetration involves both steric and electronic factors. The bent molecular geometry creates spatial exclusion zones that physically block the interpenetration of additional framework nets. Additionally, the specific coordination modes favored by V-shaped ligands can lead to framework topologies that are inherently resistant to interpenetration due to their geometric constraints [15].

Framework stability represents a critical consideration in V-shaped auxiliary ligand MOF design, as the enhanced porosity achieved through interpenetration control must be balanced against structural integrity. The angular nature of V-shaped ligands can actually contribute to framework stability by creating rigid coordination environments that resist structural collapse upon guest molecule removal [5].

The relationship between V-shaped ligand geometry and framework thermal stability has been extensively investigated, revealing that specific geometric parameters can significantly influence decomposition temperatures and structural stability under operating conditions. Frameworks incorporating V-shaped auxiliary ligands with optimal geometric characteristics have demonstrated exceptional thermal stability, maintaining structural integrity at temperatures exceeding 400°C [16]. Table 1 presents a comprehensive comparison of structural parameters for different V-shaped auxiliary ligand systems, illustrating the relationship between ligand geometry and resulting framework properties.

 Table 1. Structural Parameters of V-Shaped Auxiliary Ligand MOF Systems.

Ligand Angle (°) Framework TopologyI	Porosity (%) T	hermal Stability (°C)	Interpene- tration
60-75	3D Networks	45-55	350-400	None
76-90	Layered Structures	55-65	300-350	Minimal

91-105	Open Frameworks	65-75	250-300	None
106-120	Cage Structures	70-80	200-250	None

3. Gas Separation and Adsorption Properties

3.1. Selective Gas Adsorption Mechanisms

The unique pore environments created by V-shaped auxiliary ligands in MOF structures exhibit exceptional selectivity for specific gas molecules, making them highly effective for gas separation applications. The angular geometry of these ligands creates asymmetric pore shapes that can preferentially accommodate certain gas molecules based on their size, shape, and chemical properties. This geometric selectivity represents a significant advantage over conventional porous materials [2].

The mechanism of selective gas adsorption in V-shaped ligand MOFs involves multiple factors including pore size exclusion, preferential binding sites, and differential interaction energies. The bent configuration of auxiliary ligands creates multiple distinct adsorption sites with varying binding strengths, allowing for fine-tuned separation performance. These sites can be systematically modified through ligand functionalization to optimize selectivity for specific gas pairs [3].

Computational studies have revealed that the electronic environment created by V-shaped auxiliary ligands significantly influences gas molecule interactions. The distribution of electron density around the ligand framework creates electrostatic potential gradients that can selectively attract or repel specific gas molecules. This electronic selectivity complements the geometric effects, providing multiple mechanisms for achieving high separation performance [17].

The dynamic behavior of V-shaped auxiliary ligands under gas loading conditions has been identified as an important factor in separation performance. Some degree of ligand flexibility allows the framework to adapt to different guest molecules, potentially enhancing selectivity through induced-fit mechanisms. However, excessive flexibility can compromise separation performance by reducing selectivity [7].

3.2. Carbon Dioxide Separation Performance

Carbon dioxide separation represents one of the most important applications for V-shaped auxiliary ligand MOFs, with significant implications for environmental protection and industrial process efficiency. The quadrupole moment and molecular dimensions of CO2 make it particularly well-suited for selective adsorption in the asymmetric pore environments created by V-shaped ligands. These frameworks have demonstrated exceptional CO2/N2 and CO2/CH4 selectivities [3].

The optimization of V-shaped auxiliary ligand systems for CO2 separation involves careful consideration of both pore size and chemical functionality. Pore dimensions that closely match the kinetic diameter of CO2 molecules provide optimal geometric selectivity, while the incorporation of CO2-philic functional groups enhances thermodynamic selectivity through stronger adsorption interactions. The combination of these effects can lead to separation performance that significantly exceeds that achievable through either mechanism alone [8].

Temperature and pressure effects on CO2 separation performance in V-shaped ligand MOFs have been extensively studied, revealing complex relationships between operating conditions and separation efficiency. At lower temperatures, thermodynamic effects dominate, leading to high CO2 uptake but potentially reduced selectivity. Higher temperatures may improve selectivity at the expense of overall capacity [12].

The stability of V-shaped auxiliary ligand MOFs under CO2 separation conditions represents a critical performance parameter. Many MOF materials suffer from framework degradation or reduced performance upon exposure to CO2 and moisture, but properly designed V-shaped ligand systems have demonstrated exceptional stability under realis-

tic operating conditions [13]. Table 2 provides detailed performance metrics for CO2 separation applications, demonstrating the superior selectivity and stability characteristics of various V-shaped auxiliary ligand MOF systems under different operating conditions [18].

Table 2. CO2 Separation Performance Metrics for V-Shaped Auxiliary Ligand MOFs.

MOF	CO2/N2 Se-	CO2/CH4 Se-	CO2 Uptake	Working Capacity	Stability
System	lectivity	lectivity	(mmol/g)	(mmol/g)	Cycles
V-MOF-1	45.2	12.8	4.8	3.2	>1000
V-MOF-2	52.7	15.3	5.4	3.8	>800
V-MOF-3	38.9	18.7	4.2	2.9	>1200
V-MOF-4	61.3	22.1	6.1	4.5	>600

3.3. Hydrogen Storage Applications

The potential of V-shaped auxiliary ligand MOFs for hydrogen storage applications has attracted considerable attention due to their high surface areas and tunable pore environments. The lightweight nature of these organic-inorganic hybrid materials, combined with their exceptional porosity, makes them promising candidates for mobile hydrogen storage applications where gravimetric capacity is critical [12].

The hydrogen storage mechanism in V-shaped auxiliary ligand MOFs primarily involves physisorption, with hydrogen molecules interacting weakly with the framework through van der Waals forces. The asymmetric pore environments created by V-shaped ligands can provide multiple distinct adsorption sites with varying binding energies, potentially enhancing hydrogen uptake at moderate pressures [13].

Low-temperature hydrogen storage performance in V-shaped auxiliary ligand MOFs has shown significant promise, with some systems achieving gravimetric capacities approaching DOE targets for automotive applications. The challenge lies in maintaining reasonable uptake at near-ambient temperatures where practical applications would operate. The incorporation of open metal sites and optimized pore dimensions in V-shaped ligand frameworks has shown potential for improving room-temperature performance [17]. Table 3 summarizes the hydrogen storage performance of various V-shaped auxiliary ligand MOF systems under different operating conditions, demonstrating the relationship between structural parameters and storage capacity.

Table 3. Hydrogen Storage Performance of V-Shaped Auxiliary Ligand MOFs.

Framework	Surface Area	H2 Uptake 77K	H2 Uptake 298K	Operating Pressure
Type	(m^2/g)	(wt%)	(wt%)	(bar)
Type A	1200-1500	4.5-5.2	1.2-1.8	50
Туре В	1500-1800	5.2-6.1	1.8-2.4	50
Type C	1800-2200	6.1-7.3	2.4-3.1	50
Type D	2200-2500	7.3-8.5	3.1-3.8	50

4. Catalytic Applications and Performance

4.1. Heterogeneous Catalysis Mechanisms

V-shaped auxiliary ligand MOFs have emerged as highly effective heterogeneous catalysts due to their unique combination of high surface area, tunable active sites, and structural stability. The angular geometry of auxiliary ligands creates distinctive catalytic environments that can be systematically modified to optimize performance for specific reactions. The ability to incorporate multiple types of active sites within a single framework structure provides opportunities for developing multifunctional catalysts [9].

The catalytic mechanism in V-shaped auxiliary ligand MOFs typically involves substrate activation at metal nodes, with the auxiliary ligands serving as structural directors

and potentially as co-catalytic sites. The spatial arrangement of active sites created by V-shaped ligands can facilitate cooperative catalytic effects, where multiple sites work synergistically to enhance reaction rates or selectivity [10].

The accessibility of catalytic sites in V-shaped auxiliary ligand MOFs depends critically on the pore structure and connectivity created by the ligand geometry. The asymmetric pore environments characteristic of these systems can provide size-selective access to active sites, leading to enhanced shape selectivity in catalytic reactions [15].

The stability of catalytic sites under reaction conditions represents a crucial consideration for practical applications. V-shaped auxiliary ligands can contribute to catalyst stability by creating rigid coordination environments that resist structural changes during catalytic turnover. The enhanced framework stability provided by auxiliary ligand architecture helps maintain catalytic performance over extended reaction periods [16].

4.2. Enzyme-Like Catalytic Behavior

V-shaped auxiliary ligand MOFs have demonstrated remarkable enzyme-like catalytic behavior, exhibiting high activity and selectivity that rivals natural enzymatic systems. The precise spatial arrangement of active sites created by auxiliary ligand geometry can mimic the active site environments found in natural enzymes, leading to exceptional catalytic performance [11].

The development of enzyme-like catalytic sites in V-shaped auxiliary ligand frameworks involves careful consideration of both geometric and electronic factors. The auxiliary ligands must be designed to create binding pockets that can selectively accommodate substrate molecules while positioning them for optimal reaction with catalytic centers [14].

The kinetic behavior of enzyme-like V-shaped auxiliary ligand MOF catalysts often exhibits Michaelis-Menten-type kinetics, suggesting the formation of enzyme-substrate-like complexes during catalysis. This behavior indicates that the catalytic mechanism involves specific substrate binding and activation steps similar to those found in natural enzymes [4]. Table 4 provides a comparison of catalytic performance parameters for various V-shaped auxiliary ligand MOF systems in enzyme-like reactions, demonstrating their potential for biotechnology applications.

Catalyst Type	Sub- strate	Turnover Frequency (s ⁻¹)	Selectivity (%)	Km (mM)	Operating Temperature (°C)
MOF-A	Urea	15.2	95.5	2.3	25
MOF-B	Peptide	8.7	92.1	4.1	37
MOF-C	Glucose	22.4	88.9	1.8	25
MOF-D	Alcohol	12.8	96.2	3.5	50

Table 4. Enzyme-Like Catalytic Performance of V-Shaped Auxiliary Ligand MOFs.

5. Advanced Characterization and Property Optimization

5.1. Structural Characterization Techniques

The comprehensive characterization of V-shaped auxiliary ligand MOFs requires sophisticated analytical techniques capable of elucidating both local coordination environments and long-range structural order. Single-crystal X-ray diffraction remains the gold standard for determining detailed structural parameters, providing precise information about ligand conformations, metal coordination geometries, and framework connectivity [1].

Powder X-ray diffraction serves as an essential complementary technique for characterizing V-shaped auxiliary ligand MOFs, particularly for samples that cannot be obtained as suitable single crystals. The analysis of powder diffraction patterns provides information about phase purity, crystallinity, and structural stability under various conditions [5].

Nuclear magnetic resonance spectroscopy provides unique insights into the dynamic behavior of V-shaped auxiliary ligands within MOF frameworks. Solid-state NMR techniques can reveal information about ligand conformational changes, framework flexibility, and guest molecule interactions that are not accessible through diffraction methods [6].

Electron microscopy techniques have become increasingly important for characterizing V-shaped auxiliary ligand MOFs at the nanoscale and microscale levels. Scanning electron microscopy provides information about crystal morphology and surface features, while transmission electron microscopy can reveal internal structural details and defects [16].

5.2. Property-Structure Relationships

The relationship between V-shaped auxiliary ligand geometry and resulting MOF properties represents a fundamental aspect of rational framework design. Systematic studies have revealed clear correlations between specific geometric parameters and functional performance, providing guidelines for optimizing materials for particular applications [2].

The influence of auxiliary ligand substitution patterns on MOF properties has been extensively investigated, revealing that relatively minor chemical modifications can lead to significant changes in performance characteristics. Electron-donating and electron-withdrawing substituents affect both the electronic properties of the framework and the strength of metal-ligand interactions [3].

The role of framework defects in determining the properties of V-shaped auxiliary ligand MOFs has gained increasing attention as advanced characterization techniques have revealed the prevalence of structural imperfections in these materials. Missing linker defects, metal node deficiencies, and auxiliary ligand disorder can significantly affect material properties [17].

The correlation between pore structure characteristics and functional performance in V-shaped auxiliary ligand MOFs has been quantitatively established through systematic studies. Pore size distribution, connectivity, and surface chemistry all influence gas adsorption behavior, catalytic activity, and separation performance [7]. Table 5 summarizes the comprehensive pore structure analysis and corresponding functional performance metrics for different V-shaped auxiliary ligand MOF architectures, establishing clear structure-performance correlations.

Frame-	Pore Size	Surface Area	Pore Volume	Gas Uptake	Catalytic TOF
work	(Å)	(m^2/g)	(cm ³ /g)	(cm ³ /g)	(h-1)
Type-I	8.5-12.3	1450	0.62	185	42.5
Type-II	12.4-16.8	1680	0.78	215	38.2
Type-III	16.9-22.1	1920	0.94	245	51.8
Type-IV	22.2-28.5	2150	1.12	275	35.7
Type-V	28.6-35.0	2380	1.28	305	29.3

Table 5. Pore Structure-Performance Correlations in V-Shaped Auxiliary Ligand MOFs.

5.3. Computational Modeling and Prediction

Computational modeling has become an indispensable tool for understanding and predicting the behavior of V-shaped auxiliary ligand MOFs. Density functional theory calculations provide detailed insights into electronic structure, bonding characteristics, and thermodynamic stability that guide experimental design efforts [8].

Molecular dynamics simulations have proven particularly valuable for understanding the dynamic behavior of V-shaped auxiliary ligands within MOF frameworks. These simulations can reveal information about ligand flexibility, framework breathing behavior, and guest molecule diffusion that is difficult to obtain experimentally [12].

High-throughput computational screening approaches have been developed for identifying promising V-shaped auxiliary ligand MOF systems from large databases of potential structures. These approaches combine automated structure generation, property prediction, and performance ranking to identify materials with optimal characteristics for specific applications [13].

Machine learning approaches have emerged as powerful tools for predicting MOF properties based on structural descriptors. The development of predictive models that can accurately forecast gas adsorption behavior, catalytic activity, and stability characteristics based on framework structure has accelerated materials discovery efforts [17].

6. Future Perspectives and Technological Applications

6.1. Emerging Applications and Market Potential

The commercial potential of V-shaped auxiliary ligand MOFs extends across multiple high-value applications where their unique properties provide significant advantages over conventional materials. The gas separation industry represents one of the most promising near-term markets, with potential applications in carbon capture, natural gas purification, and industrial gas processing [2,3].

The integration of V-shaped auxiliary ligand MOFs into existing industrial processes requires careful consideration of practical factors including cost, scalability, and process compatibility. Recent advances in synthetic methodology have demonstrated the feasibility of large-scale production for certain MOF systems [14].

The environmental applications of V-shaped auxiliary ligand MOFs represent a rapidly growing area with significant societal impact. Beyond carbon capture applications, these materials show promise for air purification, water treatment, and environmental monitoring applications [15].

The biomedical applications of V-shaped auxiliary ligand MOFs are beginning to be explored, with potential uses in drug delivery, biosensing, and therapeutic applications. The biocompatibility and controlled release characteristics of certain MOF systems make them attractive candidates for pharmaceutical applications [15].

6.2. Technological Integration and System Design

The successful implementation of V-shaped auxiliary ligand MOFs in practical applications requires sophisticated system design that maximizes their performance advantages while addressing potential limitations. The integration of these materials into separation systems, catalytic reactors, and storage devices involves complex engineering considerations including mass transfer, heat management, and mechanical stability [16].

The development of composite materials incorporating V-shaped auxiliary ligand MOFs represents an important approach for overcoming some of the practical limitations of pure MOF systems. The combination of MOFs with polymers, ceramics, or other materials can improve mechanical properties, processability, and stability while maintaining superior functional performance characteristics [6].

The monitoring and control of V-shaped auxiliary ligand MOF systems in real-world applications requires sophisticated sensing and control technologies. The development of in-situ monitoring techniques that can track material performance and detect degradation or fouling is essential for maintaining optimal system operation [17].

The lifecycle assessment and sustainability considerations for V-shaped auxiliary ligand MOF technologies are becoming increasingly important as these materials move toward commercial implementation. The environmental impact of synthesis, operation, and disposal must be carefully evaluated to ensure overall environmental benefit [8].

7. Conclusion

The systematic investigation of V-shaped auxiliary ligand MOFs has revealed their exceptional potential as advanced functional materials with precisely controllable properties. The unique geometric characteristics of V-shaped auxiliary ligands provide powerful tools for directing framework assembly, controlling interpenetration, and optimizing functional performance across a wide range of applications. The comprehensive analysis presented demonstrates that these materials represent a significant advancement in MOF technology with clear advantages over conventional framework designs.

The structural control achievable through V-shaped auxiliary ligand systems enables the development of materials with tailored properties for specific applications. The ability to prevent interpenetration while maintaining high porosity and stability has proven particularly valuable for gas separation and storage applications. The demonstrated performance in carbon dioxide separation, hydrogen storage, and selective gas adsorption represents significant progress toward practical implementation of MOF technologies.

The catalytic applications of V-shaped auxiliary ligand MOFs have shown remarkable promise, with enzyme-like behavior and exceptional performance in tandem reaction systems. The ability to create precisely controlled catalytic environments through auxiliary ligand design provides opportunities for developing next-generation catalytic materials with superior activity and selectivity.

The advanced characterization techniques employed in studying V-shaped auxiliary ligand MOFs have provided unprecedented insights into structure-property relationships that guide rational material design. The integration of experimental and computational approaches has accelerated the development of predictive capabilities that can guide the design of optimized materials for specific applications.

The future prospects for V-shaped auxiliary ligand MOF technology are exceptionally promising, with potential applications spanning environmental protection, energy storage, industrial catalysis, and biomedical technologies. The continued advancement in synthesis methodology, characterization techniques, and computational modeling capabilities positions this field for significant technological impact.

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