

Mini Review

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Insights into multivariate zeolitic imidazolate frameworks

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Abstract

With the explosive growth of research focused on building units and types of crystalline materials, disruptive changes in the physical and/or chemical properties of crystals have been discovered. As the most studied subclass of metal-organic frameworks, zeolitic imidazolate frameworks (ZIFs) have shown huge potential in a wide range of applications, such as gas separation, adsorption catalysis, and so on. Specifically, when formed with multivariate (MTV) linkers or multi-metallic ions, named MTV-ZIFs, they exhibit significant differences in their thermodynamics, kinetics and properties in applications. Unraveling MTV-ZIFs, ranging from their unique structures and sequences to performance and reaction mechanisms, is crucial to further advance and expand the ZIFs. In this review, we discuss the construction methodology and properties of MTV-ZIFs, classified by MTV organic linkers and nodes, and identify challenges and opportunities, particularly linked to the chemical synthesis corresponding to their new physical chemistry. Ultimately, we outline the future direction in designing and synthesizing MTV-ZIFs to further our understanding of these promising materials.

Keywords: Zeolitic imidazolate frameworks, multivariate, chemical synthesis



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INTRODUCTION

The field of reticular chemistry and materials has been of key interest for the past 30 years. Benefiting from the flexibility of their sub-nano pore structure and specific molecular engineering, these materials showed great promise in a wide range of applications in environmental protection^[1-3], energy storage and conversion^[4-6], catalysis and cells^[7-9], sensing^[10-12], biomedicine^[13-15], chemical synthesis^[16,17], and so on^[18]. Moreover, the regulation of their topological design with the aid of chemical engineering has expanded physical chemistry and materials science into functional customization at molecular and atomic scales^[19-21]. Recently, there has been a rapid development in reticular chemistry, especially for metal-organic frameworks (MOFs) and covalent organic frameworks. For example, complex chemical entanglements (molecular weaving) have achieved breakthroughs in the design principles of traditional molecular cages^[22,23]. In addition, the emergence of glassy MOFs provided a new perspective for understanding the formation mechanism of vitrification, and a new platform for functional glasses^[24]. Meanwhile, several scalable synthetic methods and practical applications have been implemented at pilot and even on a larger scale. In Oct 2023, BASF announced a 100 tons per year level mass production of MOFs for CO₂ capture^[25]. Additionally, since using MOFs for water harvesting in 2020^[26], Yaghi *et al.* have now applied them in labs^[27], deserts^[28-31], and even when teaching in classrooms^[32]. Reticular materials have begun to penetrate into every corner of our lives. They are gaining attention due to their tailor-made multifunctional properties and potential for widespread applications.

Among the various approaches to advancing reticular materials, diversifying their building units is one of the most inspiring, resulting in a new field, multivariate MOFs (MTV-MOFs). Recently, an Editorial article from *Chemistry of Materials* also identified this direction of MOFs^[33], suggesting that future research will release more intrinsic or even produce new chemical properties of MOFs through MTV components. For carboxylate-based MOFs, there has been a lot of research on developing MTV-MOFs [Figure 1]^[34-37]. As a typical example, 1,4-benzenedicarboxylate can be modified by -NH₂, -Br, -Cl, -NO₂, or other functional groups at positions 2, 3, 5, and 6 on the benzene ring. Through the combination of these linkers, an optimized host-guest interaction, such as a 400% better selectivity for carbon dioxide compared to carbon monoxide, can be obtained [Figure 1A]^[35]. By utilizing MTV linkers, various structures, such as hierarchical pores^[38], layered pores^[39], mixed pores^[40,41], and even innovative cage structures^[42-44], have therefore been provided. Multi-metallic (secondary building unit) MTV-MOFs have been concerned as well [Figure 1B]^[45,46]. They can be obtained through the *in-situ* synthesis^[47] or post-modification^[48] method. The sequence of these metal ions can then be identified through energy-dispersive X-ray spectroscopy^[49], integrated differential phase contrast-scanning transmission electron microscopy^[50] or atom probe topography^[51]. Finally, the differences in multi-metallic MTV-MOFs lead to the multifunctional properties inherent to their specific metal sequence^[52-55].

Unsurprisingly, the above studies are mostly based on several classic carboxylate-based MOFs, such as MOF-5, MOF-808 and PCN-600. The linkers with terephthalic acid (MOF-5)^[34] or tricarboxylic acid (MOF-808)^[56] as their backbone have abundant, scalable architecture to form MTV-MOFs, while porphyrin structures (PCN-600)^[49] provide suitable anchors for metals. However, the linkers used in carboxylic MOFs have large steric hindrances in the chemical synthesis, making it difficult to obtain well-mix MTV-MOFs. The presence of functional groups will affect the steric configuration of linkers, i.e., the dihedral angle of non-coplanar binding groups and the angle of binding groups bent toward each other^[57], which could lead to disordered structure. As a subclass of MOFs, zeolitic imidazolate frameworks (ZIFs, including zeolitic triazole framework, zeolitic tetrazole framework, and so on) have unique characteristics compared to other MOFs. On the one hand, due to the zeolite-like structure, they can form various topological cages with smaller chemical linkers [Figure 2]. On the other hand, its node is a single metal ion; thus, the consideration

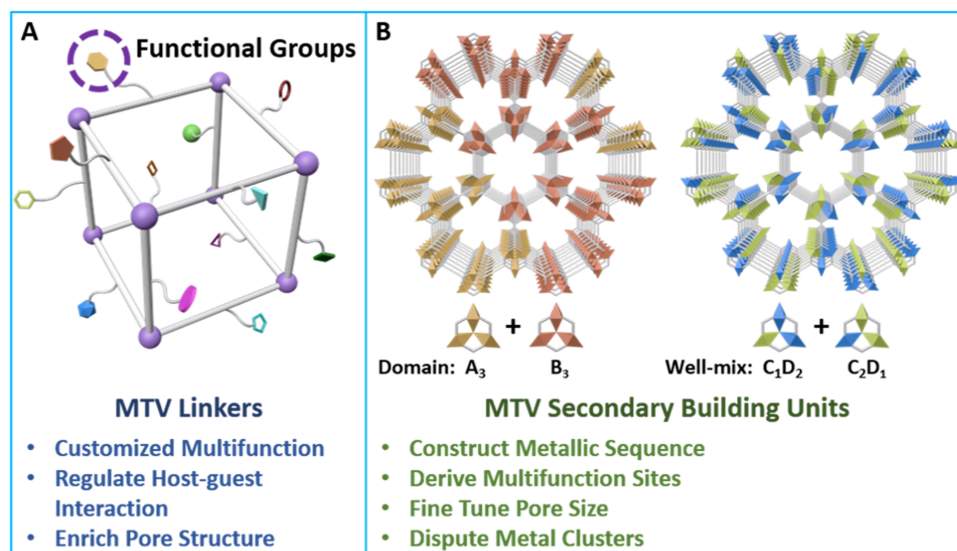


Figure 1. MTV-MOFs with different (A) functional linkers and (B) secondary building units. MTV: Multivariate; MOFs: Metal-organic frameworks.

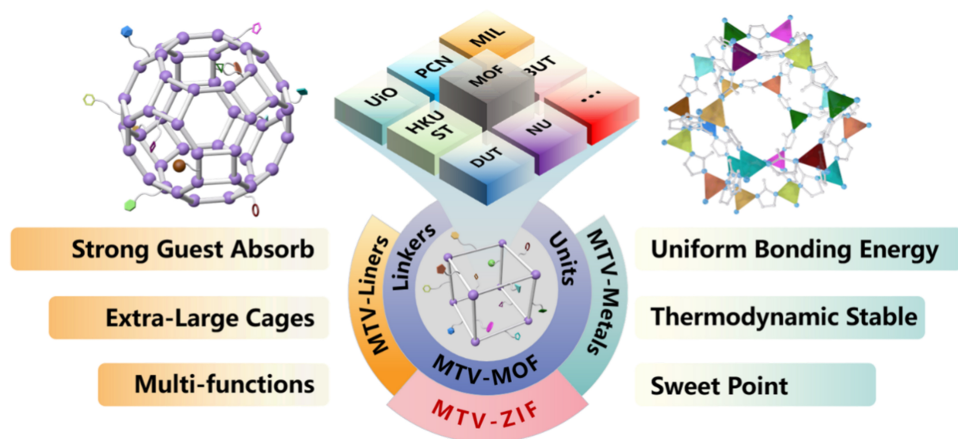


Figure 2. Diagram of MTV-ZIF. MTV: Multivariate; ZIF: zeolitic imidazolate framework.

of the variable of secondary building units becomes redundant [Figure 2]^[49]. Additionally, their chemical functions and pore size are strictly interdependent, which is different from carboxylate-based MOFs^[58]. Therefore, ZIFs are the optimal choice for exploring the intrinsic properties of MTV-MOFs. As a typical example, the combination of boron and other metal nodes in ZIFs leads to a higher volumetric CO₂ storage capacity of 81 L/L compared to their single metal counterparts. Other MTV-ZIFs were found to improve electrocatalysis, photocatalysis, photoluminescent and mechanochromic properties^[59]. Specifically, a Zeolite Lynde Type A (LTA) framework BIF-20 [Zn₂(BH(mim)₃)₂(obb)₂, obb = 4,4'-oxybis(benzoate)] exhibits a remarkable initial hydrogen uptake of 1.43 wt% at 77 K and 1 atm, which is higher than ZIF-20 [Zn(Pur)₂] with the same topology^[60]. Many other reports have also confirmed that diversified ZIFs have optimized or even displayed completely new performance^[61-66].

Herein, we present a timely review article to give a deep understanding of MTV reticular chemistry and previously yet probably ignored findings in the MTV-ZIFs. We approach its most recent advancement from

two main routes: MTV linkers and metal nodes. In addition to the structural design and synthesis strategy of MTV-ZIFs, physicochemical structure-activity relationships, and some research methods will also be proposed. Ultimately, future development directions for MTV-ZIFs or MTV-MOFs will be provided.

MTV LINKERS: UNIFICATION OF SIZE AND PROPERTIES



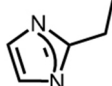

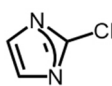
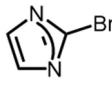
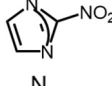
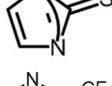
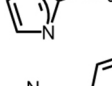
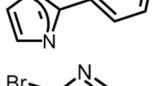
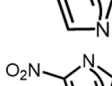
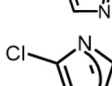
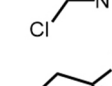
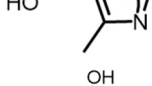
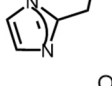
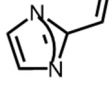
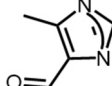
The organic linkers in MTV-ZIFs

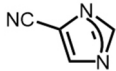
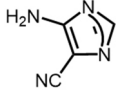
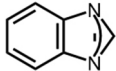
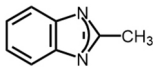
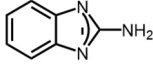
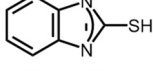
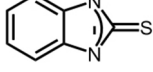
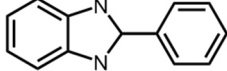
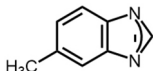
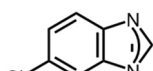
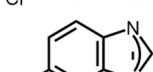
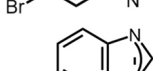
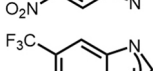
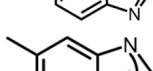
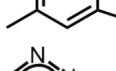
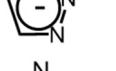

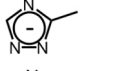
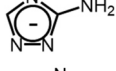
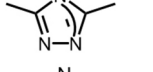
Before introducing the construction from MTV linkers into ZIFs, we first recommend imidazolate (IM)/imidazolate-type chemical linkers because they are the basic building units that have significant impacts on the structure of ZIFs^[58]. So far, the most common linkers are IM, benzimidazolate (BIM) and their analogous derivatives, which contain possible chemical substituents on the position 2, 4, or 5 sites of IM (2, 5, or 6 of BIM). They are also more controllable in synthesis due to the two active nitrogen atoms with adjacent chemical steric hindrance than other carboxylate linkers. Several studies have also used triazole or tetrazole to construct zeolite-like frameworks^[67-69]. These linkers can serve as linkers for regulating host-guest interaction in MTV-ZIFs^[70]. Nonetheless, it faces complex challenges to avoid the reactions at each nitrogen position. Therefore, several non-zeolitic frameworks have been constructed using triazole, tetrazole and pentazole^[67,71,72]. Undoubtedly, although they have innovatively expressed a new type of skeleton, indeed makes no sense to MTV-ZIFs due to the violation of the basic topological structure of zeolite (the bridging angle of linkers and nodes)^[73]. These non-zeolitic frameworks and some zeolitic non-imidazolate frameworks, such as zeolite organic frameworks^[74], will, therefore, not be considered herein. Based on such considerations, we summarized all linkers that have the potential to construct MTV-ZIFs in [Table 1](#), excluding some complex linkers.


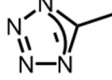
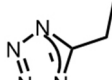
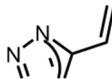
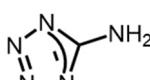
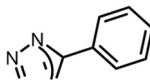
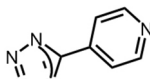

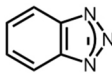
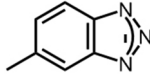
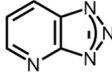
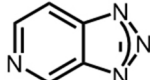
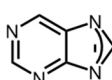
Control opening in cages

Using the above linkers, different MTV-ZIFs are built under the guidance of molecule engineering methodology. In 2008, Banerjee *et al.* first presented ten MTV-ZIFs among 25 new ZIFs prepared by high-throughput synthesis under multi-component chemical conditions, including ZIF-60 [Zn(IM)_{1.5}(2-mIM)_{0.5}], ZIF-61 [Zn(IM)(2-mIM)], ZIF-62 [Zn(IM)_{1.75}(BIM)_{0.25}], ZIF-68 [Zn(BIM)(2-nIM)], ZIF-69 [Zn(5-cBIM)(2-nIM)], ZIF-70 [Zn(IM)_{1.13}(2-nIM)_{0.87}], ZIF-73 [Zn(2-nIM)_{1.74}(5,6-mBIM)_{0.26}], ZIF-74 [Zn(2-nIM)(4,5-mBIM)], ZIF-75 [Co(2-nIM)(4,5-mBIM)] and ZIF-76 [Zn(IM)(5-cBIM)]^[79]. Subsequently, the binary combination of the above linkers controls the topological structure of ZIFs, which has also been confirmed in two studies^[86,105]. At the same time, some works reported that the different openings in cages could be controlled by the introduction of another linker, typically, i.e., replacing 2-mIM with BIM [[Figure 3A](#)]^[62,106,107]. However, constructing MTV-ZIFs remains unpredictable until a novel and rational principle is established to give design criteria for extra-large cages. In 2017, Yang *et al.* reported a direct synthesis of a large-sized ZIF with 46 Å {ZIF-412, [Zn(BIM)_{1.13}(2-nIM)_{0.62}(IM)_{0.25}]}^[58], demonstrating the functionality of MTV-ZIFs in synthesizing large cage or pore size in ZIFs. In brief, through the combination of the large ring formed by the large (steric index) linkers and the small ring by the small linkers, the much larger cages would be achieved in ZIFs^[108]. Large linkers can provide a large steric index in chemical synthesis, while it is essential for constructing large cages with large rings [[Figure 3B](#)]. This also conforms to the principle of the relationship between vertices and cages^[109]. However, to overcome the kinetics of the reaction, small linkers must be introduced based on the linkers with large steric index due to the rules of Euclidean space, as unprecedented ZIF-412 mixed IM, 2-nIM and BIM [[Figure 3C](#)]. MTV linkers are the key to exceeding the size of ZIFs. It will be more conducive to storing adsorbed gases or liquids, while breaking the adsorption restrictions on certain organic vapors^[58,105,110]. As a demonstration of absorbing volatile organic compounds, the mesoporous ZIF-412 can uptake 3.4 mmol/cm³ *p*-xylene vapors at a partial pressure of 298 K and a low relative pressure (P/P₀ = 0.1), which is higher than the commercial adsorbents and other ZIFs^[58].

Table 1. A summary of proper linkers for building MTV-ZIFs

Name	Abbreviation	Formula	Ref.
Imidazolate	IM		[75]
2-methylimidazolate	2-mIM		[75]
2-ethylimidazolate	2-eIM		[76]
2-propylimidazolate	2-pIM		[77]
2-chloroimidazolate	2-clIM		[78]
2-bromoimidazolate	2-bIM		[78]
2-nitroimidazolate	2-nIM		[79]
2-mercaptoimidazolate	2-S=IM		[80]
2-trifluoromethylimidazolate	2-CF ₃ -IM		[81]
2-phenylimidazolate	2-phIM		[82]
4-bromoimidazolate	4-bIM		[83]
4-nitroimidazolate	4-nIM		[58]
4,5-dichloroimidazolate	4,5-clIM		[79]
4-hydroxymethyl-5-methylimidazolate	4-hm-5-mIM		[84]
Imidazolate-2-carboxaldehyde	IM-2-OH		[85]
Imidazolate-2-carboxyaldehyde	IM-2-CHO		[85]
4-methylimidazolate-5-carboxaldehyde	4-mIM-5-CHO		[84]

Imidazole-4-carbonitrile	IM-4-CN		[86]
5-aminoimidazole-4-carbonitrile	5-aIM-4-CN		[84]
Benzimidazole	BIM		[75]
2-methylbenzimidazole	2-mBIM		[87]
2-aminobenzimidazole	2-aBIM		[88]
2-thiolbenzimidazole	2-SH-BIM		[89]
2-mercaptobenzimidazole	2-S=BIM		[90]
2-phenylbenzimidazole	2-phBIM		[91]
5-methylbenzimidazole	5-mBIM		[86]
5-chlorobenzimidazole	5-cBIM		[79]
5-bromobenzimidazole	5-bBIM		[58]
5-nitrobenzimidazole	5-nBIM		[86]
5-trifluorobenzimidazole	5-CF ₃ -BIM		[92]
5,6-dimethylbenzimidazole	5,6-mBIM		[79]
1H-1,2,3-triazolate	1,2,3-TZ		[67]
1,2,4-triazolate	1,2,4-TZ		[93]
3-methyl-1,2,4-triazolate	3-m-1,2,4-TZ		[94]
3-amino-1,2,4-triazolate	3-a-1,2,4-TZ		[95]
3,5-dimethyl-1,2,4-triazolate	3,5-m-1,2,4-TZ		[96]
3-amino-5-methyl-1,2,4-triazolate	3-a-5-m-1,2,4-TZ		[97]

Tetrazolate	TZ		[87]
5-methyltetrazolate	5-mTZ		[77]
5-ethyltetrazolate	5-eTZ		[98]
5-vinyltetrazolate	5-vTZ		[98]
5-aminotetrazolate	5-aTZ		[99]
5-phenyl-tetrazolate	5-phTZ		[100]
5-(4-pyridyl)-1H-tetrazolate	5-(4-pd)TZ		[101]
Pentazolate	PZ		[71]
Benzotriazole	BTZ		[102]
5-methylbenzotriazole	5-mBTZ		[103]
4-azabenzimidazole	4-ABIM		[104]
5-azabenzimidazole	5-ABIM		[104]
1H-Purine	Pur		[104]

MTV: Multivariate; ZIFs: zeolitic imidazolate frameworks.

Intensifying multifunctionality by MTV

Building MTV-ZIFs is also the best practice for constructing multifunctional ZIFs. For example, introducing 2-nIM or 2-pIM in ZIF-8 [$\text{Zn}(2\text{-mIM})_2$] can alter the physical and chemical properties on the surface and inside the pores. Many studies have demonstrated the role of functional groups in host-guest interaction^[80,88,111-113]. Moreover, there are further reports on inspiring physical and chemical properties, including hydrostability^[114,115], surface wettability^[116], interfacial compatibility^[117] and catalytic performance^[118]. Li *et al.* regulated the surface adhesion ranging from 3.62 to 15.15 nN by controlling the second linker in ZIF-L [$\text{Zn}(2\text{-mIM})_2 \cdot (2\text{-mIM})_{0.5} \cdot (\text{H}_2\text{O})_{1.5}$], including IM, 2-eIM, 2-bIM and BIM, achieving modulating superlubricity^[119]. Another interesting example is about ZIF glass where many works have confirmed that MTV-ZIFs have unique amorphous properties owing to the disordered structure and multi-components, compared to their crystalline partners^[120-122]. We systematically studied influences of MTV

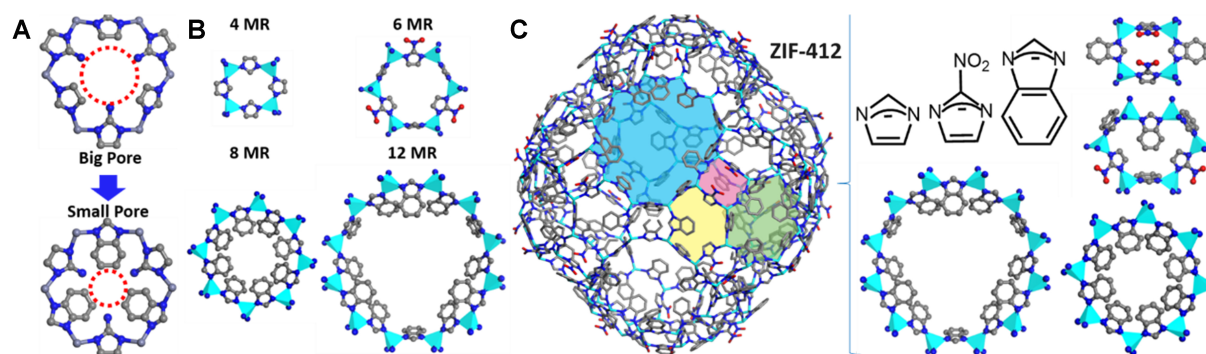


Figure 3. The principle to design and synthesize large cages in MTV-ZIFs. (A) Schematic diagram of linkers induced pore changes; (B) Linker-depended opening ring; (C) MTV towards extra-large cages. Take ZIF-412 as an example. N, blue; C, dark; O, red; and ZnN_4 units, light blue tetrahedra. H atoms are omitted for clarity. MTV: Multivariate; ZIFs: zeolitic imidazolate frameworks.

linkers on the properties including porosity, glass transition temperature and mechanics of ZIF glasses by comparing ZIF-62 [$\text{Zn}(\text{IM})_{1.75}(\text{BIM})_{0.25}$] to ZIF-4 [$\text{Zn}(\text{IM})_2$]^[24]. Therefore, by combining the linkers in Table 1, it is possible to modulate the functions of ZIFs and apply them in various environments^[123]. However, it should be noted that imidazolate-2-carboxylic acid with powerful host-guest interaction is hard to control as a linker due to the competitive reaction between carboxylic acids and IM ^[124]. Therefore, there are currently no substantial advancements in this powerful linker intensifying the function of ZIF. Therefore, we can conclude that designing and building MTV-ZIFs are able to customize more functional reticular materials.

The introduction of IM-type linkers in the latter half of Table 1 greatly alters the properties of the original ZIFs. The introduction of triazolates and tetrazolates supplies uncoordinated N sites in the frameworks, which enhance host-guest interactions^[69] and mechanical properties^[125]. Therefore, we can regulate the host-guest interactions of ZIFs by adjusting the appropriate ratio of triazolates and tetrazolates salt. Li *et al.* reported the optimal activity by encapsulating *Burkholderia cepacia* lipase (BCL) within ZIFs built by 2-mIM, 3-m-1,2,4-TZ and 5-mTZ. The optimizing results of asymmetric catalysis indicated that the best MTV-ZIF (with 23.5% 2-mIM and 76.5% 3-m-1,2,4-TZ) have a remarkable enantioselectivity of 4-phenyl-3-buten-2-ol (99% enantiomeric excess). Interestingly, a component-adjustment-ternary plot has been drawn to explore the best performance of MTV-ZIFs^[70]. However, the chemical synthesis of such MTV-ZIFs is facing certain challenges; typically, i.e., the nitrogen sites in MTV linkers arbitrarily react with the metal ions to form non-ZIF frameworks^[71,126]. Although they (or the so-called metal azolate framework, MAF) have interesting structures with impressive performance, it is not conducive to the formation of MTV-ZIFs. Therefore, beyond reports providing examples of controllable synthesis of such MTV-ZIFs^[94], we still need to ensure that the designed and synthesized MTV-ZIFs have accuracy, especially in controlling the thermodynamics and kinetics of chemical synthesis at these excess nitrogen sites.

Challenges and potential solutions in chemical synthesis

MTV-ZIFs offer unconventional apertures and customized functionality. However, achieving a harmonious equilibrium between the two linkers becomes arduous because the nature of the linkers dictates both functionality and the apertures. Another consideration is that most ZIFs have allotropes^[127-129]. Therefore, customizing MTV-ZIFs is suffering from significant difficulties in chemical synthesis, such as in controlling the function and concurrently maintaining the cage structure, which remains a daunting chemical challenge. To address the above challenges, the first strategy is to utilize organic reactions to functionalize existing linkers (named linker functionalization strategy in Figure 4A)^[85,130]. It is just suitable for several

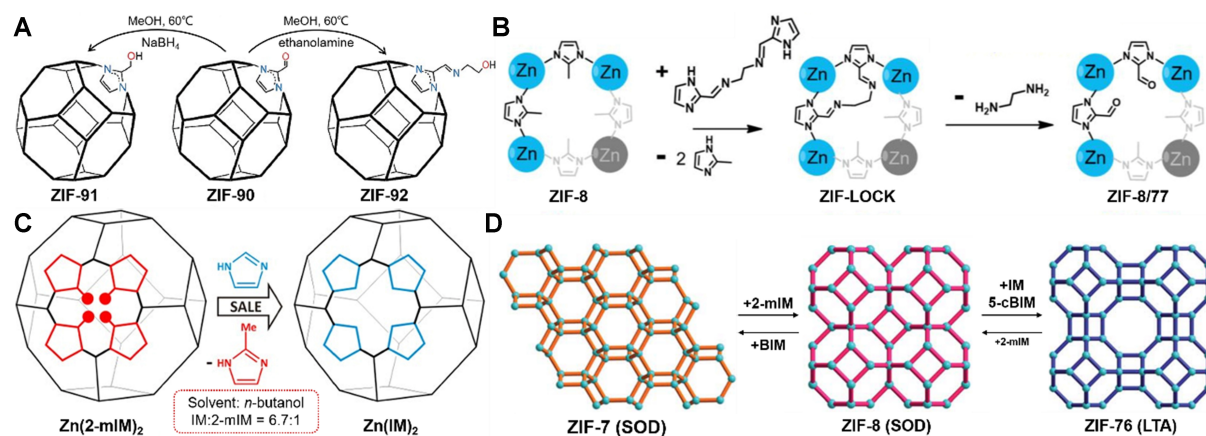


Figure 4. Diagram of (A) linker function strategy, (B) opening the lock strategy, and (C) SALE strategy; (D) Topological structure changes with linker exchange. (A) Adapted with permission^[85]. Copyright 2008, American Chemical Society; (B) Adapted with permission^[135]. Copyright 2019, American Chemical Society; (C) Adapted with permission^[138]. Copyright 2012, American Chemical Society; (D) Adapted with permission^[139]. Copyright 2020, The Royal Society of Chemistry. SALE: Solvent-assisted linker exchange.

linkers such as IM-2-CHO, while the functionalized groups have a limit of steric hindrance in an original cage^[85,131-134]. Another one is opening the lock strategy based on chemical confinement by controlling the size of the cage and considering the functional part [Figure 4B]^[135]. Specifically, it first used carbon chains to lock in two IM linkers without affecting the coordination of N, and then removed the carbon chain after forming MTV-ZIFs. This strategy can endow MTV-ZIFs with multiple channels and functional groups, while only a few linkers can be used to form carbon chains and unlock them.

Benefiting from the small ligands of ZIFs, the linker exchange strategy, especially the solvent-assisted linker exchange (SALE) strategy^[136], is currently a worthwhile and potential approach to unified functionalities and aperture [Figure 4C]. Mixing the prepared ZIFs, typically, i.e., ZIF-8 and ZIF-67 [Co(2-mIM)₂], with excess other linkers can achieve linker exchange under a certain condition^[112,137]. The 2-mIM can be substituted with almost all IM-type linkers, including IM^[138], 2-cIM^[112], 2-bIM^[112], 2-nIM^[89], 2-pIM^[89], 4-bIM^[83], 4,5-cIM^[139], 4-mIM-5-CHO^[140], 2-S=IM^[80], BIM^[141], 2-aBIM^[89], 2-SH-BIM^[89], 2-CF₃-BIM^[112], 5-cBIM^[139], 5,6-mBIM^[114], 1,2,3-TZ^[142], 3-a-1,2,4-TZ^[130], BTZ^[103], 5-mBTZ^[103] and some special linkers^[143,144]. Similar to 2-mIM, 2-nIM can also be exchanged with some linkers^[145]. However, a few of them can not ensure the formation of topological structure, causing changes in the aperture [Figure 4D]. This would make the chemical synthesis and structure uncontrollable towards MTV-ZIFs. It is relatively easy to maintain the topology of linkers with the same steric hindrance and chemical activity^[138], as can be seen in several examples of substituted BIM^[92,102]. Nevertheless, the SALE is encountering several problems, i.e., the difficulties in controlling the ratio of these MTV linkers and the lack of quantitative study describing the substitution kinetics in these SALES. Only IM to 2-mIM^[138], BIM to 2-mIM^[141], and BTZ to BIM^[102] are free to replace with the topology keeping. Moreover, it should be noted that the linker exchange in the center of the powder is hard to conduct. Marreiros *et al.* and Jiang *et al.* tried to use the vapor phase^[146] and mechanochemistry^[147] instead of the only solvent environment to solve this, respectively. However, the results still showed that characterizing and controlling the proportion of linkers in the core and shell of ZIF powder remained challenging. In brief, although there have been many attempts to replace various linkers in ZIFs, more explorations are still required in chemical synthesis and crystallography mechanisms. The above three methods are all post-synthetic modifications, while *in-situ* methods could be more effective although they suffer from a complex ionic solution environment. As artificial intelligence advances, it may help us develop a suitable *in-situ* synthesis scheme to prepare MTV-ZIFs.

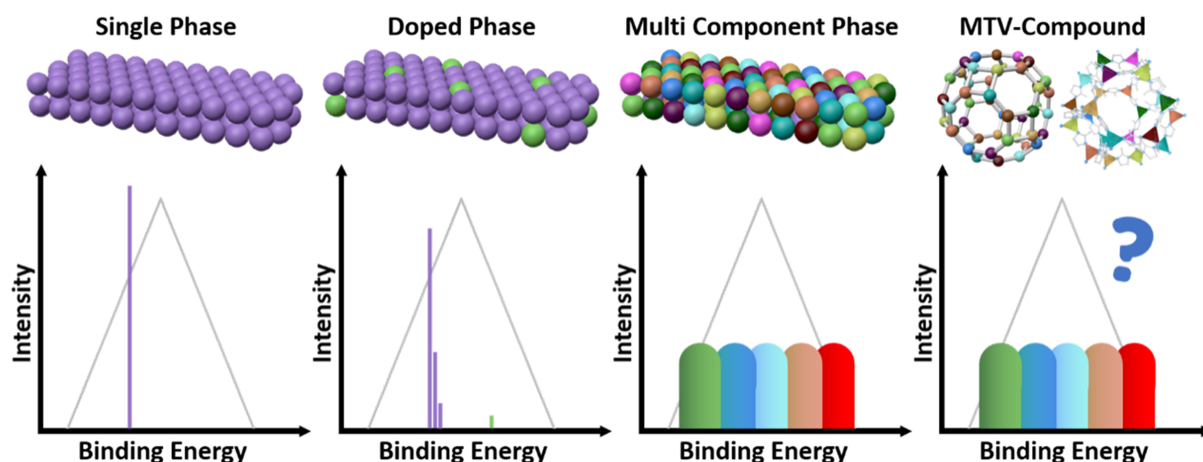


Figure 5. Diagram of the potential multifunctional catalysis through the binding energies of single phase, doped phase, multi-component phase and MTV-compound. MTV: Multivariate.

As a new frontier in MOFs, the control of multiple linkers poses a challenge to chemical synthesis. Although MTV-ZIFs are defined in material, their design, synthesis, and application are all based on reticular, coordination, and organic chemistry. With strong chemical guidance, we believe that we can obtain MTV-ZIFs with any function and pore size in theory.

MTV METAL NODES: ORDER IN DISORDER

What would be brought by MTV metals

In many MTV materials, polymetallic structures represent multiple functionalities. As a typical example, high-entropy alloy nanoparticles exhibit continuous binding energies for the on-top binding of a reactant, which was never seen in single-phase materials [Figure 5]^[148]. As the simplest MTV material, high-entropy alloys made us re-examine the role of entropy brought by the MTV metals in affecting the thermodynamics and kinetics of materials, while bringing some unprecedented peculiar properties, including the compromise between strength and toughness that has been broken by high entropy alloys, as evidenced by many pieces of literature^[149-151]. This forces us to re-emphasize the significance of MTV metal nodes in ZIFs. In an electrocatalytic reactant, whether the binding energies of these catalytic sites may be separated by the linkers, or may be consistent with those exhibited by high-entropy alloys. Moreover, as mentioned by Gagliardi and Yaghi, “What spatial arrangement is underlying this heterogeneity and could it be used to create new chemistry”^[33]. We can expect the MTV reticular chemistry to be a new frontier in MOFs, especially in a mix-well MTV nodes system.

Substantial reports have asserted the importance of the MTV metal ions in ZIFs. Metals play a decisive role in the adsorption, storage, catalysis, and detection of the guests^[45,46,152]. So far, both zinc(II) and Co(II) are the most frequent metal ions among all the nodes. These two elements can be mixed and formed into bimetallic ZIFs through various synthesis methods, such as room temperature stirring, which is different from other polymetallic ZIFs [Figure 6A]^[153]. In terms of other metals, however, there are thermodynamic limitations and kinetic hindrances inside the ZIFs. The atoms with strong coordination activity can be well mixed in ZIFs, while weak ones have a tolerant ratio in ZIFs and may even lead to the formation of other topologies [Figure 6B and C]. Our functional hybrid material lab (FHML) has previously analyzed the characteristics of CoZn-ZIF systematically^[153]. It follows that the CoZn-ZIF shows a higher activity (57%) than the single metal ZIFs (40% for Zn and 50% for Co) in the cycloaddition of CO₂ to epichlorohydrin at 353 K and 1 atm for 24 h. Whether applied in adsorption or catalysis, bimetallics have superior performance

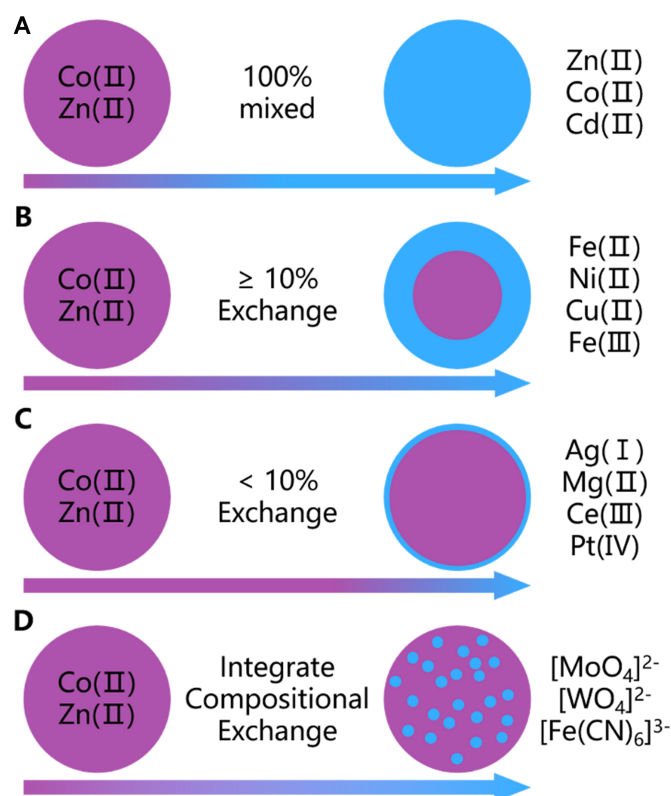


Figure 6. The schematic diagram of (A) well-mixed metals, including Zn(II), Co(II), Cd(II)^[156], and Fe(II)^[212] [Fe(II) is added with solvent-free method]; (B) secondary metals, including Mn(II)^[213], Fe(II)^[164], Fe(III)^[214], Ni(II)^[215], Cu(II)^[215], B(III)^[216] and so on [Fe(II) is added with room temperature stirring method]; (C) weak metals, including Ag(I)^[165], Pt(II)/(IV)^[217], Ce(III)/(IV)^[166], and so on; (D) integrate compositional exchange of specific metals such as Mo^[196], W^[196], Pd^[201] and Fe^[66].

over unary metals as verified by many studies^[154]. As a mechanism, the synergistic effect is often used to explain the superiority of bimetallics over single metals. Therefore, it comes to two questions: what is the highest point of synergy in MTV metal nodes, and what is the physical chemistry behind this effect?

Find “sweet spot” in MTV nodes

The ternary diagram has demonstrated the feasibility of searching for the sweet point of the ratio of azole-linkers on the adsorption enzyme, which is probably extended to MTV nodes [Figure 7]. Hou *et al.* found that a good balance of the ratio of different nodes in CoZn-ZIFs leads to the optimized crystal structure and membrane separation performance^[155]. Specifically, the trade-off at 18% of Co(II) enables MTV-ZIFs to separate C₃H₆ from C₃H₈ with a separation factor of 200. In addition to the relationship between crystalline structure and the ratio of metal nodes, there is also a balance between proportion and performance, or mixing degree^[156-158]. In a nutshell, constructing this type of graph can quickly help researchers identify the sweet point toward the desired application.

New physical chemistry behind the MTV nodes

The different sizes and intrinsic properties in metal ions are the main factors affecting MTV ZIFs, and they will result in the entire framework not being a strictly ideal and defined structure. In topologically similar ZIFs, the second metal would contribute to the enthalpy of topological formation^[159]. Specifically, a larger ionic radius leads to a tendency of the sodalite (SOD) rather than the diamondoid (*dia*) topology [Figure 8A]. Moreover, incorporation of metal nodes with higher electronegativity would increase the possibility of forming the *dia*-framework, as well as SOD. However, the effect of metal nodes on entropy

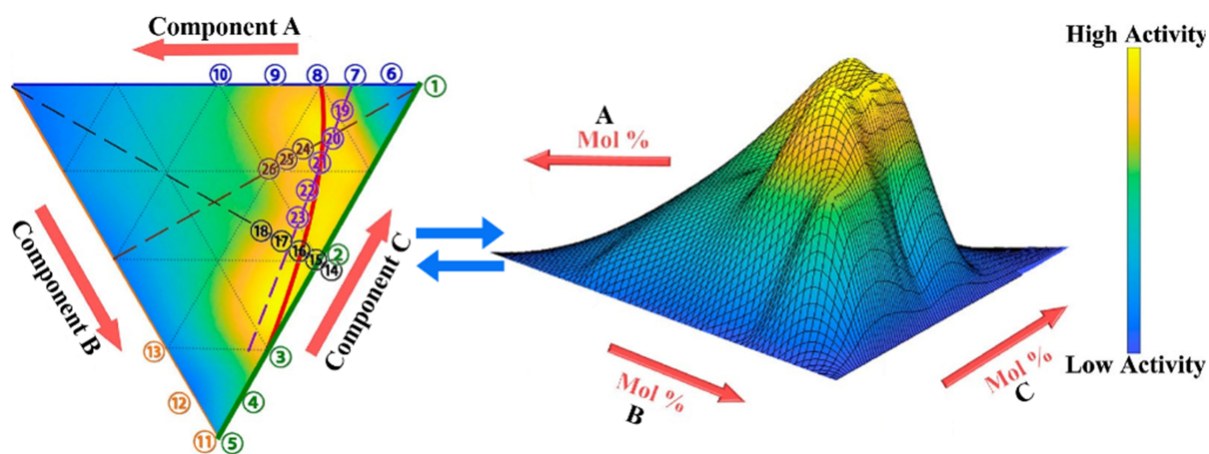


Figure 7. The ternary diagram assists in predicting the sweet points. The dark colors (red, yellow) indicate the better points, while light colors (green, blue) indicate the worse points. Adapted with permission^[70]. Copyright 2021, American Chemical Society.

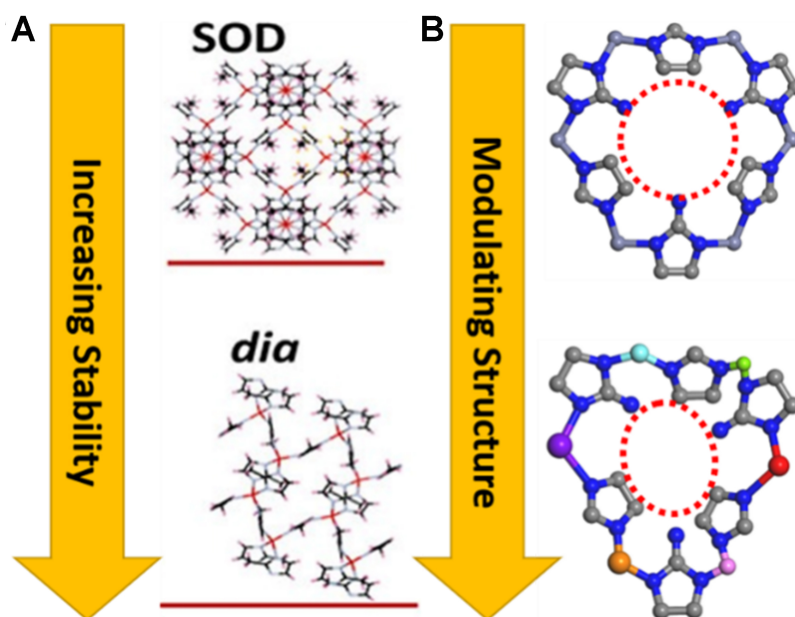


Figure 8. The schematic diagram of (A) the structural tendency of larger metal ions combined with 2-mIM and (B) a potential strategy for modulating pore structure by MTV-ZIFs. (A) Adapted with permission^[159]. Copyright 2023, American Chemical Society. 2-mIM: 2-methylimidazolate; MTV: Multivariate; ZIFs: zeolitic imidazolate frameworks.

was not considered here. It is obvious that the cooperation of larger ions will make the framework more stable due to the high-entropy effect, and at the same time, high entropy will cause the enthalpy difference between the two structures mentioned above to be ignored. Therefore, the structure disorder of ZIFs^[160] will be optimized through MTV nodes, which would be completely inconsistent with other high-entropy materials^[161]. For example, it is difficult to prepare large cage ZIFs with the MTV linkers, such as ZIF-412. By comparison, in order to get the complex reticular structure, it may be more favorable to introduce more metal ions especially the ones with larger radius. Merely, it should be noted that metals have an influence on the aperture. They will fine-tune the pore size within a very small range, which will be beneficial for our precise molecular separation or ion sieving [Figure 8B]^[162]. Back in terms of enthalpy, the synthesis of MTV

nodes brings about high mixing-enthalpy and mixing-entropy for largely improving kinetics. Therefore, it is difficult for scholars to generate MTV-ZIFs by simply multiplying the metal ions, which we would discuss in the next section.

Meanwhile, the chemical bonding character of metal and linkers can affect the properties of MTV-ZIFs, such as host-guest interaction. A previous theory calculation has proved that there is a stronger interaction between metal nodes of CdZn-ZIF and CO₂ molecules^[163]. Moreover, multiple metal ions in MTV-ZIFs may also bring stronger adsorption effects as confirmed by many works^[154,164-169]. It seems that they get the same results as the experimental verification. Furthermore, certain unique properties of specific metal ions will significantly alter the overall ZIF properties. For example, introducing the redox-active Co, Cu or Fe could enhance the electrochemical and photochemical performance at varying degrees^[170,171]. Merely, most of the research focused on these MTV-ZIFs tends to use them as an excellent template for generating powerful catalysts through high-temperature carbonization^[172-175]. Although these works provided good ideas for the atomic design of catalysts, we should focus more on ZIFs themselves.

Challenges and strategies in chemical synthesis

In addition to overcoming the thermodynamic barriers of chemical reactions in traditional ZIFs, it is also necessary to overcome the mixed Gibbs energy in MTV-ZIFs. Therefore, only a few articles focus on the trimetallic ZIFs and beyond^[176-178]. Xu *et al.* pioneered a mixture of the Ni(II), Cd(II), Co(II), Cu(II), and Zn(II) in high-entropy ZIFs by the mechanochemical synthesis^[154]. In a mechanochemical synthesis, they used extremely low energy input to enable multiple metals to only form MTV-ZIFs without excess energy to assist in the diffusion and substitution of active-less metals by highly active metals. For those extreme MTV-ZIFs, some limited approaches should be considered. We can learn, however, from high entropy alloys how to control atomic heat and mass transfer in environments where linkers are added. In addition to extremely low energy, fast-energy flashing (joule-heating^[179] or laser-synthesis^[180]) may also help form MTV-ZIFs. The high-temperature breakthrough in the thermodynamics of reactions, combined with limited reaction time, prevents atoms from aggregation, forming the metastable ZIFs^[181]. These *in-situ* preparation strategies have extremely high requirements for the synthetic conditions. In comparison, there are some promising post-synthetic modifications leading to more adjustable metallic ratios, such as cation exchange^[182].

Though cation exchange is successful in many works, we must carefully consider the thermodynamic and dynamic activity of the reactions involved. For example, in terms of that Zn(II) was exchanged by Ag(I), some reports only show that surface Zn(II) has been exchanged^[183] [Figure 9A], while there are also claims that controllable total exchange has been achieved^[184] [Figure 9B], even that the replacement of Ag(I) could damage the structure of ZIFs^[185] [Figure 9C]. On the other hand, as mentioned above, some metal complexes can cause structural changes in MTV-ZIFs^[186] [Figure 9D]. Therefore, these are what we should examine and control strictly in post-synthetic modifications. In a nutshell, there were reports about ZIF-8/ZIF-67 via cation exchange with Ti(IV)^[187], Fe(II)^[188], Ni(II)^[189], Cu(II)^[190], Mn(II)^[83,191], Cd(II)^[192], Sn(II)^[193], Li(I)^[194], Na(I)^[194], or Ag(I)^[184]. It is interesting to note that we seem to be able to achieve hyper MTV-ZIFs, which have specific metal sequences, through gradual permutation of cation exchange. We can draw conclusions from the calculation of ion reaction activity sequences and design some experiments^[195]. However, it cannot be denied that the real environment differs greatly from the molecular reactions in the calculation. There is a lot of reaction competition among them, including metal ion–metal ion *vs.* metal ion–linker, and molecule–metal *vs.* cage–metal.

The concept of exchange is not limited to cations; some works incorporate ion groups into ZIFs to achieve permutation [Figure 6D]^[196]. [MoO₄]²⁻ and [WO₄]²⁻ are currently the most reported and suitable molecules

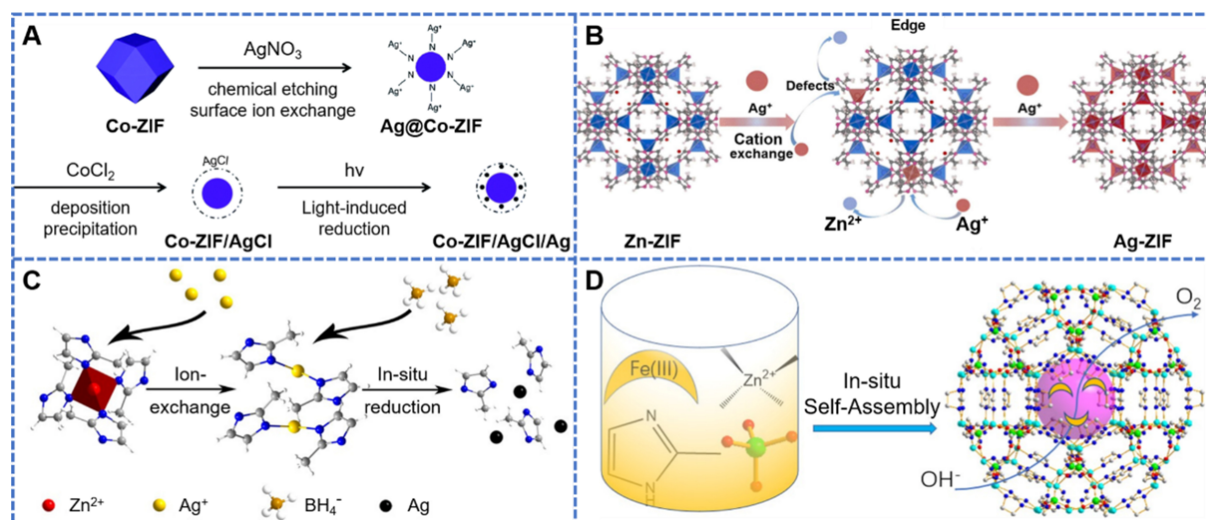


Figure 9. The schematic diagram of (A) the surface cation exchange, (B) free cation exchange, and (C) destructive cation exchange with an example of Ag⁺ replacing Zn²⁺; The schematic diagram of (D) shows a new structure in MTV-ZIFs. (A) Adapted with permission^[183]. Copyright 2020, The Royal Society of Chemistry; (B) Adapted with permission^[184]. Copyright 2022, Elsevier; (C) Adapted with permission^[185]. Copyright 2020, American Chemical Society; (D) Adapted with permission^[186]. Copyright 2022, American Chemical Society. MTV: Multivariate; ZIFs: zeolitic imidazolate frameworks.

for substitution in ZIF-8^[197,198]. However, these reports suggest that the addition of such molecules has brought greater uncontrollable topology disorder^[199,200]. Moreover, it is difficult to find suitable molecules for doping in ZIFs. In addition to the two mentioned above, only [PdCl₄]²⁻ was previously considered^[201] and an embedding of [Fe(CN)₆]³⁻ has also been reported^[66]. This integration-doped idea can significantly regulate the pore structure of ZIFs. Furthermore, it can allow us to reconsider incorporating some low-activity cations as high-activity molecules into ZIFs. In short, regarding the synthesis of ZIFs with MTV nodes, the key lies in balancing the inherent differences between ions, that is, controlling the mass transfer and coordination of metal ions.

CONCLUSION AND OUTLOOK

Overall, the functions of MTV-ZIFs, including the challenges and strategies in chemical synthesis, as well as how to interpret these effects, have been exhibited above. We believe that both materials and chemistry are moving towards diversification. MTV-ZIFs will bring reform to MOFs even in the field of materials science. Clearly, we can customize the capabilities of ZIFs through MTV linkers and nodes. Finally, some matters based on and extending beyond the MTV building units are listed.

Based on the reported and mentioned works, MTV-ZIFs can break through their own structural and thermodynamic limits theoretically in two aspects: (1) For the invisible matter, there is “new chemistry” hidden behind MTV-ZIFs which is worth exploring^[33]. The key here is to regulate the entropy and degrees of freedom in reticular materials. The series works by Andrew L. Goodwin, which focus on designing order in disorder, are inspiring and enlightening for the MTV-ZIFs^[202-204]. By utilizing the unconventional degrees of freedom rather than the sequence of building units, we may detect unexpected MTV-ZIFs. However, the invisible effects caused by the unique sequences and their corresponding interactions need advanced characterization methods to clarify the subtle molecular, atomic, and even electronic configuration differences^[51,205]. Even the relationship between the structures and properties relative to their applications is achievable through detailed analysis; (2) For visible effects, we can construct MTV phase diagrams (such as Figure 5) to help understand their potential in applications and the trade-off between MTV components^[155].

Many related studies on MTV-ZIFs lack above-mentioned systematic work.

In addition, beyond the materials itself, the chemical synthesis in MTV-ZIFs should be focused on when we persistently pursue the performance. (1) How to precisely control the crystallization and internal structure of them is the primary focus of attention. As we mentioned before, a MTV-ZIF has been obtained through mechanochemical synthesis, resulting in a powder rather than a single crystal^[154]. Although powder X-ray diffraction can also analyze the structure, it is impossible to have a clear and precise examination of the specific order and coordination structure. The hydrothermal method, however, is more suitable for growing MTV-ZIFs single crystal, despite being time-consuming^[58]; (2) The green chemical synthesis is equally crucial. Using green solvents such as water or in solvent-free conditions is a precondition for future applications instead of using N,N-dimethylformamide or similar solvents^[206]. For example, our FHML team has reported an *in-situ* thermal solvent-free synthesis to increase the yield of ZIFs to 95% and achieve linker recovery with one step of heating^[207,208]; (3) Recently, MOF assistants based on artificial intelligence (ChatGPT) have been reported to guide not only the discovery and prediction of the synthesis of new MOF^[209,210], but also the details of experiments such as the crystallization^[211]. Undoubtedly, the artificial-intelligent model trained by vast existing experiments will provide us with innovative ideas and solutions for the accurate, efficient, and green synthesis of MTV-ZIF.

As a new concept, although MTV-ZIF is full of unknowns and challenges, there are also opportunities and potential for new materials and chemistry. We hope that the theoretical discussions on the reticular chemistry of MTV-ZIF could provide inspiration for the future innovation of MOFs. We believe that a bright future for MTV-ZIFs and MTV materials will certainly be achieved by the continued exploration of the above challenge.

DECLARATIONS

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Authors' contributions

Supervision and preparation: Kou, Z.

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Methodology and writing original draft: Zhang, X.

Visualization: Zhang, X.; Li, X.

Validation: Qiao, A.; Verpoort, F.; Kou, Z.

Writing - review and editing: Zhang, X.; Li, X.; Wang, Z.; Chaemchuen, S.; Koo-amornpattana, W.; Qiao, A.; Bu, T.; Verpoort, F.; Wang, J.; Mu, S.; Kou, Z.

Availability of data and materials

Not applicable.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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