Molecularly Imprinted Porous Aromatic Frameworks for Molecular Recognition

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ABSTRACT: Porous aromatic frameworks (PAFs) are an important class of porous materials that are well-known for their ultralarge surface areas and superb stabilities. Basically, PAF solids are constructed from periodically arranged phenyl fragments connected via C–C bonds (generally), which provide vast accessible surfaces that can be modified with functional groups and intrinsic pathways for rapid mass transfer. Molecular imprinting technology (MIT) is an effective method for producing binding sites with a specific geometry and size that complement a template object. This review focuses on the integration of MIT into PAF structures via state-of-the-art coupling chemistry to expand the application of porous materials in the fields of metal ion extraction (including the nuclear element uranium) and selective catalysis. Additionally, a concise outlook on the rational construction of molecularly imprinted porous aromatic frameworks is discussed in terms of developing next-generation porous materials for broader applications.

INTRODUCTION

In biological and chemical processes, molecular recognition is a crucial step that governs the capabilities of enzymes and receptors in biological functions.1 In principle, the subtle structure (including a tailored configuration and well-designed functional groups) forms specific interactions with the target substrate.2 Because of this precise combination, organisms achieve efficient catalysis or response processes. As a consequence, the design and manufacture of enzyme and receptor mimics have long been pursued to efficiently identify and convert substrate molecules for chemical production, drug testing, artificial organs, and other applications.

Inspired by nature, a variety of synthetic systems have been developed for molecular recognition including supramolecular amphiphiles, cavity inclusion, and dynamic combinatorial/metallo-capsule/polymer receptors, etc.3−5 Significantly, molecularly imprinted technology is considered to be an effective and efficient approach for realizing the molecular recognition abilities.6 Generally, this technology is achieved through the following steps (Figure 1): (i) The template (ion, molecule, macromolecular assembly, and microorganisms) and functional groups form an imprinted complex by a self-assembly process involving multiple interactions such as van der Waals forces, hydrogen bonding, π–π interactions, ionic interactions, and coordination bonds. (ii) The imprinted complexes are incorporated into a bulk polymer through cross-linking agents, which facilitates the fixing of the position of the respective group. (iii) After the removal of the template, the final structure contains cavities that are capable of recognizing and rebinding the target objects and their analogues.7,8 Correspondingly, the generated molecularly imprinted polymers (MIPs) exhibit several leading edges, including high physical stability, specific recognition, a predictable structure, and universal application.9,10 Thus, molecularly imprinted technology has attracted widespread attention for applications such as...
chromatographic separations, artificial antibodies, sensing, artificial immunosays, drug delivery, and catalysis.11 Despite the tremendous success in molecular recognition, many problems with respect to its performance restrict its wide utilization.8−10 (1) The microrheology of the polymer distorts the spatial position of the functional groups, which then lose their selective capability to recognize specific template molecules. (2) Due to the dense structure of the polymer originating from the flexible skeleton, few imprinted sites are exposed on the particle surfaces, and a large number of imprinted sites are entrapped in the interior of the grains, thus greatly reducing the utilization of the imprinted sites. (3) Target objects with a relatively large diameter cannot be effectively transported through the channels. Although MIPs reveal significant characteristics for diverse applications, they suffer from some burning issues, including template leakage, a low binding capacity, and a slow diffusion velocity.

Porous materials with nanometer-size pore cavities are regarded to be a hot research topic in the chemistry and material science fields.12,13 This Outlook aims to reveal the unique advantages of porous materials for the application in the molecular recognition field. Zeolites were the first well-studied member and opened the door for the systematic investigation of composition, structure, properties, and functions.14 The excellent performances of zeolites in gas adsorption and separation and in catalysts15 have motivated the rapid development of other porous solids including aluminophosphates (AlPOs), mesoporous materials (OMMS), and metal−organic frameworks (MOFs).16−18 These porous materials with open architectures have numerous accessible surfaces and large storage spaces, enabling full host−guest interactions and rapid diffusion velocities. Based on these unique physical and chemical characteristics, they not only exhibit excellent properties in gas separations, molecular storage, and catalysis but also can be used in an extensive range of applications, such as monitoring, drug release, photoelectricity, etc.19−23 For instance, a copper MOF composed of 3,6-di(pyridin-4-yl)-1,2,4,5-tetrazine (dptz) as the ligand shows a negative solvatochromic behavior with a blue-shift of the absorption band with the increase of solvent polarity (Figure 2).24,25

Figure 1. Five main types of molecular imprinting: (i) noncovalent, (ii) electrostatic or ionic, (iii) covalent, (iv) semicovalent, and (v) metal coordination. An imprinted complex composed of the target object and ligands with functional groups is formed through several binding patterns: (I) hydrogen bonding, van der Waals, and π−π interactions; (II) electrostatic or ionic interactions; (III) a covalent bond; (IV) a covalent bond with a spacer; and/or (V) ligand−metal coordination. The ligand Y contains a reactive group for the cross-linking reaction. Then, the imprinted complex with the linker molecules is copolymerized to form the polymer matrix (gray). After removing the template object, an imprinted site is left behind with functional groups fixed on the polymer walls. Finally, the imprinted site with the tailored structure and well-designed functional groups rebinds the target objects. Reprinted with permission from ref 10. Copyright 2014, Royal Society of Chemistry.

Figure 2. UV−vis spectra and photograph of an MOF-based sensor containing solvent molecules. Reprinted with permission from ref 24. Copyright 2011 American Chemical Society.

Porous materials with diversified structural units can be endowed with talented architectures (including specific groups and suitable pore/window sizes), which direct the unique roles for guest molecules via several types of interactions: (i) van der Waals interactions with the porous channels, (ii) coordination bonds with metal centers, (iii) hydrogen bonding interactions with the framework surface, and (iv) π−π interactions with phenyl fragments.26 Two remarkable examples demonstrate the conventional strategies for achieving specific objectives in porous materials (Figure 3). (1) The ultra-micropores in zeolites allow us to rationally adjust to arrange multiple small gas molecules exclusively by size, which can be used to achieve
gas separation into high purity grades for practical industrial applications.27,28 (2) MOF samples are gifted with open metal sites that can strongly interact with guest molecules such as hydrogen and acetylene.29−33 Therefore, they could potentially be capable of separating gas molecules and sensing neutral and ionic species. However, these materials have many drawbacks, such as low selectivity, complicated synthesis procedures, and limited universality.

Inspired by nature, the specific interaction for a target object is realized through the assembly and arrangement of various functional groups in a confined cavity. Scientists aim to mimic this mechanism by decorating the porous scaffold with the functional groups using two strategies—pre- and postsynthesis modification (Figure 4).34−37 Presynthesis modification uses a building block with functional groups; after the cross-linking process, the functional groups are introduced into the porous network. Nevertheless, functional groups sometimes interfere with the reactivity and reduce the degree of the polymerization reaction to some extent. Furthermore, some prereactions (for instance, acid−base reactions and metal−ligand coordination) between the different functional groups lead to the deficiency of their activity.38−41 In addition, the cooperative effect of multiple functional groups is difficult to achieve when they are randomly distributed in the porous matrix. In postsynthesis modification, the porous framework is constructed first, and then, the architecture is decorated with functional groups.52−54 Some problems are inevitably encountered in this method: (1) The diffusion of functional groups through a porous solid results in a concentration gradient, and the amount of functional groups is high on the outside of the particle and low inside the particle. (2) The functional groups are randomly distributed, making it difficult to achieve cooperative effects. These problems make it an extravagant hope to fix functional groups in an angled and related pattern for molecular recognition.

As previously mentioned, MIT technology could be used to integrate various functional groups into tailor-made binding sites to complement the geometrical shape and chemical composition of target molecules. However, a molecularly imprinted complex consisting of multiple functional groups and a template object has an asymmetrical structure that does not match the fragment of the porous framework, leading to the distortion of the porous skeleton. After ditching the template molecules, the dynamic skeleton in classical materials (MOFs, for instance) undergoes a self-assembly process. Subsequently, the relative positions of the functional groups in the imprinted sites change, resulting in the deactivation of the imprinted pattern.

Recently, porous organic materials have attracted much attention because they combine the advantages of polymer materials and inorganic porous materials. They have several unique advantages, such as a low density, a large specific surface area, structural diversity, and tailorability. Their geometry can be predefined by the rational design of the size, structure, and connectivity of the building blocks. This control enables the fine regulation of the chemical and physical properties of the porous skeleton, resulting in an ideal environment for gas adsorption/separation and molecular storage, and the accessible and definable surface argues in favor of catalysis and sensing applications.55−60

In these cases, constructing diverse structures becomes the main concern for the state-of-the-art applications. With the help of the different preparation methods, porous organic materials connected by covalent bonds can be classified as hyper-cross-linked polymers (HCPs),61,62 polymers of intrinsic microporosity (PIMs),63,64 covalent organic frameworks (COFs),65−67 conjugated microporous polymers (CMPs),68,69 covalent triazine frameworks (CTFs),70 porous aromatic frameworks (PAFs),71 covalent organic polymers,72 and porous polymers,73−75 etc.,76−83 in a timed sequence (Figure 5). These materials have various structural characteristics: (1) HCPs are obtained by pillaring solvent-swelled polymers, generally through Friedel−Crafts alkylation, to expand the dense structure of flexible polymers. The pore cavities of PIMs with a 1D polymer skeleton are generated by distorting rigid fragments. COFs are prepared via the reversible cross-linking of polyhedral monomers to form a thermody-
Monomers are usually composed of phenyl fragment-based structural composition, pore size, and channel environments. The tetrahedral building block tetrakis(4-bromophenyl)methane was cross-linked to itself via a C–C Ullmann coupling reaction to obtain PAF-1 with a specific surface area of 5600 m² g⁻¹ based on the Brunauer–Emmett–Teller (BET) model (Figure 6). PAF-1 exhibits excellent thermal stability (>520 °C in air) and solvent stability in moist/acidic/basic environments. Due to the ultralarge surface area, it demonstrates high uptake capacities for hydrogen (10.7 wt % at 77 K, 48 bar) and carbon dioxide (1300 mg g⁻¹ at 298 K, 40 bar).

Based on this solid foundation, the synthesis of PAF materials has been continually developing by adjusting the structural composition, pore size, and channel environments. From a synthesis perspective, the building monomers are usually composed of phenyl fragment-based building units with highly rigid and symmetrical geometries, including linear, trigonal, tetragonal, and tetrahedral structures. Regarding the polymerization reactions, a diversity of linking approaches, such as Yamamoto-type Ullmann, Suzuki, Sonogashira–Hagihara, and Heck coupling reactions, are effective for the construction of PAF networks. The wide availability of monomers together with the diversity of coupling modes provides convenient conditions for the design and synthesis of various PAF solids.

PAF materials have inherent advantages for combining them with MIT including the following: (1) The large surface area provides numerous positions for imprinted sites. (2) The hierarchical pore structure is conducive to mass transfer. (3) The stable skeleton can withstand the process of template removal and practical utilization. Compared with crystalline materials (MOFs and COFs) with a dynamic skeleton, the irreversible covalent linking pattern of PAFs provides inherent structure for the safe existence of imprinted sites. When the building monomers polymerize around the imprinted complex, the expanded network provides a large number of channels for the transfer of guest molecules toward imprinted sites. The rigid skeleton overcomes the microrheology of the flexible polymer, which is conducive to maintaining the structure of the imprinted sites.

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Incorporating single-function imprinted sites for the removal of heavy metals

Lead is an essential substance in the battery and paint industries. With the rapid development of industrialization, the global demand for refined lead increases sharply by 1.29 million tons from 2012 to 2016. According to statistics, only 4.9% of total consumption is effectively recycled for commercial usage every year. Correspondingly, a large number of lead ions are discharged into the natural environment, causing serious environmental pollution and lead poisoning. Due to the great pressure of environmental and economic problems, the establishment of an efficient lead recycling system is urgently needed. The authors mixed methacrylic acid (MAA), 4-vinylpyridine (4-VP), and Pb(NO₃)₂ together to obtain a Pb-imprinted complex (Figure 7a). The pyridyl group acts as a monodentate ligand to bind to a Pb²⁺ ion through a coordination bond. The deprotonated carboxyl group also coordinates to the Pb²⁺ ion, which leads to the cooperative effects of the pyridine ring and carboxyl group in the Pb-imprinted complex. This Pb-imprinted complex contains vinyl groups at the end, which can undergo a Heck coupling reaction to realize a PAF skeleton extending around the imprinted complex. The Pb-imprinted complex was used as a building unit, replacing the original monomer (divinylbenzene) at molar ratios varied from 0%, 10%, 20%, 30%, to 40% to obtain PAF-10a, PAF-10b, PAF-10c, PAF-10d, and PAF-10e, respectively (Figure 1b). After removing the Pb²⁺ centers from the PAF structure to obtain MIPAF, the carboxylic acid and pyridine groups are fixed in the PAF framework by rigid structural segments.
These MIPAF materials maintain the large porosity of the parent PAF sample (613 m² g⁻¹) with surface areas ranging from 87 to 427 m² g⁻¹. The utilization of the imprinted sites varies from 96% for PAF-10b to 57% for PAF-10e. The largest sorption uptake of 90.36 mg g⁻¹ is achieved with MIPAF-10d (utilization of imprinted sites ∼89%). In contrast, the traditional polymer-based MIP powder with the same amount of imprinted complexes has a specific surface area of 37 m² g⁻¹. Its capacity for Pb²⁺ ions is only 5.5 mg g⁻¹, corresponding to the utilization of 14% of the imprinted sites. The capacity of MIPAF-10d is ca. 15 times higher than that of MIP. All these results demonstrate that the large accessible surface in the MIPAF architecture provides a larger number of imprinted sites for Pb²⁺ ion binding (Figure 7). The selectivity coefficient of MIPAF-10d for Pb²⁺ ions exceeds 1.5 × 10² (Pb²⁺/Cu²⁺), which is more than 3 times that of MIP (Figure 7c). This phenomenon is attributed to the fact that the rigid scaffolds of PAFs effectively keep the relative positions of the functional groups as they were in the imprinted complex, and the two functional groups cooperatively and selectively bind lead ions again.

Because of the pure organic composition of the PAF network, the MIPAF-10d powder can be charged into a CH₂Cl₂ solution of poly(methyl methacrylate) (PMMA) to prepare a composite pipe using MIPAF-10d as an interior coating. This MIPAF-10d-doped composite pipe selectively adsorbs emitted Pb²⁺ ions with high efficiency (>99.94%). The Pb²⁺ ion concentration in the outflow decreases from an original 20 ppm to ∼0.008 ppm, satisfying the standard issued by the U.S. Environmental Protection Agency (0.015 ppm).⁹⁰⁻⁹¹

Figure 7. (a) Structural units for MIPAF materials 10b–10e. (b) Ion adsorption of PAF-10d against various interfering ions. (c) Comparison of selectivity and saturation uptake for Pb²⁺ ion. Reprinted with permission from ref 106. Copyright 2018, Royal Society of Chemistry.

### Incorporating Dual-Function Imprinted Sites for Tandem Functions

Natural enzymes have specific recognition and catalytic properties toward substrate molecules. A variety of enzymes cooperate to achieve efficient metabolism in the human body. Many scientists hope to mimic these interrelated and collaborative catalytic processes to realize an efficient production technology. However, different functional groups might interact with each other to form acid–base or coordination adducts, rendering them ineffective and preventing them from occupying specific fixed positions. Using the MIT method to preassemble the functional groups needed for various target capabilities, different types of imprinted sites may be embedded in the large accessible surface of the PAF platform to perform tandem or cooperative behaviors that originate from the rigid skeleton and large accessible surface.

Organophosphorus compounds are a kind of nerve agent that can seriously damage the nervous system of the human body. In order to degrade this poison, the authors designed a hydrolysis site for organophosphorus that simulates the binuclear zinc centers in the natural enzyme.¹⁰⁷ A series of MIPAFs were synthesized via a Heck coupling reaction with 10%, 20%, 30%, and 40% of hydrolysis site (molar ratio of hydrolysis complex to p-divinylbenzene) and are denoted as MIPAF-1, MIPAF-2, MIPAF-3, and MIPAF-4 (Figure 8a). The BET surface areas of MIPAF-1, MIPAF-2, MIPAF-3, and MIPAF-4 are 503, 390, 275, and 160 m² g⁻¹, respectively. For MIPAF-1 through MIPAF-3, the hydrolysis speed gradually increases, and then, it decreases with the increased content of hydrolysis sites in MIPAF-4. The hydrolysis rate of MIPAF-3 is 9.5 × 10⁻⁶ m min⁻¹, which is ~26 times higher than that of the traditional MIP with a nonporous structure.
Using the hydrolysate (p-nitrophenol) as a template, a product adsorption complex is obtained by preassembly with zinc dimethacrylate. Similarly, MIPAF-5, MIPAF-6, MIPAF-7, and MIPAF-8 possess 10%, 20%, 30%, and 40%, respectively, of product adsorption complex. The BET surface areas of MIPAF-5, MIPAF-6, MIPAF-7, and MIPAF-8 are 507, 400, 283, and 191 m² g⁻¹, respectively. Regarding the capacity for p-nitrophenol, MIPAF-7 exhibits the largest uptake of 32.06 μg mg⁻¹ among other MIPAF samples, which is a 4.6-fold increase relative to that of the traditional polymer-based MIP (5.7 μg mg⁻¹). Both the substrate hydrolysis and product adsorption experiments make evident that PAF scaffolds will open up the interior spaces and enable the high utilization of imprinted sites.

As previously reported, the in situ separation of products from catalytic sites can effectively accelerate the catalytic process. In order to increase the catalytic rate, 30% substrate hydrolysis sites and 5% product adsorption sites were incorporated into a PAF scaffold to give MIPAF-9. The MIPAF-9 powder hydrolyzes the organophosphorus molecules and transports the hydrolysate (p-nitrophenol) from the catalytic site. The resulting catalytic system converts 17% of the paraoxon-ethyl in 2.5 h, which surpasses 14 times the rate of natural organophosphorus hydrolase (Flavobacterium sp.)
strain ATCC 27551). It is worth mentioning that, with the stable PAF as a scaffold, the MIPAF-9 sample exhibits excellent stability at high temperatures; in the presence of heavy metals, acids/bases, and organic solvents; and with cycling (Figure 8). The integration of multiple functional units in the porous architecture facilitates the realization of efficient catalysis. In addition, the porosity and stability of the PAF platform provide more possibilities for practical applications.

**POSTMODIFICATION OF RECOGNITION SITES FOR SELECTIVE HALOGENATION**

As previously observed, the topological structure of an imprinted complex is quite different from the segment of the porous framework. As imprinted sites are introduced into the architecture, the skeletal fragments are distorted, and the pore channels are blocked, which damages the integrity of the framework structure. Therefore, the specific surface area of PAF materials gradually decreases with increasing content of imprinted sites. After exceeding a certain upper limit (molar ratio of 40–50%) of the imprinted complex loading, the yields of the resulting solids decrease sharply. Maintaining the integrity of the porous architecture and increasing the amount of imprinted sites for high-performance molecular recognition is a goal that is currently being pursued.

Halide molecules are important ingredients in the fields of pharmaceuticals and chemical production. However, phenyl rings have multiple reactive sites, which introduces additional challenges to selectively synthesizing phenyl halides. To increase the effective payload of imprinted sites, the authors designed a fully fluorinated PAF solid, denoted as PAF-63. PAF-63 exhibits unique advantages (Figure 9), including the following: (i) The C–F bond has a higher energy than the C–H bond, helping to maintain the structural integrity and avoid the side reactions, and (ii) the hierarchical porosity facilitates the postsynthesis reactions for the recognition units.

The PAF architecture was subsequently decorated with three CD molecules (α-CD, β-CD, and γ-CD) with internal diameters varying from 0.57 to 0.95 nm. The CD-modified PAF frameworks have hydrophobic cavities that can incorporate aromatic substrates due to the CD internal chamber. Under the protection of the hydrophobic cavities, the meta and ortho positions are embedded in the CD-PAF materials, and only the para position is exposed for halogenation reactions (Figure 9f). This study illustrates the postsynthesis modification method for obtaining a large number of imprinted sites while also maintaining the integrity of the PAF framework. This strategy prevents the collapse of the framework and the blockage of the mass transfer channels, greatly improving the molecular recognition performance of these materials.

**TACKLING AN INTERNATIONAL PROBLEM: URANIUM EXTRACTION**

Currently, nuclear power provides 13% of the world’s electrical energy; and the International Atomic Energy Agency (IAEA) predicts that it will supply more than half of the electricity consumed globally in the next few decades. Uranium is an indispensable raw material in the nuclear energy industry.
However, the uranium reserves that are easy to mine on land are about to be exhausted in the coming decades. Seawater contains the largest amount of uranium in the world and can help meet energy needs and ensure developments in the long run. Nevertheless, two major problems, i.e., the ultralow concentration (∼3.3 ppb) and presence of vast competing...
PAF materials featured by large specific surface areas and high stabilities show great potential in the enrichment of radioactive ingredients. Using PAF-1 (BET surface area ~5600 m$^2$ g$^{-1}$) as the scaffold, Prof. Ma decorated the PAF-1 framework with amidoxime groups to obtain PAF-1-CH$_2$AO, and Prof. Dai loaded polycrylonitrile into its channels to prepare amidoxime-PAF-1. Both materials reveal an excellent capability in terms of adsorption capacities and rates, but the selectivity still needs to be improved.

According to the specific coordination mode of uranyl ions, the authors synthesized a UO$_2$$^{2+}$-imprinted complex consisting of salicylaldoxime, MAA, 4-VP, and UO$_2$(NO$_3$)$_2$ via free assembly (Figure 10a). Through the Heck coupling reaction, the UO$_2$$^{2+}$-imprinted complex together with 1,3,5-tris(4-bromophenyl)benzene and p-divinylbenzene was used to construct a series of PAF networks (Figure 10b,c). After removing template UO$_2$$^{2+}$ ions, the carbosyl and pyridine groups in the PAF structure are fixed on the PAF structure through covalent bonds, whereas the salicylaldoxime units maintain their spatial positions through hydrogen bonding and π-π interactions.

The combination of MIT technology and porous materials effectively enables the highly efficient and selective adsorption of uranyl ions. MIPAF structures provide numerous accessible imprinted sites with a nearly 4-fold increase in the capacity relative to that of conventional MIPs. To our knowledge, the MIPAF-11c material has the highest reported selectivity (selective coefficient of greater than 746) of all uranium adsorbents (Figure 10). In addition, composite devices, including fibers, films, and coatings, can be fabricated by a flexible operation using the UO$_2$$^{2+}$-imprinted PAF powder, enabling facile and convenient industrial utilization.

To improve the adsorption capacity while maintaining the high selectivity, the authors designed an imprinted complex (uranyl-specific bis-salicylaldoxime entity) by the ion coordination template strategy. The bis-salicylaldoxime entity is tethered on the PAF skeleton through hydrogen bonding (Figure 11a). Regarding its spatial structure, the configuration of the uranyl-specific bis-salicylaldoxime entity is consistent with the fragment of PAF-1, and the imprinted sites maintain their structural integrity via π-π interactions (Figure 11b). Because the imprinted sites preserve the architecture and direction for uranium rebinding, the resulting adsorbent exhibits ultrahigh selectivity for uranium ions with a selectivity coefficient of greater than 113 (uranium/vanadium) and an outstanding capture capacity of 5.79 mg g$^{-1}$ from real seawater in 56 days. Professor Daniel T. Sun wrote a critical article, stating that the porous aromatic framework material with uranyl-specific bis-salicylaldoxime entities has fascinating properties and is expected to solve the world problem of uranium extraction from seawater.

Having established a porous adsorbent with both high selectivity and capacity, conductive chains (phenylacetylene, PPA) were incorporated into the channels of porous adsorbents into PPA@MISS-PAF-1. Under the asymmetrical alternating current electrochemical (AACE) method, the expended electrical field covers the micrometer-sized particles, which guide the migration and enrichment of uranyl ions (see Figure 12). Based on the merits of both porous

![Figure 12. Chemical structure for phenylacetylene-doped MISS-PAF-1 and PPA@MISS-PAF-1, and physical processes occur under AACE extraction. Reprinted with permission from ref 114. Copyright 2020, Elsevier.](https://dx.doi.org/10.1021/acscentsci.0c00311)

**SUMMARY AND PERSPECTIVE**

This Outlook illustrates the important progress in the construction of molecularly imprinted PAFs. MIPAF solids are breaking new ground by combining MIT technology with the construction of PAF networks to attain the large surface area, hierarchical porosity, and rigid skeleton of PAFs and the specific molecular recognition of MIT. Significant progress in porous ion adsorbents and porous artificial enzymes has been achieved with MIPAFs, which exhibit outstanding performances in terms of capacity and selectivity. The materials overcome the bottlenecks and disadvantages of traditional molecularly imprinted polymers, such as the difficult elution of template molecules, low binding ability, slow mass transfer, inability to imprint macromolecules, etc. As compared with other strategies for molecular recognition, the MIPAFs render some leading edges such as strong recognition capability, high physical/chemical stability, excellent usage for functional sites, rapid mass transfer, and universal application. Consequently, they might expand the applications of porous organic materials into artificial antibodies, purification and separation, drug delivery, chemo-/biosensing, catalysis, and other fields.

Future work should focus on the following: (1) changing the hydrophilicity, hydrophobicity, and polarity of the pore surfaces of PAF materials to simulate the catalytic environments of biomimetic enzymes; (2) tuning pore structures to regulate the speed for mass transfer; (3) rationally assembling multiple imprinted sites to reasonably combine multiple functions; (4) utilizing conjugated building units with electrical or photophysical properties for chemo-/biosensing fields; and (5) imprinting macromolecules, such as biological enzymes, DNA, proteins, etc., for their monitoring and separation.

**Safety Statement.** No unexpected or unusually high safety hazards were encountered.
These materials overcome the bottlenecks and disadvantages of traditional molecularly imprinted polymers, such as the difficult elution of template molecules, low binding ability, slow mass transfer, inability to imprint macromolecules, etc.

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**Notes**
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