



Porous ionic polymers: Design, synthesis, and applications

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ABSTRACT

Porous ionic polymers (PiPs) are newly emerging organic porous polymers. In sharp contrast to charge-neutral porous polymers, ionic moieties are either incorporated into the polymer backbone, or are covalently attached to a polymeric framework. Therefore, their physicochemical properties, functional groups, and active sites can be easily modified through screening of building blocks and ionic tectons. Meanwhile, their surface area, pore size, and pore volume can also be tuned by counterions exchange. Intrinsic functionalization further broadens the application range of the PiPs. This review will describe the recent advancements with regard to PiPs, and their development in the area of gas adsorption, catalysis, antibacterial applications and water purification.

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Abbreviations: AA, acrylic acid; ABN, sanionic microporous polymer networks; AN, acrylonitrile; BF, Blm5,6-bis(4-formylbenzyl)-1,3-dimethyl-benzimidazolium bromide; BIPY, bipyridinium; BPIB1, 3-bis(4-bromophenyl)imidazolium bromide; cCTF, charged covalent triazine framework; CCU, carbon capture and utilization; CMP, sconjugated microporous polymers; COF, scovalent organic frameworks; CPN, cationic microporous polymer network; CPP, scharged porous polymers; CT, charge-transfer; CTF, scovalent triazine frameworks; DMF, *N,N*-dimethylformamide; DMSO, dimethyl sulfoxide; EB, ethidium bromide; HCPs, hypercrosslinked polymer; HER, hydrogen evolution reaction; HIP, hypercrosslinked ionic polymers; i-COP, sionic covalent organic polymers; ICOF, ionic covalent organic framework; iCONs, ionic covalent organic nanosheets; IL, ionic liquid; IMPP, ionic-metal porous polymer; LTIPB, lithium tetrakis(4-iodophenyl)borate; MO, methyl orange; N-CMP, naphthyl containing conjugated microporous polymer; NIPAM, *N*-isopropylacrylamide; OER, oxygen evolution reaction; PAF, sporous aromatic frameworks; PBC, polyborane carbazole; PC-COF, polycationic covalent organic framework; PCIF, porous cationic frameworks; PCPs, porous cationic polymers; PCu-NHC, Cu(II)-coordinated polyN-heterocyclic carbenes; PhIm, 2-phenylimidazoline; PILs, poly(ionic liquid)s; PlmCl, polyimidazolium chloride; PIMs, polymers of intrinsic microporosity; PIONs, nanoporous ionic organic networks; PIONs, nanoporous ionic organic networks; PiPs, porous ionic polymers; PNHC, polyN-heterocyclic carbenes; POPs, porous organic polymers; POSS, polyhedral oligomeric silsesquioxane; PPN, porous polymer networks; PSF, polysulfone; PXRD, powder X-ray diffraction; PyTTA, 4,4',4''-(pyrene-1,3,6,8-tetrayl) tetraaniline; S_{BET} , Brunauer-Emmett-Teller specific surface area; TATA, thiophene-functionalized triazatriangulenium cations; TBBT, 2,4,6-tribromo-benzene-1,3,5-triol; TBPE, tetrakis-(4-bromophenyl) ethylene; TEPM, tetrahedral tetrakis(4-ethynylphenyl) methane; TFP, 1,3,5-triformylphloroglucinol; TOF, turnover frequency; TON, turnover number; TTSBI, 5,5,6,6-tetrahydroxy-3,3,3,3-tetramethyl-1,1'-spirobisindane.

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1. Introduction

Porous materials attracted significant attention, regardless of whether they occur naturally or are artificially designed [1–9]. Due to their distinct advantages, such as fascinating architectures, high surface areas, and large pore volumes, porous materials have become increasingly important for various applications ranging from gas storage and separation to catalysis, to water purification, to energy conversion and storage [10–20]. Among porous materials, organic porous polymers are unique because they possess the properties of both porous materials and of polymers. These porous polymers include covalent organic frameworks (COFs) [21–24], conjugated microporous polymers (CMPs) [25–28], hypercrosslinked polymers (HCPs) [29–31], polymers of intrinsic microporosity (PIMs) [32–35], covalent triazine frameworks (CTFs) [36–39], porous aromatic frameworks (PAFs) [40–43], porous polymer networks (PPN) [44,45], and others [46,47]. One unique characteristic of porous polymers is the wide range of chemical reactions and synthetic methods that are available to prepare them. Another characteristic is a considerable flexibility in monomer design at the atomistic level. Rigid building blocks with diverse structures and compositions have already been synthesized. These two advantages facilitate the optimization of the pore sizes and maximization of the surface area of porous polymers. More importantly, the rational design of porous polymers with diverse structures and functions renders them suitable for specific applications, such as gas storage, separation, water treatment, as heterogeneous catalysts, and in the photovoltaic and luminescent fields [48–58].

Due to the remarkable advantages of porous polymers, “function-led design” or “target-motivated” approaches can be used to prepare various functional porous polymers [59–67]. The above-mentioned functional porous polymers are all charge-neutral. Bringing ionic moieties into the porous polymer backbone will no doubt diversify the types of porous polymers and further broaden the application range of porous polymers [68]. PiPs are a new class of porous polymers that permit the linking of building blocks with both ionic bonds and chemical bonds. Thus, the ionic moieties are not only attached to the pore wall but are embedded in the pore wall. More importantly, their physicochemical properties, functional groups, and active sites can be easily tuned by counterion exchange. Due to the high charge density, high surface area, and well-defined porosity, as well as the synthetic diversity, PiPs will become a rising star in the area of material science.

The topic of PiPs is steadily broadening as an increasing number of diverse applications are demonstrated. Here, we aim to assemble a review of the significant scientific progress in the synthesis and applications of PiPs. We will discuss the advances in molecular design, synthesis, and the potential applications of PiPs in the area of gas storage and separation, in catalysis and for antibacterial applications, as well as in the water purification process.

2. Design and synthesis of PiPs

In general, a wide range of chemical reactions and synthetic methods, such as Schiff-base reaction [69,70], dehydration of boronic acid [71], Suzuki cross-coupling reaction [72], Yamamoto

reaction [73,74], Sonogashira-Hagihara reaction [75,76], Friedel-Crafts reaction [77,78], C–O cross-coupling reactions [79] and others [80–82] are available for the synthesis of PiPs. To date, various ions, such as quaternary ammonium cations [83], borate anions [84], quaternary phosphonium cations [85] and other cations have been involved in the synthesis of PiPs. Consequently, the Brunauer-Emmett-Teller specific surface area (S_{BET}) can be tuned by the molecular design such as neutral porous polymers. Furthermore, the pore diameters can be easily adjusted at an angstrom scale just through ion exchange. However, due to the charge interaction, intermolecular packing easily occurs, which decreases the S_{BET} and the pore volume. Therefore, functional monomer with rigidity and contortion are prerequisites for maintaining the interconnected porosity of PiPs. Based on recent publications, mainly four strategies that were developed for the synthesis of PiPs will be discussed in detail in the following section.

2.1. Direct polymerization of ionic building unit

The direct synthesis from ionic tectons is an efficient method for obtaining PiPs with high charge density. Typical monomers for the direct synthesis of PiPs are listed in Table 1. The free radical polymerization approach is the most commonly employed reaction. A series of quaternary phosphonium cations containing polymers were prepared based on this method. For example, Xiao et al. reported a hierarchical porous ionic organic polymer (with an S_{BET} of $758 \text{ m}^2 \text{ g}^{-1}$ and a total pore volume of $0.59 \text{ cm}^3 \text{ g}^{-1}$) prepared from vinyl-functionalized quaternary phosphonium cationic monomers [86]. They also prepared a hierarchically porous polymer (with an S_{BET} of $1123 \text{ m}^2 \text{ g}^{-1}$ and pore volume of $2.1 \text{ cm}^3 \text{ g}^{-1}$) from 5,5'-divinyl-2,2'-bipyridine via a free-radical polymerization reaction [87]. Based on the same method, Meng et al. prepared vinyl-functionalized quaternary phosphonium cation-based PiPs via a free-radical solvothermal polymerization reaction. The obtained PIP-Me-I, PIP-Et-Br, and PIP-Bn-Cl showed surface areas of 402, 625, and $758 \text{ m}^2 \text{ g}^{-1}$, respectively [88]. Recently, Ji and co-workers prepared a series of ionic-metal porous polymers (IMPP:DVB@ISA (Al(salen)) and DVB@ISZ (Zn(salen))) through the one-pot free-radical copolymerization of metallosalen complexes containing IL moieties and divinylbenzene (DVB) under solvothermal conditions [89]. The S_{BET} and the total pore volume of DVB@ISA were found to be $590 \text{ m}^2 \text{ g}^{-1}$ and $0.55 \text{ cm}^3 \text{ g}^{-1}$, respectively. In addition, Zhu et al. prepared quaternary pyridinium-type PAFs (PAF-50 with a surface area of $384 \text{ m}^2 \text{ g}^{-1}$ and pore sizes of 0.5 nm) through the condensation of 4-pyridinylboronic acid and cyanuric chloride [71]. Han et al. have reported porous organic polymers synthesized from triazatriangulenium cations (TAPOPs) via FeCl_3 -promoted oxidative polymerization [77]. The obtained TAPOP-1 had an S_{BET} of $940 \text{ m}^2 \text{ g}^{-1}$.

Charged covalent triazine framework (cCTF) was recently reported by Coskun et al. [90]. A cCTF with an S_{BET} of $1247 \text{ m}^2 \text{ g}^{-1}$ was synthesized from cyanophenyl substituted viologen dication via ionothermal trimerization reaction at 500°C . Similarly, Yoon et al. synthesized 1,3-bis(pyridyl) imidazolium-based CTFs (bpim-CTFs), which showed an S_{BET} of $1556 \text{ m}^2 \text{ g}^{-1}$ and a total pore volume of $0.75 \text{ cm}^3 \text{ g}^{-1}$, respectively [91].

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