Coordination Chemistry Reviews 261 (2014) 1–27

Contents lists available at ScienceDirect

Coordination Chemistry Reviews

journal homepage: www.elsevier.com/locate/ccr

Review

Metal-organic frameworks based upon non-zeotype 4-connected topology

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Abbreviations: ABTC, 3,3′,5,5′-azobenzenetetracarboxylic acid; ADA, adipic acid; ADB, azobenzen-4,4′-dicarboxylic acid; ADC, 1,3-adamantine-dicarboxylic acid; 5-AIP, 5-aminoisophthalate; ATZ, 5-amino-tetrazole; BD DC, 5,5′-(buta-diyne-1,4-diyl) diisophthalic acid; BPTC, biphenyl-2,5,2′,5′-tetracarboxylic acid; BPE, 1,2-bis(4-pyridyl)ethane; BPDC, biphenyl-4,4′-dicarboxylic acid; 2,4′-H_2BPDC, 2,4′-biphenyldicarboxylic acid; BPEA, biphenylethene-4,4′-dicarboxylic acid; BPG, bi(4-pyridyl) glycol; BPPD, 1,3-bis(4-pyridyl)-1,3-propane dionate; BPP, 1,3-bis(4-pyridyl)-propane; BPIPA, N,N′-bis-4-pyridyl-isophthalamide; 4,4′-BPY, 4,4′-bipyridine; BPS, 4,4′-dipyridyl sulfide; BDC, benzenedicarboxylate; BIX, 1,4-bis(imidazol-1-ylmethyl)benzene; BIIM-4, 1,1′-(1,4-butanediyl)bis(imidazole); BIMB, 4,4′-bis(imidazol-1-ylmethyl)biphenyl; BMB, (1,4-bis(2-methylbenzimi dazol-1-ylmethyl) benzene); BPMP, bis(4-pyridylmeth-yl)piperazine; BTEC, 1,2,4,5-benzenetetracarboxylic acid; BENZTB, *N,N,N′,N′*-benzidinetetrabenzoate; 4,4′-BPDZ, 4,4′-bipyridazine; BTMB, 4,4′-bis(1,2,4-triazol-1-ylmethyl)biphenyl; BIE, 2,2′-bis(1Himidazolyl)ether; BIIM-4, 1,1′-(1,4-butanedidyl)bis-(imidazole); BTZ, benzotriazolate; BZTPY, 1,2,4,5-tetra(4-pyridyl)benzene; BPYM, 5,5′-bipyrimidine; 1,4-BPEB, 1,4-bis[2-(4-pyridyl)ethenyl] benzene; CPT, 4-(4-carboxyphenyl)-1,2,4-triazole; CMB, 2-(carboxymethoxy)benzoic acid; CHDC, cis-/trans-1,4-cyclohexanedicarboxylic acid; DPT24, 3-(2-pyridyl)-5-(4-pyridyl)-1,2,4-triazolate; DABCO, diaza bicyclo-[2,2,2]octane; dadpm, 4,4′-diaminodiphenylmethane; DCMP, 2.5-dicarboxy-1-methylpyridinium; DDM, p,p′-diamino-diphenyl-methane; DEF, N,N-diethylformamide; DHTP, 2,5-dihydroxyterephthalate; DMA, dimethyl acetamide; DMF, N,N′-dimethylformamide; DMP, 2,6-dimethylphenol; DPA, 4,4′-dipyridylamine; DPE, 1,2-di(4-pyridyl)ethylene; EBTC, 1,1′-ethynebenzene-3,3′,5,5′-tetracarboxylate; EN, ethylenediamine; ET, 5-ethyltetrazole; 2-ETIM, 2-ethylimidazole; H_2DPA, diphenic acid; H_3IEDC, 2-ethyl-1H-imidazole-4,5-dicarboxylic acid; H_3PhIDC, 2-phenyl-1H-imidazole-4,5-dicarboxylic acid; HMPH, homophthalic acid; IDC, imidazole-4,5-dicarboxylic acid; IN, isonicotinic acid; IPT, 5-isopropyltetrazole; ISOP, benzene-1,3-dicerboxylic acid; MDPT 24, 3-(3-methyl-2-pyridyl)-5-(4-pyridyl)-1,2,4-triazolate; MTPO, 5-methyl-1,2,4-triazolo[1,5-a]pyrimidin-7(4H)-one; MT, 5-methyltetrazole; MBIX, 1,3-bis(imidazol-1-ylmethyl-)-benzene; NDC, 2,6-naphthalenedicarboxylic acid; ODPT, 4,4′-oxidiphthalate; OHIP, 5-hydroxyisophthalate; OQA, 4-oxo-1(4H)-quinolineacetic acid; PBT, 5′-(pyridin-2-yl)-2H,4′H-3,3′-bi(1,2,4-triazole); PN, 1,3-diaminopropane; PTZ, 5-(3-pyridyl)tetrazole; PMC, pyrimidine-5carboxylate; PPE, poly(1,4-phenylene ether); PYEB, bis(4-[2-(4-pyridyl)ethenyl] benzoic acid); PYBZ, 4-(pyridin-4-yl)benzoic acid; PMA, pyromellitic acid; PMPMD, N′-bis(4-pyridylmethyl)phenyldiimide; PMTZ, 5-(pyrimidyl)-tetrazolato; PHDA, phenylenediacetic acid; PHDP, 1,4-phenylenedipropionate; PDA, phenylenediacetic acid; PA, 4,4′-methylenebis[3-hydroxy-2-naphthalene carboxylic acid; PHT, phthalate; QUIN-6-c, quinoline-6-carboxylate; (S)-IMP, (S)-2-(1H-imidazole1-yl) propionate); SDBA, 4,4′-dicarboxybiphenyl-sulfone; TCPP, 4-(1H-1,3,7,8-tetraazacyclopenta[l]phenanthren-2-yl)phenol; TEA, triethanolamine; TMA, trimesate; TPOM, tetrakis(4-pyridyloxymethylene)methane; TPP, tetraphenylphosphonium; TPYP, meso-tetra(4-pyridyl)porphyrin; TZC, 5-carboxylatotetrazolato; TZDC, 1,2,3-triazole-4,5-dicarboxylate; TTG, *N,N′,N″*-1,3,5-triazine-2,4,6-triyltris-glycine.

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0010-8545/$ – see front matter
http://dx.doi.org/10.1016/j.ccr.2013.11.004

ARTICLE INFO

Article history:
Received 13 August 2013
Accepted 4 November 2013
Available online 14 November 2013

Keywords:
Metal-organic frameworks
Non-zeotype 4-connected net
Adsorption
Catalysts
Luminescence
Magnetism

ABSTRACT

In order to further design and successfully prepare for the functional metal-organic framework materials, it is essential to understand the fundamental correlations between the composition, physical properties and topology of the underlying nets. In this review, we focus on recent advances in metal-organic frameworks (MOFs) that possess more common non-zeotype 4-connected topological nets (such as **sql**, **kag**, **nbo**, **lvt**, **cds**, **qtz**, **dia**, **lon**, **pts**, etc.), and discuss the synthetic strategies of non-zeotype 4-connected MOFs and their related properties. In particular, we emphasize how to establish basic design principles and synthetic methodology to construct the same topological MOFs with different functions using specifically designed organic linkers.

1. Introduction

Metal-organic frameworks (MOFs) have recently emerged as a novel class of promising inorganic–organic hybrid materials for diverse structure-related applications [1–7]. Conceptually, MOFs are crystalline porous materials built from metal ions/polynuclear building units connected by polytopic organic linkers. Their exact structures and physical properties associated with their structural features can be well characterized by advanced diffraction techniques, spectroscopic methods and theoretical simulations [8–10]. To some extent, MOFs have higher a degree of designability and adjustability in their structures and functions, as well as acceptable thermal stability when compared with other porous solid materials such as zeolites. Insufficiency and disfigurement problems have been associated with zeolite porous materials in the chemical process. Whereas the MOFs as novel functional materials, especially for those possessing analogous zeolite 4-conncected topological net, have shown superior properties. Thus, the exploration of their performance for practical applications is attracting intense interest in the fields of chemistry, chemical engineering, materials science, and others [11]. In principle, diversified structures of MOFs are usually assembled by the metal ions (or transition-metal carboxylate clusters as secondary building units (SBUs)) and bridging organic tectonics utilizing different synthetic strategies [12–14]. However, designing a target structure with specific properties is always very difficult, in comparison with a typical organic reaction. Moreover, these complex structures are usually showing complicated structures that are linking with special properties. Therefore, it is very important to understand the fundamental correlations between their composition, physical properties and topology of the underlying nets.

In the assembly process of various MOFs, topological control has served as a facile and effective approach to guide the direct-synthesis of functional MOFs with desirable features [15]. That is, the topology analysis could simplify complicated structures to straightforward networks and help one to analyze the packing trends of diverse MOFs [16]. Furthermore, understanding the structural topology will help on the structural design of new functional MOFs [17]. IUPAC approved Structure Commission of the International Zeolite Association assigned three letter codes to each unique zeolite structure and published them in a web-based database, O'Keeffe, Yaghi and co-workers have also adopted three letter codes for each net in MOFs that corresponding to a similar system used for zeolites. Accompanied by the rapid development of MOFs, a variety of network topologies have been discovered, such as 3-, 4-, 6-, 8- and even higher-connected and mixed-connected networks [18,19]. Among them, nodes of 4-connectivity, possessing zeolite analog structures, are very common in crystal chemistry and various kinds of such net topologies have been realized by 3D coordination networks [20,21]. Significantly, such exceptional nets not only possess fascinating structures but also show excellent properties for potential applications in the area of gas storage, separation, luminescence and magnetism. Some early monographs [22], though, have briefly mentioned those different 4-connected topological types; they are very limited due to fewer available entries. Some other reviews focused on 4-connected zeotype MOFs have also been published [23,24]. In this context, a further summary for MOFs with non-zeotype 4-connected topologies, particularly for their design strategies, is vitally essential for an in-depth investigation of these structure-related materials. In this review, we do not intend to provide a comprehensive collection of literature, but to focus on the progress in the design, synthesis, and properties of such non-zeotype 4-connnected MOFs with selected representative examples from the literature, which, in some extent, may be applied as a feasible approach in preparing such crystalline materials in the future.

2. Building strategy of 4-connected nets

According to the principle of topology, many high dimensional MOFs can be simplified with diverse topological nets considering the single metal ions (or metal clusters) as "nodes" and organic ligands as "linkers" through coordination bonds. From the viewpoint of tectonics, the prerequisite constituents of constructing 4-connected nets are the organic linkers and metal ions possessing 4-connected nodes, or 2-conneted metal linker associating with 4-connected organic nodes as well as the metal ions as 4-connected nodes and organic ligands as 2-conneted linkers, respectively (Chart 1), which should be pre-designed using the organic ligands and metal ions in the assembly process of 4-connected MOFs. These factors will be summarized and discussed in the following.

2.1. The typical non-zeotype 4-connected nets

The assembly of a zeotype framework is based on the connectivity between tetrahedral nodes. Moreover, there are a number of non-zeotype 4-connected networks built from square nodes or tetrahedral nodes or mixed square and tetrahedral nodes (Chart 2). Over twenty types of 4-connected 3D nets have been observed in the assembly of MOFs, such as **dia**, **nbo**, **cds**, **pts**, **sra**, **sod**, **qtz**, **crb**, **gis**, **lon**, **irl**, **pcb**, **pcl**, **neb**, **unh**, **cag**, **gsi**, **mmt**, **zni**, **lvt**, **tcb**, **ptt**, **mog**, **asv**, etc. Other two common 2D 4-connected underlying nets,