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Review

# Coordination polymers and metal-organic frameworks built up with poly(tetrazolate) ligands

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In the past two decades the chemistry of coordination polymers (CPs) and metal-organic frameworks (MOFs) has known an incessant development at both the academic and industrial level: the diversity they show in chemical composition, structural features and chemico-physical properties makes them attractive for a wide range of applications of technological, economical and/or environmental interest. Within this scenario, this review focuses the attention on the transition-metal-ion CPs and MOFs built up with bis-, tris- and tetrakis(5-substituted tetrazolate) ligands. After a short introduction on CPs and MOFs, and the chemistry of tetrazole and its derivatives, the reader is provided with a brief description of the synthesis, structural aspects and, whenever studied, functional properties of the numerous transition-metal poly(5-substituted tetrazolate) frameworks appeared in the literature to the end of 2017.
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*Abbreviations:* Alq3, tris(8-hydroxyquinoline)aluminum; BCEE, bis(2-cyanoethyl)ether; BET, Brunauer-Emmett-Teller; bpe, 1,2-bis(4-pyridyl)ethane; 4,4′-bpy, 4,4′-bipyridine; CP, coordination polymer; DCA, 9,10-dicyanoanthracene; dcbipy, 6,6′-dicyano-2,2′-bipyridine; DEF, *N,N′*-diethylformamide; DMA, *N,N′*-dimethylacetamide; DMF, *N,N′*-dimethylformamide; DMSO, dimethylsulfoxide; EtOH, ethanol; Htz, 1*H*-tetrazole; $H_2$Andolast, *N*-4-(1*H*-tetrazol-5-yl)phenyl-4-(1*H*-tetrazol-5-yl)benzamide; $H_2$BDMPX, 1,4-bis((3,5-dimethyl-1*H*-pyrazol-4-yl)methyl)benzene); 1,4-$H_2$BDP, 1,4-bis(1*H*-pyrazol-4-yl)benzene; 1,2-$H_2$BDT, 1,2-bis(1*H*-tetrazol-5-yl)benzene; 1,3-$H_2$BDT, 1,3-bis(1*H*-tetrazol-5-yl)benzene; 1,4-$H_2$BDT, 1,4-bis(1*H*-tetrazol-5-yl)benzene; $H_2$BDTM, 1,2-bis((1*H*-tetrazol-5-yl)methyl)benzene; $H_2$BDTri, 1,4-bis(1*H*-1,2,3-triazol-5-yl)benzene; $H_2$BPDT, 4,4′-biphenyl-bis(1*H*-tetrazol-5-yl); $H_2$BPZ, 4,4′-bipyrazole; $H_2$bta, *N,N*-bis(1(2)*H*-tetrazol-5-yl)amine; $H_2$btzb, 1,2-bis(1*H*-tetrazol-5-yl)butane; $H_2$btzbp, 6,6′-bis(1*H*-tetrazol-5-yl)-2,2′-bipyridine; $H_2$btze, 1,2-bis(1*H*-tetrazol-5-yl)ethane; $H_2$btzop, 1,5-bis(1*H*-tetrazol-5yl)-3-oxapentane; $H_2$btzp, 1,2-bis(1*H*-tetrazol-5-yl)propane; $H_2$-BTZPX, xylene-bis(1*H*-tetrazol-5-yl); $H_2$cdtp, 2,3-camphoryl-5,6-bis(1*H*-tetrazol-5-yl)pyrazine; $H_2$DMPMB, 4,4′-bis((3,5-dimethyl-1*H*-pyrazol-4-yl)methyl)biphenyl; $H_2$DTA, 9,10-bis(1*H*-tetrazol-5-yl)anthracene; $H_2$dtp, 2,3-bis(1*H*-tetrazol-5-yl)pyrazine; $H_2$MeBDT, 2-methyl-1,4-bis(1*H*-tetrazol-5-yl)benzene; $H_2$ox, oxalic acid; $H_2$ptp, 2,3-bis(pyridin-2-yl)-5,6-bis(1*H*-tetrazol-5-yl)pyrazine; $H_3$BTC, 1,3,5-benzenetricarboxylic acid; $H_3$BTP, 1,3,5-tris(1*H*-pyrazol-4-yl)benzene; $H_3$BTT, 1,3,5-tris(1*H*-tetrazol-5-yl)benzene; $H_3$BTT·2HCl, 1,3,5-tris(1*H*-tetrazol-5-yl)benzene dihydrochloride; $H_3$BTTri, 1,3,5-tris(1*H*-1,2,3-triazol-5-yl)benzene; $H_3$tatb, 4,4′,4″-*s*-triazine-2,4,6-triylbenzoic acid; $H_3$TPB-3tz, 1,3,5-tri-*p*-(1*H*-tetrazol-5-yl)phenylbenzene; $H_3$TPT-3tz, 2,4,6-tri-*p*-(1*H*-tetrazol-5-yl)-phenyl-s-triazine; $H_4$btc, biphenyl-3,3′,4,4′-tetracarboxylic acid; $H_4$tta, 1,3,5,7-tetrakis(1*H*-tetrazol-5-yl)adamantane; $H_4$ttpa, 1,3,5,7-tetrakis((1*H*-tetrazol-5-yl)phenyl)adamantane; $H_4$ttpm, tetrakis(4-tetrazolylphenyl)methane; $H_4$TTPP, 5,10,15,20-tetrakis(4-(1*H*-tetrazol-5-yl)phenyl)porphyrin; $H_4$ttps, tetrakis(4-tetrazolylphenyl)silane; IPN, isophthalonitrile; MeOH, methanol; Mn(III)Cl-ttzpp, 5,10,15,20-tetrakis[4-(2,3,4,5-tetrazolyl)phenyl]-porphyrinato manganese(III) chloride; MOF, metal-organic framework; OLEDs, organic light emitting diodes; PCP, porous coordination polymer; $PPh_3$, triphenylphosphine; py, pyridine; pyz, pyrazine; PXRD, powder X-ray diffraction; UV–Vis, ultraviolet–visible; XPS, X-ray photoelectron spectroscopy.

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## 1. Introduction

Coordination polymers (CPs) [1–3] and metal-organic frameworks (MOFs) [4–9] are hybrid inorganic/organic compounds with extended crystal structures built up by metal ions or metal-containing clusters bridged, through coordination bonds, by poly(-dentate) spacers. Both classes of compounds have known an incessant growth during the past two decades, raising the interest of chemists, material scientists and physicists at the academic and industrial level.

The variety of chemical composition, crystal structures and functional properties they show makes CPs and MOFs powerful platforms for their exploitation in fields of economical, technological and/or environmental interest ranging from luminescence [10–15] to microelectronics [16], conductivity [17–20], electrochemistry [21], high-energy materials [22], catalysis [23–29], magnetism [30–34], capture, storage and separation [35–43], sensing [15,16,44–46], drug delivery [47–49] or biomedicine [50–52].

The structural and functional diversity is granted by a careful selection of inorganic and organic building blocks [53–57], also under the guidance of computational chemistry methods [58–62]: main-group metal ions [1,4,63], transition metal ions and lanthanides [64,65] have been coupled to a great variety of organic ligands [66] typically containing oxygen [67], nitrogen [67–76] and phosphorus [77] as donor atoms.

Within this vast and appealing scenario, this review is aimed at gathering the transition-metal-ion CPs and MOFs built up with poly(tetrazolate) ligands that have been reported to the end of 2017. Together with poly(pyrazolates) and poly(imidazolates), poly(tetrazolates) are one of the three most employed classes of azolates in the synthesis of coordination polymers [67-–76]. There is a vast number of tetrazole ligands used as spacers to build up coordination frameworks [71,73,75]. In the following, we will systematically concentrate on CPs and MOFs with linkers possessing two to four 5-substituted tetrazoles. As detailed in Section 2, 5-substituted tetrazoles can be prepared following a safe, convenient and environmentally friendly synthetic path. In addition, 5-substituted tetrazolates bear four nitrogen donor atoms *per* heteroaromatic ring, this occurrence increasing the coordination modes of the ligand, hence enriching and diversifying the class of poly(tetrazolate) 1D to 3D coordination frameworks in terms of node-and-spacer connectivity, structural motif and functional properties.

Charts 1 and 2 collect the molecular structure and the acronym of the bis-, tris- and tetrakis(tetrazoles) employed in the preparation of the CPs and MOFs briefly described hereafter. To facilitate consultation, comparison and grouping, Table 1 provides the reader with the list of compounds presented in this review, together with details on network dimensionality, metal center stereochemistry and ligand hapticity. In the text, for each derivative a brief description is reported on the synthesis, structural aspects, thermal behaviour and, whenever studied, functional properties. To increase legibility, the investigated functional properties are collectively highlighted in Table 2. The compounds have been organized according to the transition metal ion, starting from the most populated family, namely: zinc(II), copper(I,II), manganese(II,III), cadmium(II), cobalt(II) and iron(II). The last section collects the few derivatives isolated with other metal centers [chromium(II,III), nickel(II), silver(I) and mercury(II)]. Schemes have been introduced to highlight the role of reaction conditions modulation. Figures are provided to illustrate the reader the most recurrent or fascinating structural units or topologies, or pictorially describe frequently studied functional properties.

Before briefly introducing the chemistry of tetrazole, the following advice appears to us necessary: rigorously, CPs are coordination compounds in which the building blocks – metal ions or metal-containing clusters, acting as nodes, and organic ligands, acting as spacers – are reciprocally linked by means of coordination bonds and develop repetitively into one, two or three dimensions [78]. When (permanent) porosity is present, they are referred to as either metal-organic frameworks (MOFs) or porous coordination polymers (PCPs). There exists a long-lasting debate in the scientific community (see e.g. [79,80]) about the use of these terms. Entering into or detailing upon such a debate is out of the scope of this review. In the following, we will use CP when dealing with non-porous compounds, MOF in all of the cases in which the material is porous.

## 2. Tetrazole and its derivatives – a brief overview

Since the accidental discovery of the first tetrazole-containing metal complex in 1885 by Bladin [81], who proposed the term

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