

Short communication

Diverse architectures and luminescence properties of three low-dimensional Zn(II)/Cd(II) coordination polymers based on a pyridine-imidazole ligand

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ABSTRACT

Based on a rigid ligand 3,5-bis(imidazole-1-yl)pyridine (bip), three new low-dimensional Zn(II)/Cd(II) coordination polymers $[\text{Zn}(\text{bip})(\text{Cl})_2]_n$ (**1**), $[\text{Zn}(\text{bip})(\text{Cl})_2]_n$ (**2**) and $[\text{Cd}(\text{bip})(p\text{-bdc})_{0.5}(\text{NO}_3)(\text{H}_2\text{O}) \cdot \text{H}_2\text{O}]_n$ (**3**) ($p\text{-H}_2\text{bdc} = 1,4\text{-benzenedicarboxylic acid}$) have been successfully obtained. Compounds **1** and **2** both feature zero-dimensional (0D) structures, whose asymmetric units are the same. Interestingly, the weak $\pi\text{-}\pi$ stacking arrangement in **1** and **2** is significantly different in molecular orientation, resulting in two entirely different three-dimensional (3D) supramolecular structures. Compound **3** is characterized as an infinite one-dimensional (1D) chain structure formed by the linkage of $\text{Cd}_2(\text{bip})_2$ units and linear $p\text{-bdc}^{2-}$ ligands, and is further extended into a 3D supramolecular architecture via weak $\pi\text{-}\pi$ stacking interactions. It can be observed by careful investigation of the two structures that different packing modes in **1** and **2** can afford different networks. The photoluminescence of compounds **1–3** has been investigated in the solid state at ambient temperature.

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The self-assembly of coordination networks will continue to be a trend due to not only their intriguing structural characteristics but also potential applications in photochemical areas, drug delivery, gas storage and separation, catalysis, and so on [1–8]. As compared to high dimensional frameworks which were produced more easily due to the high affinity of ligands to metal ions, the 3D supramolecular structures of low dimensional coordination polymers (CPs) have become a research interest in crystal engineering [9–12]. They can be sustained by weak interactions such as hydrogen bonding, $\pi\text{-}\pi$ stacking, and van der Waals interactions, and thus easily produce structure diversity during crystallization as well as interesting unique electro-conductive, magnetic, and non-linear optical properties which are different from those of high dimensional ones. When constructing low dimensional CPs, N-donor linear bridging ligands such as 4,4'-bipyridine and pyrazine were among the first ligands used [13,14]. In addition, the angular-shaped bridging ligands can also afford low dimensional structures. For instance, assembly of the rigid ligand 3,5-bis(imidazole-1-yl)pyridine (bip) with different transition metal ions generated a series of 1D and 2D CPs with promising ion exchange and luminescence properties [15].

As is known, there are many different factors that can affect the final structures of CPs, such as metal ions, organic ligands, temperature,

solvent system, pH value and various templates [16–20]. Significantly, the rational selection of organic ligands provides a possibility for the assembly of structurally controllable CPs. N-donor ligands (pyridine, imidazole, triazole, triazine, or tetrazole) are often employed to fabricate CPs because of their strong coordination abilities and versatile coordination geometries [21–26]. The bip ligand, which acts as a multi(pyridine-imidazole) ligand, integrates the features of both pyridine and polyimidazole, and has been widely used to construct CPs with diverse dimensional networks. What's more, utilizing mixed ligands has been proved to be an effective route to construct intriguing CPs [27–31]. It has been demonstrated that polycarboxylate ligands are excellent co-ligands and have been widely and profoundly employed, because carboxylate groups have versatile coordination modes to satisfy the geometric coordination requirement of metal centers [32–34]. Such a dual-ligand strategy gives the opportunity to construct attractive structures due to the distinct donors which can coordinate with metal centers through different coordination modes.

Considering all of the above-mentioned, we have successfully obtained three new low-dimensional coordination polymers based on the bip ligand, $[\text{Zn}(\text{bip})(\text{Cl})_2]_n$ (**1**), $[\text{Zn}(\text{bip})(\text{Cl})_2]_n$ (**2**) and $[\text{Cd}(\text{bip})(p\text{-bdc})_{0.5}(\text{NO}_3)(\text{H}_2\text{O}) \cdot \text{H}_2\text{O}]_n$ (**3**) ($p\text{-H}_2\text{bdc} = 1,4\text{-benzenedicarboxylic acid}$). Compounds **1** and **2** feature both 0D structures. Though they have the same asymmetric units, the whole 3D supramolecular structures of **1** and **2** are entirely different, which are caused by the different packing modes. Compound **3** possesses a 1D chain structure with the help of terephthalic acid which acts as auxiliary ligand. All the structures

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were determined by single-crystal X-ray diffraction analyses and further characterized by elemental analyses, infrared spectra (IR), powder X-ray diffraction (PXRD) and thermogravimetric analyses (TGA). We report herein the syntheses [35], structural characterization [36] and photoluminescent properties of the three low-dimensional CPs.

Single-crystal X-ray analysis reveals that compound **1** crystallizes in the orthorhombic space group of *Pbca*, while compound **2** belongs to the triclinic system with the space group *P*-1. It should be noticed that the asymmetric units of compounds **1** and **2** are the same, and both of them contain one Zn(II) ion, one bip ligand and two terminal Cl⁻ ions. Herein, the asymmetric unit of **1** as a representation is described in detail. As depicted in Fig. 1a, Zn(II) sits in a distorted tetrahedral geometry defined by two nitrogen atoms (N2 and N4A) from two different bip ligands and two chlorine ions where one chlorine ion (Cl1) composes the equatorial plane and another one (Cl2) is at the axial site. The Zn—N bond distances are consistent with the previously reported values [37–39], ranging from 1.998 Å to 2.009 Å for **1** and 2.001 to 2.021 Å for **2**.

To reduce the steric hindrance and increase structural stabilization, the three aromatic rings of bip ligand are non-planar, each outer imidazole group is twisted with respect to the central pyridyl ring. The dihedral angles between the central pyridyl ring and two imidazole groups are 33.290°, 46.157° for **1** and 42.472°, 52.698° for **2** (Figs. 1b and 2b). There are uncoordinated outer pyridyl group, so bip in both **1** and **2** acts as bidentate-bridging ligand linking two Zn(II) ions with two of the imidazole N-donors to generate the [Zn₂(bip)₂Cl₄] macrocycles.

The sizes of the [Zn₂(bip)₂Cl₄] rings are about 6.183 × 10.244 Å² for **1** and 5.967 × 10.501 Å² for **2**. The terminal Cl⁻ ions terminate the Zn(II) ions on two sides to yield the above-mentioned dinuclear ring, preventing the linkage between the neighbouring rings into a higher dimensional structure.

The two compounds display interesting and remarkably different molecular packing features as the result of their different dihedral angles of the bip ligands. Structural analyses of **1** and **2** reveal that weak π-π stacking interactions exist in the two compounds, which play an essential role in constructing the resulting three-dimensional (3D) supramolecular framework (Figs. 1c and 2c). As shown in Figs. 1d and 2d, the weak π-π stacking arrangement is significantly different in molecular orientation and centroid-centroid distances of imidazole rings are 4.020 Å for **1** and 4.051 Å for **2**, respectively.

The structure of compound **3** was solved by single-crystal X-ray diffraction in the triclinic space group *P*-1 and the asymmetric unit is composed of one crystallographically independent Cd(II) ion, one bip ligand, one coordinated and one free water molecule, half one *p*-bdc²⁻ and one NO₃⁻ by which charge neutrality is achieved. As depicted in Fig. 3a, the Cd center in **3** have a seven coordinated geometry, in which the different positions are occupied by two imidazole nitrogen atoms from two different bip ligands, two oxygen atoms from the same *p*-bdc²⁻, two oxygen atoms of one coordinated NO₃⁻ and one water molecule. The Cd—N and Cd—O (carboxyl) bond lengths are 2.230 Å, 2.258 Å and 2.377 Å, 2.394 Å, respectively, which correspond well with previously reported values [40,41]. The bond lengths of Cd—O (NO₃⁻) with 2.667 Å and

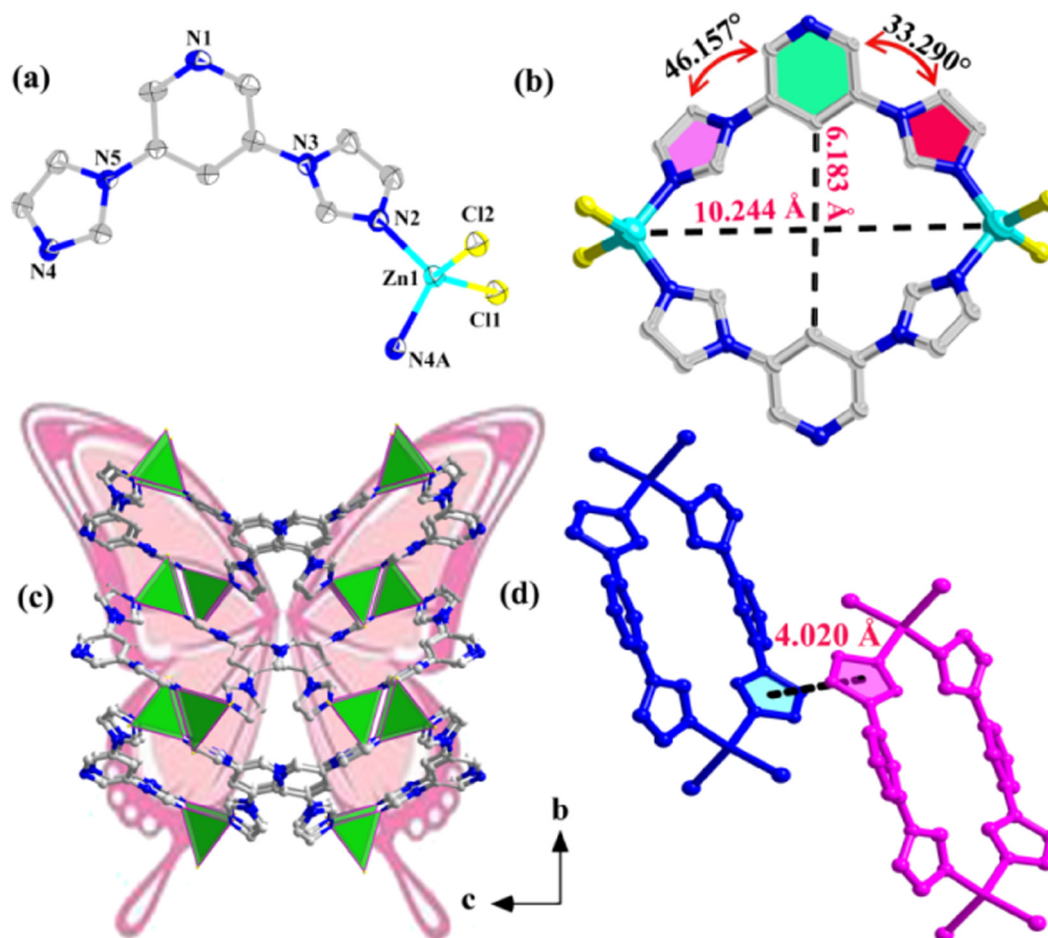


Fig. 1. (a) View of the coordination environments of Zn(II) ions in compound **1**; (b) The dihedral angles in [Zn₂(bip)₂Cl₄] ring; (c) The 3D butterfly-shaped supramolecular architecture along *a* axis; (d) Weak π-π interactions in **1**.

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