

Ligand configuration introduced into a 3D self-interpenetrated coordination polymer as visible-light-driven photocatalyst



Xiao Wang^{a,b}, Quan-Guo Zhai^{a,*}, Xiang-Yang Hou^b, Shu-Ni Li^a, Yu-Cheng Jiang^a, Man-Cheng Hu^{a,*}

^a Key Laboratory of Macromolecular Science of Shaanxi Province, School of Chemistry & Chemical Engineering, Shaanxi Normal University, Xi'an, Shaanxi 710062, PR China

^b Department of Chemistry and Chemical Engineering, Shaanxi Key Laboratory of Chemical Reaction Engineering, Yan'an University, Yan'an, Shaanxi 716000, PR China

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ABSTRACT

The combination of 1,4-bis(imidazol-1-ylmethyl)benzene (BIYB) and anthraquinone-2,7-disulfonic acid (H₂AQDA) ligands leads to a copper coordination polymer, namely, $[\text{Cu}(\text{BIYB})_2(\text{AQDA}) \cdot \text{H}_2\text{O}]_n$ (**1**). The *cis*-BIYB ligands link one unique Cu(II) atoms to give 1D double chains, but *trans*-BIYB connects the other Cu(II) centers to generate 2D (4,4)-layers. Each H₂AQDA ligand acts as a μ_2 -bridging linking the double chains and (4,4)-layers to generate the 3D framework of **1**. Cu(1) and Cu(2) can be viewed as 6- and 4-connected nodes, which lead to a (4,6)-connected self-penetrated network. Notably, complex **1** represents the rare example of coordination polymer-based visible-light-driven photocatalyst, and shows good degradation efficiency of organic dye.

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The designed construction of coordination polymers has been of tremendous interest, due to their esthetically pleasing structural features [1], as well as their promising applications in many areas including photocatalysis [2], luminescence [3], atypical magnetic [4] and so on. Carboxylate, N-containing ligands or their co-ligand systems have received considerable attention [5], owing to their versatile coordination models for the design of multifunctional coordination polymers. However, compared with them, little attention has been paid to the sulfonate-based assemblies, despite the fact that the sulfonate group bears diversified coordinating modes and ligating sensitivity to the nature (hardness) of the metal ions. Up to now, the ligands containing sulfonate group have been successfully used to investigate the lanthanide contraction effect [6] and the coordination chemistry of the “soft” metals such as Ag(I) [7]. To the best of our knowledge, the sulfonate-based ligands and N-containing ligands are rarely combined to construct coordination polymers. On the other hand, photocatalysis is a “green” technology for the treatment of contaminants, which has many advantages over other traditional methods [8]. However, the relatively wide band gap limits further application of such material in the visible-light region [9]. The advantages of coordination polymers as photocatalyst lie in the fact that the presence of organic linkers and transition metal centers, resulting in different ligand-to-metal charge-transfer transitions. Furthermore, coordination polymers usually exhibit absorption bands in the visible region, which indicate that they may undergo photochemical processes and exhibit responses upon visible-light excitation. Most recently, there have only been a few reports on the development of such new coordination polymer photocatalyst

materials [10]. As an extension of our research on the coordination chemistry of sulfonate ligands [7b] and azole ligands, we try to combine these two ligand systems to explore novel coordination polymer-based photocatalyst. Herein, the anthraquinone-2,7-disulfonic acid (H₂AQDA) was firstly selected to be mixed with 1,4-bis(imidazol-1-ylmethyl)benzene (BIYB), which led to a novel (4,6)-connected self-interpenetrated coordination polymer triggered by the *cis*- and *trans*-configurations of BIYB, namely, $[\text{Cu}(\text{BIYB})_2(\text{AQDA}) \cdot \text{H}_2\text{O}]_n$ (**1**) [11]. Under visible light irradiation, complex **1** showed good degradation efficiency of methylene blue.

Single-crystal X-ray diffraction analyses reveal that compound **1** crystallizes in triclinic *P*-1 space group [12]. As shown in Fig. 1, the asymmetric unit of **1** consists of two bivalent Cu ions, three independent BIYB ligands, one AQDA anion, and one crystallization water molecule. Both Cu(1) and Cu(2) ions are surrounded by four imidazole nitrogen atoms (N(5), N(7), N(5)#1 and N(7)#1 for Cu(1); N(1), N(1)#3, N(4)#2 and N(4)#4 for Cu(2)) from four different BIYB ligands to form square sphere. The Cu–N lengths and N–Cu–N angles are of 1.995(2)–2.022(2) Å and 89.27(9)–90.73(9)°, respectively. It should be noted that two of the three independent BIYB ligands are of *trans*-configuration, and the third one is of *cis*-configuration. As depicted in Fig. 2(a), two *trans*-BIYB ligands link Cu(1) atoms to give a 2D (4,4) layer in the *b*, *c*-plane. Notably, $[\text{Cu}_4(\text{BIYB})_4]$ large metal–organic rectangle, that is $[\text{Cu}_4(\text{benzene})_4(\text{CH}_2)_8(\text{imidazole})_8]$ or $[\text{Cu}_4\text{C}_36\text{N}_8]$ 48-membered rings are observed with dimensions of about 16.8×25.2 Å². On the other hand, Cu(2) is linked by *cis*-BIYB ligands to form a 1D double chain along the *a*-axis direction. $[\text{Cu}_2(\text{BIYB})_2]$ 26-membered metal–organic rings are also observed with Cu–Cu separations of 12.451(6) Å (Fig. 2(b)). As depicted in Fig. 2(c), each $[\text{Cu}_4(\text{BIYB})_4]$ rectangle from 2D net is enclosed by two $[\text{Cu}_2(\text{BIYB})_2]$ rings. The π - π

* Corresponding authors. Tel.: +86 29 81530767; fax: +86 29 81530727.
E-mail addresses: zhaiqg@snnu.edu.cn (Q.-G. Zhai), hmch@snnu.edu.cn (M.-C. Hu).

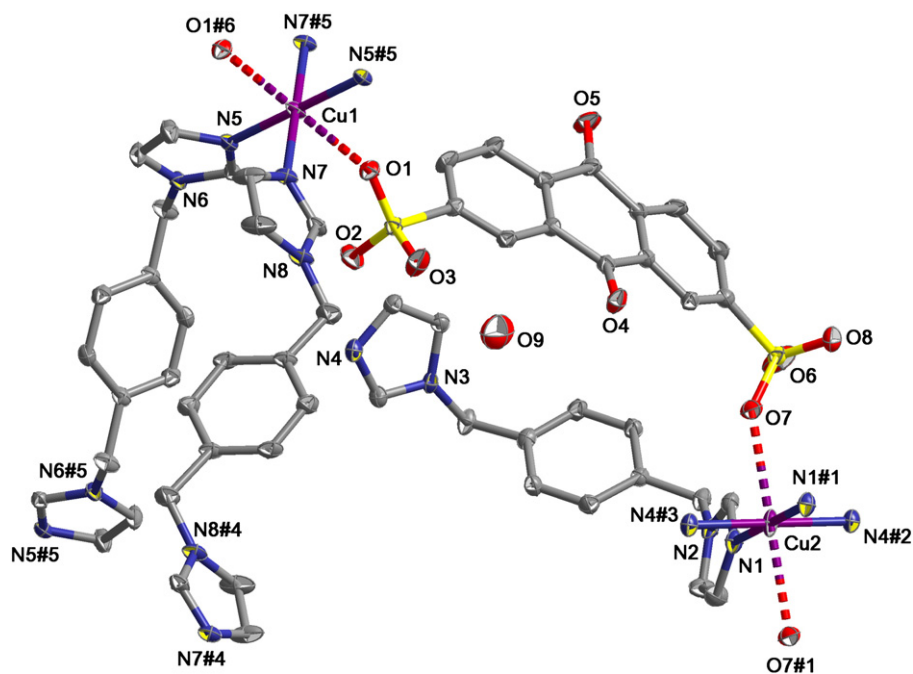


Fig. 1. The local coordination environments of 1. Symmetry codes: #1: $2 - x, -y, 1 - z$; #2: $1 + x, y, z$; #3: $1 - x, 2 - y, 1 - z$; #4: $1 - x, 2 - y, 1 - z$; #5: $-x, 1 - y, 1 - z$; #6: $-x, 1 - y, -z$.

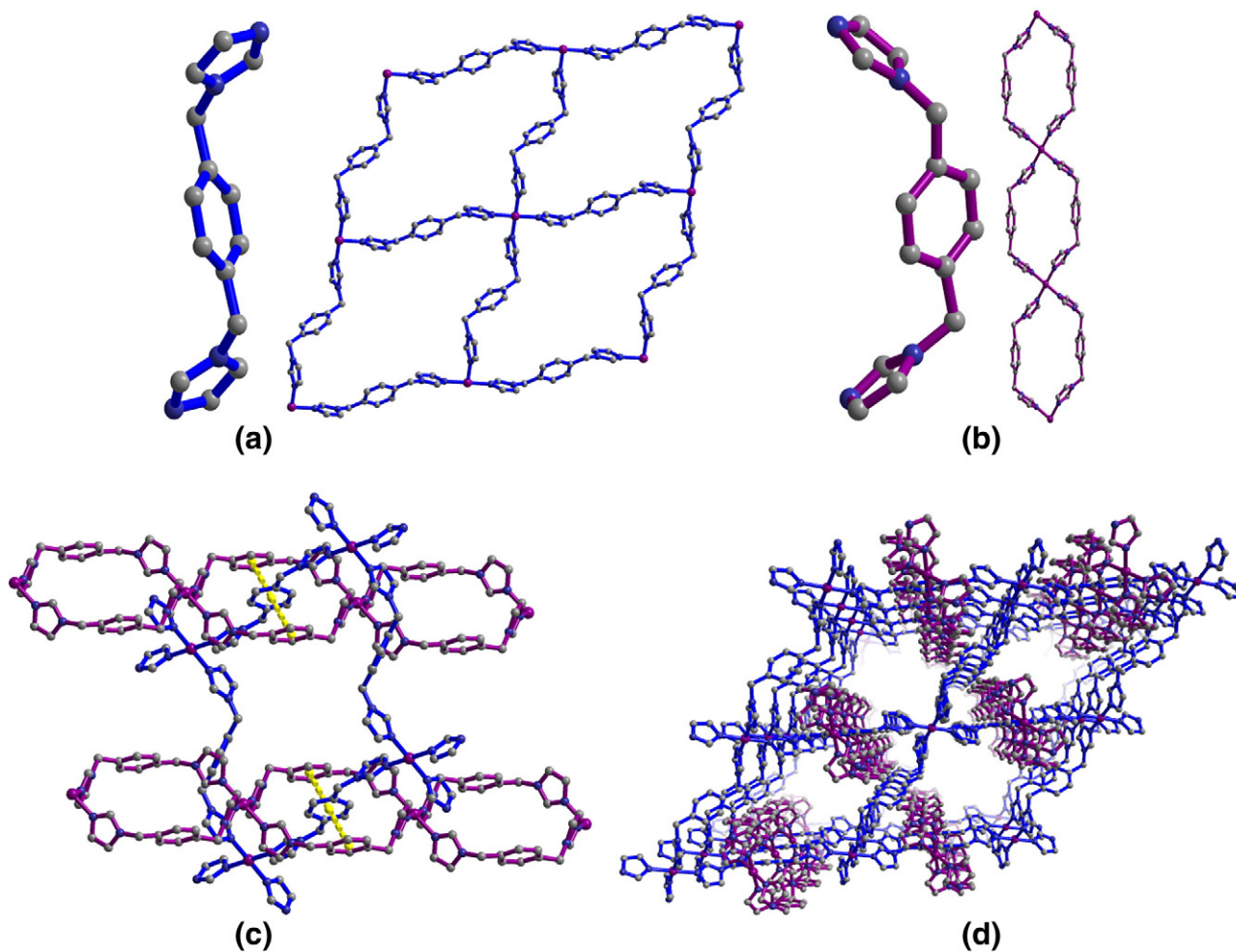


Fig. 2. (a) The 2D layer generated from Cu(1) and *trans*-BIYB ligands; (b) the 1D double chain generated from Cu(2) and *cis*-BIYB ligands; (c) the interpenetration modes exist in 1; (d) the porous framework of 1 generated by interpenetration.

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