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Novel L4(6)6(6) water layers trapped in an organic host framework 3-(1H-imidazol-1-yl)-1-phenylpropan-1-one

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ABSTRACT

Using a flexible imidazole derivate ligand 3-(1H-Imidazol-1-yl)-1-phenylpropan-1-one (L), novel 2D (two-dimensional) L4(6)6(6) water layers can be observed, which consist of fused cyclic water tetramers and hexamers. These 2D water layers are also trapped in supramolecular architecture $L\cdot 3H_2O$ (**1**). It is interesting that the C–H···π interactions obviously play an important role in stabilizing the packing structure, which also reveal the cooperative interactions between these supramolecular contacts in the construction of 2D water layers.

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1. Introduction

During the past few years much attention has been paid to theoretical and experimental studies of water due to their unusual properties and unique applications in many physical, chemical and biological processes [1–3]. Many small molecular water clusters such as tetramers [4], hexamers [3c,5,6] and octamers [7,8] also have been widely investigated because they can be seen as the basic building blocks of liquid water and ice. However there still needs more investigations about how these small water clusters are assembled into bulk water/ice. Therefore further attention also has been paid to multi-dimensional water morphologies, which structurally lie between small water clusters and bulk water/ice. The precise investigation of their structures helps to further understand the formation and anomalous behavior of bulk water/ice [9–16].

Recent studies also reveal that 2D water layers are quite important for the formation of bulk water or ice [3a,17,18]. For example, Janiak et al. systemically investigated 2D water/ice from 273 K down to 20 K, which reveals 2D water layers with 12-membered rings with a dangling D_2O molecule are important in the construction of 2D ice [10]. Ghosh et al. also use a cryptand ligand to construct novel 2D water layers containing alternatively fused 4- and 8-membered cyclic water clusters, in which ligand plays a key template factor for the formation of novel 2D water layers [11]. However, it is noted that though cyclic tetrameric and hexameric water clusters are often reported and well illustrated, only a few water layer structures simultaneously consisting of them as subunits are reported [2,19].

In this work, a flexible ligand, 3-(1H-Imidazol-1-yl)-1-phenylpropan-1-one (L), with plenty of active hydrogen-bonding sites has been synthesized. Its rich proton acceptor-donor groups encouraged us to utilize it as a host template to encapsulate novel water aggregates within its crystal lattice and to investigate the interactions between guest water molecules and organic host framework. With this in mind, a novel supramolecular compound $L\cdot 3H_2O$ (**1**) is isolated, in which novel two-dimensional water layers can be observed. These 2D water layers consist of fused cyclic water tetramers and hexamers, which can be classified as L4(6)6(6) according to Infantes and Motherwell [2]. The structure of **1** contains the subunits of T4(1) water chains similar to previous examples [19], which also arise our interest. On the other hand, it is also interesting that weak C–H···π interactions obviously play a key role in the stabilization of the supramolecular architecture, which is also scarcely reported in the previous examples in the construction of 2D water layers.

2. Experimental

2.1. General

The ligand 3-(1H-Imidazol-1-yl)-1-phenylpropan-1-one (L) was synthesized according to the literature method [20]. All other reagents were commercially available and used without further purification. The C, H, and N elemental analyses were carried out with a CE-440 (Leemanlabs) analyzer. Fourier Transform (FT)-IR

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spectra (KBr pellets) were taken on an AVATAR-330 (Nicolet) spectrometer.

2.2. Preparation

2.2.1. Synthesis of L·3H$_2$O (**1**)

The ethanol solution (15 mL) of L (21 mg, 0.1 mmol) was added into a solution of Mn(OAc)$_2$·4H$_2$O (24.5 mg, 0.1 mmol) in ethanol–H$_2$O (10 mL, 1:1) with vigorous stirring for *ca* 30 min. The reaction solution was filtered and left to stand at room temperature. Colorless block-like single crystals of **1** suitable for X-ray diffraction were obtained after several days by slow evaporation of the solvent (yield 46%). Elemental analysis (%) for **1**: found: C, 56.67; H, 6.21; N, 11.07%. Calc. for $C_{12}H_{18}N_2O_4$: C, 56.68; H, 6.18; N, 11.02%.

2.3. X-ray crystallography

Single-crystal X-ray diffraction data collections of complexes **1** (0.24 × 0.22 × 0.18 mm) were performed on a Bruker Apex II CCD diffractometer at 293(2) K with Mo-Kα radiation ($\lambda = 0.71073$ Å). A total of 2373 reflections were collected, of which 1929 are independent. Both structures were solved by direct methods and refined by full-matrix least-squares with SHELXTL package (semi-empirical absorption corrections were applied using SADABS program). All non-hydrogen atoms were located in successive difference Fourier syntheses. The final refinement was performed by full-matrix least-squares methods with anisotropic thermal parameters for non-hydrogen atoms on F^2. H atoms of organic ligands were generated with assigned isotropic thermal parameters, and those of coordinated water were first found in difference electron density maps, and then placed in the calculated sites and included in the final refinement. The crystallographic data is listed in Table 1. Crystallographic data (excluding structure factors) for the crystal structures reported in this paper has been deposited with the Cambridge Crystallographic Data Center (CCDC No. 686965). This material can be obtained free of charge via www.ccdc.cam.ac.uk/conts/retrieving.html (or from the CCDC, 12 Union Road, Cambridge CB2 1EZ, UK; fax: +44 1223 336033; e-mail: deposit@ccdc.cam.ac.uk).

3. Results and discussion

3.1. Description of crystal structures

Single-crystal X-ray diffraction analysis reveals that **1** crystallizes in space group *P*2(1)/*c*, the asymmetric unit of **1** contains one L ligand and three independent lattice water molecules (O2, O3 and O4) (Fig. 1). The imidazole and phenyl rings of L are not coplanar, the corresponding dihedral angles formed by the least-squares planes of phenyl and imidazole rings is 85.06(5)°.

As is shown in Fig. 2(a), a novel 2D water layer consisting of cyclic tetramers and hexamers can be observed along the crystallographic [001] direction. Each tetrameric water cluster is made up of O2, O2A, O3 and O4, in which the four oxygen atoms are nearly coplanar. While each hexameric water cluster consists of O2A, O2B, O3, O3A, O4A and O4B, in which they are also nearly

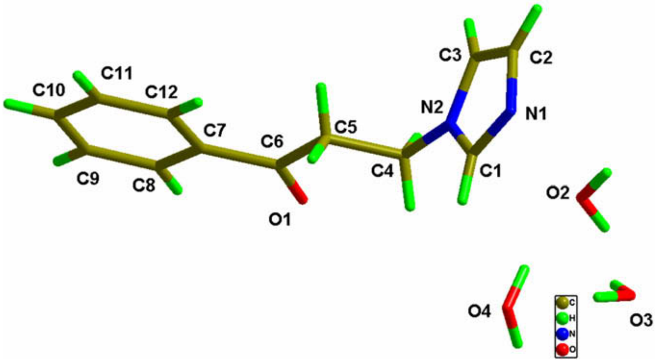

Fig. 1. The representation of **1** with selected numbering scheme.

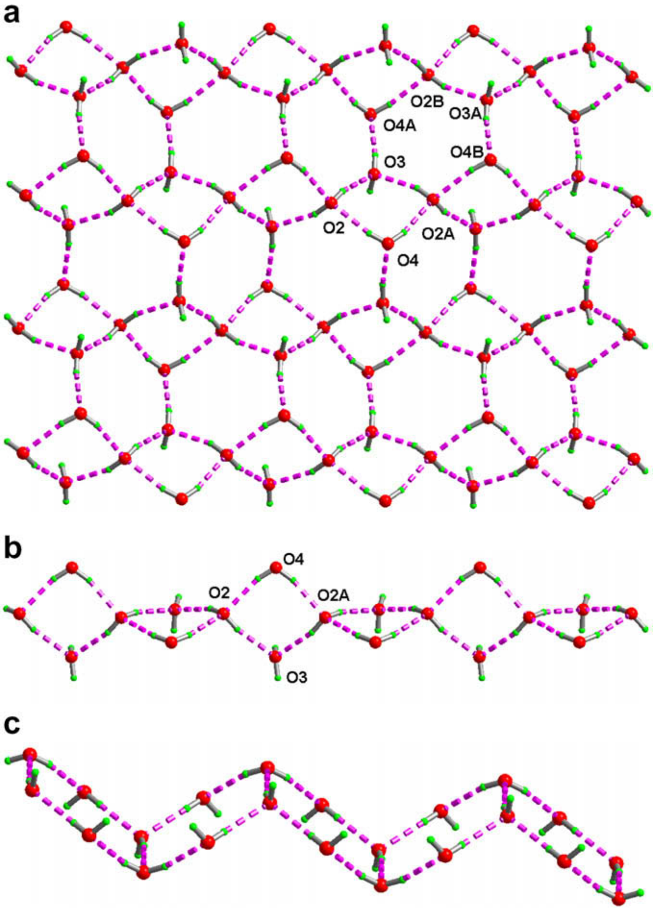

Fig. 2. (a) Neutral 2D L4(6)6(6) water layers. (b) The subunit of 1D water tape formed by perpedicular alternatively linked tetrameric water clusters. (c) 1D ladder-like water tape formed by planar hexameric water clusters.

Table 1
Crystallographic data and structural refinement for compound **1**

Empirical formula	$C_{12}H_{18}O_4N_2$
Formula weight	254.28
Crystal system	Monoclinic
Space group	*P*2(1)/*c*
T (K)	293(2)
a (Å)	16.147(11)
b (Å)	10.205(7)
c (Å)	8.224(6)
β (°)	96.898(8)
V [Å^3]	1345.4(16)
Z	4
F(000)	544
ρ [Mg/m^3]	1.255
GOF	1.034
$R_1(I > 2\sigma(I))$	0.0339
$WR_2(I > 2\sigma(I))$	0.0913
R_1 (all data)	0.0421
WR_2 (all data)	0.0971

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