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The crystal structure of 1,4-bis(1*H*-imidazol-3-ium-1-yl)benzene dinitrate, $C_{12}H_{12}N_4^{2+} \cdot 2(NO_3^-)$

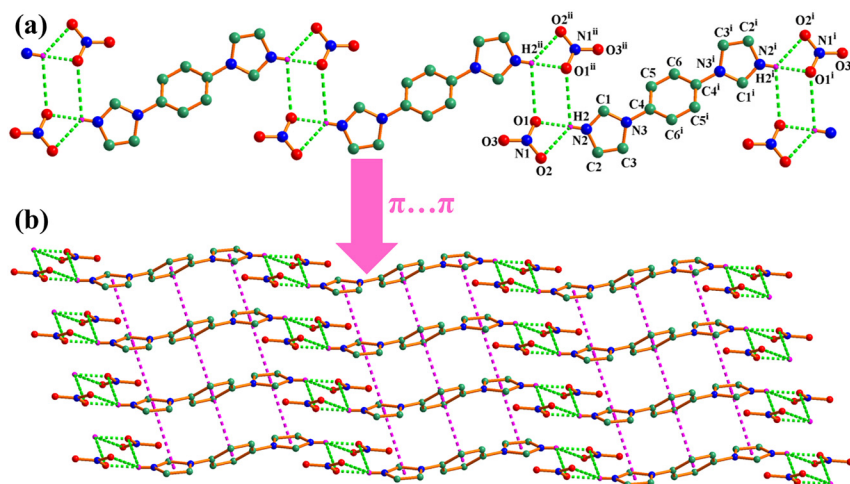


Figure 1: The one-dimensional hydrogen bonding chain formed between the cations $[H_2bib]^{2+}$ and the anions $[NO_3^-]$, and further assembled to a two-dimensional layer structure with the $\pi \cdots \pi$ interactions in I. (a) The one-dimensional hydrogen bonding (N–H $\cdots$ O hydrogen bonds as green dashed lines) structure of I, showing the part atom numbering scheme (symmetry codes: i 1–x, 1–y, 2–z; ii 2–x, 1–y, 1–z); (b) View of the 2D $\pi \cdots \pi$ interactions and N–H $\cdots$ O hydrogen bonded framework in I. The $\pi \cdots \pi$ interactions and N–H $\cdots$ O hydrogen bonds show as dashed lines.

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Abstract

$[H_2(C_{12}H_{10}N_4)]^{2+} \cdot 2(NO_3^-)$, monoclinic, $P2_1/c$ (no. 14), $a = 3.6937(3)$ Å, $b = 15.5641(10)$ Å, $c = 12.3322(8)$ Å, $\beta = 93.201(7)^\circ$, $V = 707.86(9)$ Å³, $Z = 2$, $R_{gt}(F) = 0.0377$, $wR_{ref}(F^2) = 0.1076$, $T = 298.15$ K.

CCDC no.: 2357529

Table 1: Data collection and handling.

Crystal:	Colorless block
Size:	0.25 × 0.18 × 0.12 mm
Wavelength:	Mo K α radiation (0.71073 Å)
μ :	0.13 mm ^{−1}
Diffractometer, scan mode:	XtaLAB Mini (ROW), ω
θ_{max} , completeness:	25.1°, >99 %
$N(hkl)_{measured}$, $N(hkl)_{unique}$, R_{int} :	2,511, 1,267, 0.023
Criterion for I_{obs} , $N(hkl)_{gt}$:	$I_{obs} > 2\sigma(I_{obs})$, 1,026
$N(param)_{refined}$:	109
Programs:	CrysAlis ^{PRO} , ¹ Olex2, ² SHELX, ^{3,4} PLATON ⁵

Table 1 contains crystallographic data and Table 2 contains the list of the atoms including atomic coordinates and displacement parameters.

1 Source of materials

All chemicals were purchased from commercial sources and used as received. A mixture of $Zn(NO_3)_2$ (2.0 mL 0.10 mol L^{−1} $Zn(NO_3)_2$ solution, 0.20 mmol), 2,3-quinoline dicarboxy-acid (0.0221 g, 0.10 mmol), 1,4-bis(1*H*-imidazol-1-yl)benzene (1,4-bis(1*H*-imidazol-1-yl)benzene is abbreviated as bib, 0.0212 g, 0.10 mmol), NaOH (2.0 mL 0.10 mol L^{−1} NaOH solution, 0.20 mmol), and H₂O/anhydrous ethanol/DMF (3.0 mL/4.0 mL/2.0 mL) was added to a 25 mL Teflon-lined stainless steel reactor and heated at 413 K for 3 days. After cooling to room temperature at a rate of 283 K h^{−1}, colorless block-shaped crystals of I were collected by filtration, washed with anhydrous ethanol and dried in air. Phase pure crystals were obtained by manual separation (Yield: 11.77 mg, ca. 35 %

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Table 2: Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters ($\text{\AA}^2$).

Atom	<i>x</i>	<i>y</i>	<i>z</i>	$U_{\text{iso}}^*/U_{\text{eq}}$
C1	0.6582 (5)	0.54507 (11)	0.70841 (13)	0.0324 (4)
H1	0.745922	0.489362	0.702393	0.039*
C2	0.4930 (5)	0.67712 (12)	0.66923 (15)	0.0394 (5)
H2A	0.448646	0.727489	0.630157	0.047*
C3	0.4323 (5)	0.66430 (10)	0.77402 (14)	0.0343 (4)
H3	0.338443	0.703857	0.821496	0.041*
C4	0.5181 (4)	0.53909 (10)	0.90101 (13)	0.0267 (4)
C5	0.4148 (5)	0.45401 (10)	0.90532 (13)	0.0315 (4)
H5	0.357859	0.423640	0.841695	0.038*
C6	0.3967 (5)	0.41441 (10)	1.00516 (13)	0.0312 (4)
H6	0.328092	0.357069	1.009354	0.037*
N1	0.9625 (4)	0.65064 (9)	0.38770 (11)	0.0332 (4)
N2	0.6320 (4)	0.60271 (9)	0.63025 (12)	0.0378 (4)
H2	0.692946	0.594713	0.564720	0.045*
N3	0.5365 (4)	0.58078 (8)	0.79788 (10)	0.0280 (4)
O1	0.8518 (5)	0.58010 (8)	0.42392 (11)	0.0520 (4)
O2	0.9613 (4)	0.71553 (8)	0.44573 (10)	0.0492 (4)
O3	1.0731 (4)	0.65285 (8)	0.29470 (10)	0.0469 (4)

based on 1,4-bis(1*H*-imidazol-1-yl)benzene). Anal. Calc. for I: $C_{12}H_{12}N_6O_6$ (%) (Mr = 336.28): C, 42.82; H, 3.57; N, 24.98. Found: C, 42.80; H, 3.59; N, 24.97 (CCDC number 2357529).

2 Experimental details

CrysAlisPro 1.171.39.46 (Rigaku Oxford Diffraction, 2018) was used for empirical absorption correction using spherical harmonics, implemented in SCALE3 ABSPACK scaling algorithm.¹ Using Olex2,² the structure was solved with the ShelXT³ structure solution program and refined with the ShelXL⁴ refinement package. Nitrogen or carbon-bound hydrogen atoms were placed in calculated positions $d = 0.86 \text{ \AA}$ for NH, and $d = 0.93 \text{ \AA}$ for CH and were included in the refinement in the riding model approximation, with $U_{\text{iso}}(\text{H})$ set to $1.2U_{\text{eq}}(\text{N or C})$ for $-\text{NH}$ and $-\text{CH}$. The structure was examined using the ADDSYM subroutine of PLATON⁵ to ensure that no additional symmetry could be applied to the models.

3 Comment

The choice and design of organic ligands are of great importance in the construction of MOFs. Neutral N-donor bridging ligand 1,4-bis(1*H*-imidazol-1-yl)benzene and its derivatives have been widely used to explore new

POM-based metal-organic frameworks (polyoxometalate abbreviated as POM).^{6–8} Three crucial points should be considered in choosing ligands: (i) The coordination ability between ligands and transition metal (TM) ions. Initially, pyridine-based bridging ligands were widely used in the construction of POM-based networks. Later, imidazole-based and multiazole-based ligands were shown to have stronger coordination ability, which increased the number of POM-based networks.^{9–11} (ii) The rigidity and flexibility of the ligands. Generally, using rigid ligands is essential in constructing frameworks, because flexible ligands can easily cause distortion and instability of the frameworks, producing an entangled network, and finally making pores disappear.^{12–15} (iii) The length of bridging ligands should be similar to the diameter of the Keggin-type anions. If the organic ligand is too short, the pores of the metal-organic frameworks will be occupied or terminated by POM anions, making it harder to generate frameworks.¹⁶ Using 1,4-bis(1*H*-imidazol-1-yl)benzene as the structural model for retrieval, there were found 570 crystal structure hits in the Cambridge Crystal Structural Database (ConQuest 2022.3.0, CSD), in which there are a total of 42 crystal structures containing 1,4-bis(1*H*-imidazol-3-ium-1-yl)benzene. On the basis of the above analysis, it has been further found that 1,4-bis(1*H*-imidazol-1-yl)benzene often acts as both a rigid μ_2 bridging and a divalent hum salt cation in constructing POM-based MOFs.^{17–19} To the best of our ability, only 2 crystal structures with 1,4-bis(1*H*-imidazol-3-ium-1-yl)benzene crystallization in inorganic anion with balanced charge, were searched in the CSD, i.e. 1,4-bis(1*H*-imidazol-3-ium-1-yl)benzene diperchlorate²⁰ and 1,1'-(*p*-phenylene)-bis(1*H*-imidazol-3-ium) hexafluorosilicate(iv).²¹ More importantly, 1,4-bis(1*H*-imidazol-1-yl)benzene and its derivatives have been found to possess widespread biological activities, many of them have been developed as medical and functional materials. Based on the above considerations, the title compound, (I), (Figure 1) was obtained unexpectedly as the product of an attempted synthesis of a network complex of Zn(II) using mixed H_2O /anhydrous ethanol/DMF as the solvent.

The crystallographic asymmetric unit of (I) comprises a nitrate anion $[\text{NO}_3^-]$, in a general position, and half a protonated 1,4-di(1-imidazolyl)benzene cation $[1,1'-(p\text{-phenylene})\text{bis}(1\text{H-imidazol-3-ium})]$, as this is disposed about a centre of inversion. In I, the two imidazolyl rings (5-membered ring, Cg1: C1/N2/C2/C3/N3) and (5-membered ring, Cg3: C1^{*i*}/N2^{*i*}/C2^{*i*}/C3^{*i*}/N3^{*i*}, symmetry code: $i = 1 - x, 1 - y, 2 - z$) are coplanar, but the benzene ring (6-membered ring, Cg2: C4/C5/C6/C4^{*i*}/C5^{*i*}/C6^{*i*}) and imidazolyl rings are seriously twisted. The dihedral angle between the benzene and imidazolyl rings is $39.00(7)^\circ$, which is larger than the reported similar compounds, i.e. 1,1'-

(*p*-phenylene)bis(1*H*-imidazol-3-ium) hexafluoridosilicate(IV) (27.80(11)°)²¹ and 1,4-(di(hydrogen-imidazolyl)benzene)bis perchlorate (34.88°).²⁰ The bond lengths and angles are within normal ranges. The closeness in the N2–C1, C2 bond lengths [1.316(2) & 1.365(2) Å] confirms that proton transfer occurred during co-crystallisation; see below for further comment. The angles subtended at the N2 atom [C1–N2–C2 = 109.52(15)°] is unequivalent to that subtended at the N3 atom [C3–N3–C4 = 126.03(13)°], confirming the N2 atom is partially protonated. In the 1,4-bis(1*H*-imidazol-3-ium-1-yl)benzene analogue of (I), the comparable angles were disparate at 113.09(9) and 104.80(9)°, with the former corresponding to the protonated imidazole-N atom.^{22,23} The imidazole ring is planar to –0.006(1) Å and an evaluation of the bond lengths within the five-membered ring is consistent with the significant delocalisation of π -electron density over the five atoms. Thus, the C1–N2 [1.316(2) Å] and C2–N2 [1.365(2) Å] bond lengths are experimentally equivalent as the C1–N3 [1.336(2) Å] and C3–N3 [1.383(2) Å] bonds; C2–C3 = 1.339(2) Å. This observation provides further support for the protonation at each terminal nitrogen (N2 and N2^b) site of the imidazole ring. The structure model of the NO₃[–] in (I) represents a flat triangle with an N atom at the center and three O atoms at the corners; see below for further validation. The angles between the bonds are nearly identical and measured to be approximate 120° [O3–N1–O1 = 118.81(14)°, O2–N1–O3 = 121.62(14)°, O2–N1–O1 = 119.57(14)°], and the distances $d(N1–O1)$ = 1.2620(18) Å, $d(N1–O2)$ = 1.2380(18) Å, and $d(N1–O1)$ = 1.2392(18) Å. The partial electric charges of N and O atoms in the nitrate ion were determined to be qN = 0.5741e, qO = –0.5247e (e is the elementary charge).²⁴ As a whole, the ion showed the electric charge = –1e.

In the crystal of (I), all potential hydrogen bond acceptors and donors participate in charge-assisted hydrogen bonding interactions. Interestingly, the cations and anions associate about a 2-fold axis via imidazole-N–H $\cdots$ O (nitrate) $\cdots$ H–N–imidazole hydrogen bonds leading to three edge-sharing hydrogen-bonded rings, i.e. a four-membered R_4^4 { $\cdots H \cdots O \cdots H \cdots O \cdots$ } (R_4^4 hydrogen bonded ring) and two four-membered R_2^4 { $\cdots H \cdots O \cdots N \cdots O \cdots$ } synthons.²⁵ In detail, the two protonated nitrogen atoms (N1 and N1^b) of imidazole group and nitrate oxygen atoms (O1, O2 and O1ⁱⁱ, symmetry code: ii 2 – x, 1 – y, 1 – z) are the hydrogen-bonding donor and acceptor (N1–H2 $\cdots$ O1: $d(H2 \cdots O1)$ = 1.88(2) Å, $d(N1 \cdots O1)$ = 2.737(2) Å and N1–H2 $\cdots$ O1 = 177.01(2)°; N1–H2 $\cdots$ O2: $d(H2 \cdots O2)$ = 2.61(4) Å, $d(N1 \cdots O2)$ = 3.170(2) Å and N1–H2 $\cdots$ O2 = 123.41(5)°; N2–H2 $\cdots$ O1ⁱⁱ: $d(H2 \cdots O1^{ii})$ = 3.19(7) Å, $d(N1 \cdots O1^{ii})$ = 3.510(2) Å and N1–H2 $\cdots$ O1ⁱⁱ = 104.27(7)°, respectively, resulting in three significant N–H $\cdots$ O hydrogen bonds in (I). These R_4^4 and R_2^4 hydrogen bonded rings parallel alternate arrangement among the 1,4-bis(1*H*-imidazol-3-ium-1-yl)benzene cations, leading to a 1D zigzag

hydrogen bonding chain by translation along the *c* axis. Moreover, these chains are further interconnected by weak intermolecular C–H $\cdots$ O hydrogen bonds (C1–H1 $\cdots$ O1ⁱⁱ, C1–H1 $\cdots$ O3ⁱⁱ, C3–H3 $\cdots$ O2ⁱⁱⁱ, C3–H3 $\cdots$ O3ⁱⁱⁱ, C5–H5 $\cdots$ O3^{iv}, C6–H6 $\cdots$ O2^v, symmetry codes: iii –1 + x, 1.5 – y, 0.5 + z; iv 1 – x, 1 – y, 1 – z; v 1 – x, –0.5 + y, 1.5 – z) involved in the three nitrate O-atoms (acceptor). The separations of $d(O \cdots H)$ and $d(C \cdots O)$, and the angle of C–H $\cdots$ O are ranged from 2.31 and 3.156(2) Å, and 133 % to 2.55 and 3.433(2) Å, and 173°. In addition, the salt lies in two kinds of $\pi \cdots \pi$ stacking interactions between the imidazole rings (Cg1/Cg1^{vi}, Cg1/Cg1^{vii}, symmetry codes: vi –1 + x, y, z; vii 1 + x, y, z), and between the phenyl rings (Cg2/Cg2^{vi}, Cg2/Cg2^{vii}, Cg2/Cg2^{viii}, Cg2/Cg2^{ix}, symmetry codes: viii –x, 1 – y, 2 – z; ix 2 – x, 1 – y, 2 – z), both related to the 1,4-bis(1*H*-imidazol-3-ium-1-yl)benzene cations. The dihedral angles between the Cg1/Cg1^{vi} or Cg1/Cg1^{vii} is 0.02(11)°, with a centroid-to-centroid distance of 3.6937(11) Å, and a perpendicular distance of 3.5445(7) Å. The dihedral angles between the Cg2/Cg2^{vi} or Cg2/Cg2^{vii} or Cg2/Cg2^{viii} or Cg2/Cg2^{ix} is 0.00°, with a centroid-to-centroid distance of 3.6937(10) Å, and a perpendicular distance of 3.4207(15) or 3.4580(13) Å. It is worth mentioning that the two Cg $\cdots$ Cg distances and their dihedral angles are almost the same. However, their contribution to the overall lattice energy must be very small. Thus a supramolecular 3D network fragment is formed by N–H $\cdots$ O, C–H $\cdots$ O, and $\pi \cdots \pi$ interactions stabilizing the compound. Furthermore, PLATON⁵ analysis shows that the unit cell contains no residual solvent accessible void.

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