Crystal structure of 1,1'-(1,4-phenylenebis(methylene))bis(2-methyl-1*H*-benzo[*d*]imidazol-3-ium) 2,5-dicarboxyterephthalate, $C_{17}H_{14}N_2O_4$

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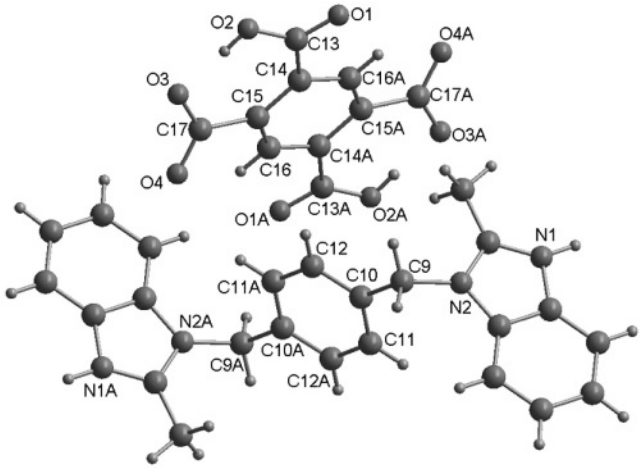
Received November 14, 2014, accepted April 16, 2015, available online April 21, 2015, CCDC no. 1267/4304



Abstract

$C_{17}H_{14}N_2O_4$, triclinic, $P\bar{1}$ (no. 2), a = 7.025(1) Å, b = 9.607(2) Å, c = 10.749(2) Å, α = 97.47(3)°, β = 92.10(3)°, γ = 91.01(3)°, V = 718.6 Å^3, Z = 2, $R_{gt}(F)$ = 0.0564, $wR_{ref}(F^2)$ = 0.1251, T = 293 K.

Table 1. Data collection and handling.

Crystal:	colourless blocks, size 0.17×0.22×0.36 mm
Wavelength:	Mo K_α radiation (0.71073 Å)
μ:	1.04 cm^{-1}
Diffractometer, scan mode:	Bruker P4, ω
$2\theta_{max}$:	51°
$N(hkl)_{measured}$, $N(hkl)_{unique}$:	7549, 2661
Criterion for I_{obs}, $N(hkl)_{gt}$:	$I_{obs} > 2\,\sigma(I_{obs})$, 2360
$N(param)_{refined}$:	210
Programs:	SHELX [10]

Source of material

A mixture of 1,2,4,5-benzenetetracarboxylic acid (0.1 mmol, 19.5 mg), $Cd(OAc)_2{\cdot}2H_2O$ (0.1 mmol, 238 mg), THF (10 mL), NaH solution (5 mL), 2-methyl-1*H*-benzoimidazole (0.3 mmol, 132 mg), 1,4-bis-(2-bromo-ethyl)-benzene (0.1 mmol, 292 mg) and H_2O (10mL) was heated in a 25 mL Teflon-lined autoclave under autogenous pressure at 413K for four days. After cooling to room temperature, colourless crystals were collected by filtration and washed with distilled water.

Discussion

Over the past few decades, supramolecular chemistry depending upon hydrogen bonding and intermolecular weak interactions have been a field of rapid growth due to potential application such as catalysis, magnetism, semiconductor, nonlinear optical materials, biological activity [1–6]. Benzimidazole and its derivatives, which are one kind of important building modules of N-containing heterocyclic ring compounds, may provide supramolecular recognition sites for N–H···O, C–H···O, C–H···π and π···π interactions to construct supramolecular architectures. On account of its various coordination modes, multi-carboxylate ligands are extremely popular as efficient linkers in the construction of supramolecular compounds. Thereinto, the deprotonated analoga of the 1,2,4,5-benzenetetracarboxylic acid (bta) as unsymmetrical carboxylate bridging ligands possess two or more coordination sites with differing donor abilities. Its carboxylate group can also be used as hydrogen-bond acceptor. To our knowledge, the discrete subunits or low-dimensional entities have been linked higher dimensional supramolecular compounds through hydrogen bonds, π···π stacking and host-guest interactions [7–9]. In this contribution, we have report on a supramolecular network constructed via hydrogen bonds and π···π interactions. The title structure comprises half of a 1,1'-(1,4-phenylenebis(methylene))bis(2-methyl-1*H*-benzo[*d*]imidazol-3-ium) dication and a one half of a 2,5-dicarboxyterephthalate per asymmetric unit. The carboxylate groups of the acid are incomplete protonated. Several kinds of hydrogen bonding are observed in the crystal structure: (a) the hydrogen bonding between oxygen atoms of carboxylate /carboxy motif (d(O···O): 2.398(3) Å and 2.682(3) Å, and O–H···O are 177.3° and 170.4°, respectively; (b) the hydrogen bonding between carboxylate oxygen atoms and nitrogen atoms of 1,1'-(1,4-phenylenebis(methylene))bis(2-methyl-1*H*-benzo[*d*]imidazol-3-ium) moiety (d(O···N): 2.652(3) Å and N–H···O is 167.9°). These hydrogen bonds form a one-dimensional chain structure. Weaker π···π interactions are also observed between the 1,1'-(1,4-phenylenebis(methylene))bis(2-methyl-1*H*-benzo[*d*]imidazol-3-ium) moiety with the centroid–centroid distance of 3.792 Å, further consolidating the stability of the chain structure.

Table 2. Atomic coordinates and displacement parameters (in Å^2).

Atom	Site	x	y	z	U_{iso}
H(1)	2*i*	0.3494	0.2152	1.0440	0.044
H(16)	2*i*	0.6575	0.3619	0.3512	0.040
H(9A)	2*i*	0.1106	0.1316	0.6384	0.047
H(9B)	2*i*	0.0848	–0.0310	0.6362	0.047
H(11)	2*i*	0.4728	–0.1189	0.6719	0.045
H(12)	2*i*	0.2730	0.1599	0.4569	0.044
H(2)	2*i*	0.1229	0.5845	0.2903	0.102
H(4)	2*i*	0.2858	–0.0170	1.1731	0.057
H(7)	2*i*	0.0947	–0.2218	0.7654	0.055
H(6)	2*i*	0.0887	–0.3480	0.9344	0.068
H(1A)	2*i*	0.3653	0.3837	0.8884	0.081
H(1B)	2*i*	0.2255	0.3425	0.7719	0.081
H(1C)	2*i*	0.4402	0.3015	0.7648	0.081
H(5)	2*i*	0.1813	–0.2486	1.1332	0.070

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Table 3. Atomic coordinates and displacement parameters (in Å^2).

Atom	Site	x	y	z	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
N(2)	2*i*	0.2212(2)	0.0623(2)	0.7932(1)	0.0296(9)	0.040(1)	0.0242(8)	0.0006(7)	0.0041(7)	–0.0002(7)
N(1)	2*i*	0.3087(2)	0.1549(2)	0.9821(2)	0.034(1)	0.044(1)	0.0289(9)	0.0032(8)	–0.0007(7)	–0.0060(8)
C(14)	2*i*	0.3210(3)	0.5553(2)	0.4686(2)	0.031(1)	0.029(1)	0.033(1)	0.0014(8)	–0.0052(8)	0.0032(8)
C(16)	2*i*	0.5927(3)	0.4177(2)	0.4123(2)	0.036(1)	0.032(1)	0.031(1)	0.0039(9)	–0.0007(8)	–0.0037(8)
C(8)	2*i*	0.1994(3)	–0.0425(2)	0.8703(2)	0.026(1)	0.038(1)	0.032(1)	0.0064(8)	0.0088(8)	0.0004(9)
C(9)	2*i*	0.1737(3)	0.0476(3)	0.6577(2)	0.035(1)	0.057(1)	0.026(1)	0.003(1)	0.0013(9)	0.002(1)
C(11)	2*i*	0.4831(3)	–0.0708(2)	0.6026(2)	0.044(1)	0.043(1)	0.026(1)	0.006(1)	0.0036(9)	0.0093(9)
C(10)	2*i*	0.3456(3)	0.0244(2)	0.5777(2)	0.032(1)	0.038(1)	0.023(1)	0.0013(8)	–0.0003(8)	–0.0007(8)
C(15)	2*i*	0.4173(3)	0.4674(2)	0.3774(2)	0.031(1)	0.029(1)	0.028(1)	0.0002(8)	–0.0043(8)	0.0032(8)
O(4)	2*i*	0.4260(3)	0.3099(2)	0.1911(2)	0.071(1)	0.066(1)	0.0399(9)	0.023(1)	–0.0184(8)	–0.0203(9)
C(17)	2*i*	0.3496(3)	0.4156(2)	0.2427(2)	0.040(1)	0.038(1)	0.033(1)	–0.002(1)	–0.0060(9)	0.0016(9)
O(3)	2*i*	0.2248(3)	0.4816(2)	0.1886(2)	0.069(1)	0.060(1)	0.0393(9)	0.0149(9)	–0.0218(8)	0.0035(8)
C(12)	2*i*	0.3639(3)	0.0953(2)	0.4746(2)	0.038(1)	0.042(1)	0.031(1)	0.0121(9)	0.0007(9)	0.0062(9)
O(2)	2*i*	0.0656(3)	0.6378(2)	0.3410(2)	0.070(1)	0.078(1)	0.053(1)	0.039(1)	–0.023(1)	0.001(1)
C(3)	2*i*	0.2550(3)	0.0176(2)	0.9905(2)	0.026(1)	0.043(1)	0.033(1)	0.0062(9)	0.0068(8)	0.0044(9)
C(13)	2*i*	0.1353(3)	0.6313(2)	0.4527(2)	0.038(1)	0.040(1)	0.047(1)	0.008(1)	–0.010(1)	–0.001(1)
C(2)	2*i*	0.2879(3)	0.1793(2)	0.8645(2)	0.026(1)	0.039(1)	0.036(1)	0.0008(8)	0.0026(8)	0.0009(9)
O(1)	2*i*	0.0589(2)	0.6892(2)	0.5441(2)	0.051(1)	0.079(1)	0.058(1)	0.0306(9)	–0.0072(9)	–0.014(1)
C(4)	2*i*	0.2491(3)	–0.0576(3)	1.0923(2)	0.036(1)	0.073(2)	0.037(1)	0.012(1)	0.007(1)	0.016(1)
C(7)	2*i*	0.1339(3)	–0.1812(2)	0.8457(2)	0.035(1)	0.041(1)	0.059(2)	0.003(1)	0.011(1)	–0.006(1)
C(6)	2*i*	0.1302(4)	–0.2546(3)	0.9470(3)	0.045(2)	0.042(1)	0.088(2)	0.005(1)	0.017(1)	0.018(1)
C(1)	2*i*	0.3337(4)	0.3135(3)	0.8184(3)	0.049(2)	0.047(1)	0.068(2)	–0.004(1)	–0.006(1)	0.017(1)
C(5)	2*i*	0.1864(4)	–0.1946(3)	1.0676(3)	0.044(2)	0.068(2)	0.072(2)	0.013(1)	0.016(1)	0.039(2)

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