Co-Crystal structure of 1,3-phenylenediacetic acid and 2,5-bis(4-pyridyl)-1,3,4-oxadiazole (1:1), $C_{22}H_{18}N_4O_5$

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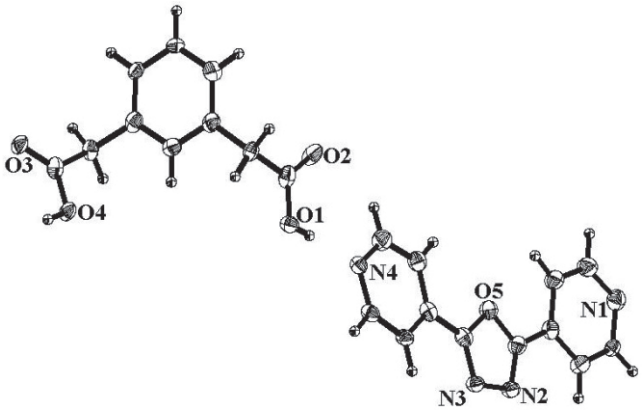

Abstract

$C_{22}H_{18}N_4O_5$, monoclinic, $P2_1/c$ (no. 14), a = 17.153(1) Å, b = 13.4635(8) Å, c = 8.8010(6) Å, β = 90.807(2)°, V = 2032.3 Å^3, Z = 4, $R_{gt}(F)$ = 0.0414, $wR_{ref}(F^2)$ = 0.1148, T = 296 K.

Table 1. Data collection and handling.

Crystal:	colourless blocks, size 0.18×0.25×0.29 mm
Wavelength:	Mo K_α radiation (0.71073 Å)
μ:	0.99 cm^{-1}
Diffractometer, scan mode:	CCD area detector, φ and ω
$2\theta_{max}$:	50.54°
$N(hkl)_{measured}$, $N(hkl)_{unique}$:	13030, 3663
Criterion for I_{obs}, $N(hkl)_{gt}$:	$I_{obs} > 2\ \sigma(I_{obs})$, 2457
$N(param)_{refined}$:	283
Programs:	SHELX [10]

Source of material

The mixtures of 1,3-phenylenediacetic acid (0.1 mmol, 19.4 mg), 2,5-bis(4-pyridyl)-1,3,4-oxadiazole (0.1 mmol, 22.4 mg), NaOH (0.1 mmol), and H_2O (6.0 ml) were placed in a 23ml Teflon lined stainless steel reactor. The vessel was heated to 393K for 4 days, and then cooled at 5°C h^{-1} to room temperature. Colourless crystals were obtained, and further crystals were filtered off, washed with mother liquid, and dried under ambient conditions. Yield 56 %.

Discussion

Cocrystals represent a class of compounds that are attracting increasing attention because they are readily accessible, amenable to crystal engineering, and offer great diversity in terms of composition and properties [1–4]. These components or cocrystal formers coexist as a stoichiometric ratio of a target molecule or ion and a neutral molecular cocrystal former(s). That there has been a recent surge in the interest in cocrystals can be attributed to both scientific and practical matters: crystal engineering has made cocrystals amenable to design (i.e., through CSD statistics and supramolecular synthons) in a way that other crystal forms such as polymorphs, hydrates, and solvates [5] are not; pharmaceutical scientists realized that physicochemical properties of practical importance such as solubility and stability can be dramatically impacted or even controlled via cocrystal formation. In this contribution, we select the 1,2-phenylenediacetic acid with long-spanning carboxyl groups. Featuring two carboxyl groups in the title molecule may provide hydrogen-bonding interactions which may endow enormous potential for assembling multi-dimensional supramolecular architectures [6, 7]. On the other hand, the introduction of nitrogen heterocyclic compound can alter the electron density in different parts of the molecules due to the prototropy and conjugation between aromatic nitrogen heterocyclic [8, 9]. Formation of the 1:1 cocrystal of 1,3-phenylenediacetic acid and 2,5-bis(4-pyridyl)-1,3,4-oxadiazole, results in the formation zig-zag chains by virtue of O4–H4···N1$^{\#1}$ and O1–H1···N4 hydrogen bonding interactions in the title structure. Their N···O distances are 2.684(2) and 2.693(2) Å, respectively. Strong $\pi\cdots\pi$ stacking interactions (the face to face distance is 3.30 Å) existing both pyridine rings of 2,5-bis(4-pyridyl)-1,3,4-oxadiazole ligands from two adjacent zig-zag chains, result in a 2D thick layer. The adjacent 2D layers stack in a slightly off-set parallel fashion with the weak effect of van der Waals force. Clearly, hydrogen bonds and $\pi\cdots\pi$ stacking interactions play a vital role in the formation of 3D supramolecular structure.

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

Atom	Site	x	y	z	U_{iso}
H(8)	4*e*	0.0114	0.6680	0.7007	0.058
H(14)	4*e*	0.6078	0.5908	0.0886	0.066
H(4)	4*e*	–0.1868	0.5954	0.7266	0.107
H(1)	4*e*	0.2220	0.5467	0.7051	0.118
H(19)	4*e*	0.4350	0.5818	0.3552	0.064
H(12)	4*e*	0.6701	0.3208	–0.0585	0.064
H(11)	4*e*	0.7650	0.3947	–0.1987	0.068
H(22)	4*e*	0.3837	0.3015	0.4686	0.078
H(21)	4*e*	0.2895	0.3616	0.6245	0.085
H(15)	4*e*	0.7033	0.6565	–0.0615	0.070
H(20)	4*e*	0.3376	0.6328	0.5132	0.073
H(2A)	4*e*	0.1547	0.7244	0.9807	0.076
H(2B)	4*e*	0.0994	0.6370	0.9317	0.076
H(9A)	4*e*	–0.1268	0.8168	0.5004	0.080
H(9B)	4*e*	–0.0837	0.7145	0.4849	0.080
H(4A)	4*e*	0.1174	0.8902	0.9190	0.087
H(6)	4*e*	–0.0677	0.9440	0.6514	0.097
H(5)	4*e*	0.0303	0.9992	0.8106	0.115

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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}
O(5)	4*e*	0.52348(7)	0.46250(9)	0.2129(1)	0.0461(7)	0.0515(7)	0.0513(8)	–0.0020(6)	0.0080(6)	0.0006(6)
C(18)	4*e*	0.4196(1)	0.4347(1)	0.3910(2)	0.043(1)	0.058(1)	0.044(1)	–0.0086(9)	0.0009(9)	–0.0009(9)
C(17)	4*e*	0.4779(1)	0.3969(1)	0.2879(2)	0.045(1)	0.053(1)	0.050(1)	–0.0079(9)	0.0031(9)	0.0022(9)
C(8)	4*e*	0.0147(1)	0.7353(1)	0.7241(2)	0.051(1)	0.044(1)	0.051(1)	0.0017(9)	0.0162(9)	0.0038(8)
N(1)	4*e*	0.74313(9)	0.5324(1)	–0.1445(2)	0.049(1)	0.061(1)	0.057(1)	0.0022(8)	0.0057(8)	0.0052(8)
C(13)	4*e*	0.6283(1)	0.4477(1)	0.0325(2)	0.043(1)	0.054(1)	0.045(1)	–0.0004(9)	–0.0006(9)	0.0030(9)
C(14)	4*e*	0.6389(1)	0.5494(1)	0.0305(2)	0.052(1)	0.054(1)	0.058(1)	0.0028(9)	0.010(1)	–0.0050(9)
O(4)	4*e*	–0.15212(8)	0.6189(1)	0.6745(2)	0.0579(9)	0.070(1)	0.088(1)	0.0095(7)	0.0261(8)	0.0115(8)
O(1)	4*e*	0.18883(9)	0.5618(1)	0.7671(2)	0.069(1)	0.077(1)	0.091(1)	–0.0145(8)	0.0353(8)	–0.0167(9)
O(3)	4*e*	–0.22218(8)	0.7547(1)	0.6991(2)	0.0587(9)	0.073(1)	0.098(1)	0.0114(8)	0.0275(9)	0.0041(8)
C(16)	4*e*	0.5695(1)	0.4023(1)	0.1282(2)	0.048(1)	0.053(1)	0.048(1)	0.0025(9)	0.0041(9)	–0.0017(9)
C(19)	4*e*	0.4057(1)	0.5353(1)	0.4077(2)	0.050(1)	0.057(1)	0.054(1)	–0.0051(9)	0.003(1)	0.0041(9)
N(4)	4*e*	0.3044(1)	0.5026(1)	0.5839(2)	0.059(1)	0.077(1)	0.059(1)	–0.0080(9)	0.0123(9)	–0.0105(9)
C(10)	4*e*	–0.1662(1)	0.7129(2)	0.6497(2)	0.041(1)	0.067(1)	0.048(1)	0.002(1)	0.0034(9)	0.005(1)
N(2)	4*e*	0.5538(1)	0.3095(1)	0.1479(2)	0.071(1)	0.055(1)	0.074(1)	–0.0039(9)	0.023(1)	–0.0009(9)
C(7)	4*e*	–0.0390(1)	0.7996(1)	0.6589(2)	0.041(1)	0.058(1)	0.059(1)	0.0017(9)	0.0169(9)	0.0149(9)
C(12)	4*e*	0.6761(1)	0.3894(1)	–0.0563(2)	0.056(1)	0.049(1)	0.056(1)	0.0058(9)	0.008(1)	0.0017(9)
N(3)	4*e*	0.4933(1)	0.3062(1)	0.2532(2)	0.069(1)	0.055(1)	0.073(1)	–0.0065(9)	0.021(1)	–0.0022(9)
C(3)	4*e*	0.0730(1)	0.7669(1)	0.8227(2)	0.041(1)	0.055(1)	0.058(1)	0.0025(9)	0.0185(9)	0.0016(9)
C(1)	4*e*	0.1886(1)	0.6574(2)	0.7858(2)	0.040(1)	0.067(1)	0.059(1)	0.008(1)	0.0077(9)	0.018(1)
C(11)	4*e*	0.7325(1)	0.4345(1)	–0.1409(2)	0.055(1)	0.060(1)	0.057(1)	0.010(1)	0.011(1)	0.005(1)
C(22)	4*e*	0.3753(1)	0.3697(2)	0.4753(2)	0.069(1)	0.058(1)	0.070(2)	–0.014(1)	0.021(1)	–0.004(1)
C(21)	4*e*	0.3190(1)	0.4064(2)	0.5688(3)	0.071(2)	0.073(2)	0.070(2)	–0.022(1)	0.024(1)	–0.007(1)
O(2)	4*e*	0.2338(1)	0.7108(1)	0.7235(2)	0.085(1)	0.072(1)	0.147(2)	0.0158(9)	0.065(1)	0.038(1)
C(15)	4*e*	0.6966(1)	0.5880(2)	–0.0598(2)	0.059(1)	0.051(1)	0.066(1)	–0.003(1)	0.010(1)	–0.001(1)
C(20)	4*e*	0.3474(1)	0.5652(2)	0.5039(2)	0.058(1)	0.063(1)	0.062(1)	0.002(1)	0.003(1)	–0.004(1)
C(2)	4*e*	0.1287(1)	0.6934(2)	0.8944(2)	0.053(1)	0.080(1)	0.056(1)	0.011(1)	0.011(1)	0.009(1)
C(9)	4*e*	–0.1043(1)	0.7618(2)	0.5571(2)	0.054(1)	0.089(2)	0.056(1)	0.001(1)	0.008(1)	0.019(1)
C(4)	4*e*	0.0783(1)	0.8668(2)	0.8539(3)	0.052(1)	0.063(1)	0.104(2)	–0.005(1)	0.008(1)	–0.012(1)
C(6)	4*e*	–0.0324(1)	0.8989(2)	0.6933(3)	0.053(1)	0.054(1)	0.137(2)	0.011(1)	0.011(1)	0.020(1)
C(5)	4*e*	0.0261(1)	0.9317(2)	0.7893(4)	0.071(2)	0.043(1)	0.174(3)	0.000(1)	0.007(2)	–0.007(2)

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