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3,4-(Methylenedioxy)benzaldehyde benzoylhydrazone ethanol solvate

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Key indicators

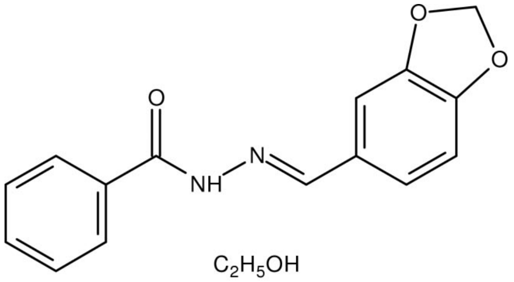
Single-crystal X-ray study
T = 297 K
Mean σ(C–C) = 0.004 Å
R factor = 0.045
wR factor = 0.136
Data-to-parameter ratio = 9.8

For details of how these key indicators were automatically derived from the article, see http://journals.iucr.org/e.

The title compound, $C_{15}H_{12}N_2O_3 \cdot C_2H_6O$, was synthesized by the condensation of 3,4-(methylenedioxy)benzaldehyde with benzoylhydrazine. The molecule deviates more from planarity than is usual for hydrazones. In the crystal structure, hydrogen bonds link 3,4-(methylenedioxy)benzaldehyde benzoylhydrazone molecules to ethanol solvent molecules; aromatic stacking interactions are also found.

Comment

Various benzoylhydrazones derived from arylaldehydes, phenylalkyl aldehydes and phenylalkyl ketones, as well as some related compounds, have been evaluated for anticonvulsant activity (Katyal & Dutt, 1975). In order to study the relationship between the structure and biological activities of these compounds, several hydrazone Schiff base compounds have been synthesized (He *et al.*, 2002; He, Yang *et al.*, 2003; He, Chen *et al.*, 2003). We report here the synthesis and crystal structure of 3,4-(methylenedioxy)benzaldehyde benzoylhydrazone, obtained by the condensation of 3,4-(methylenedioxy)benzaldehyde with benzoylhydrazine as an ethanol solvate.



(I)

The molecular structure is shown in Fig. 1 and the crystal packing in Fig. 2. In the 3,4-(methylenedioxy)benzaldehyde benzoylhydrazone molecule, the dihedral angles between the 3,4-methylenedioxybenzene group and the benzoyl and hydrazone group planes are 28.66 (3) and 23.89 (3)°, respectively; these angles are much larger than the values of 6.04 (3) and 6.84 (3)° for 3,4-(methylenedioxy)benzaldehyde 2,4-dinitrophenylhydrazone (Wang & Jia, 2004), showing that the molecule of the title compound deviates markedly from planarity.

The plane defined by atoms C11–C16, O10, O20 and C10 in the 3,4-methylenedioxybenzene group has a maximum deviation of 0.0303 (19) Å for atom C10. The O10—C10 and O20—C10 bond lengths are 1.418 (3) and 1.424 (3) Å, respectively, longer than the adjacent O10—C14 and O20—

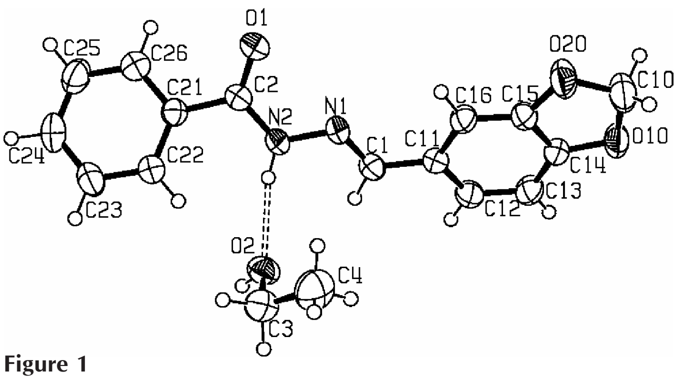


Figure 1
The structure of the asymmetric unit, with displacement ellipsoids drawn at the 50% probability level. The dashed lines represent a hydrogen bond.

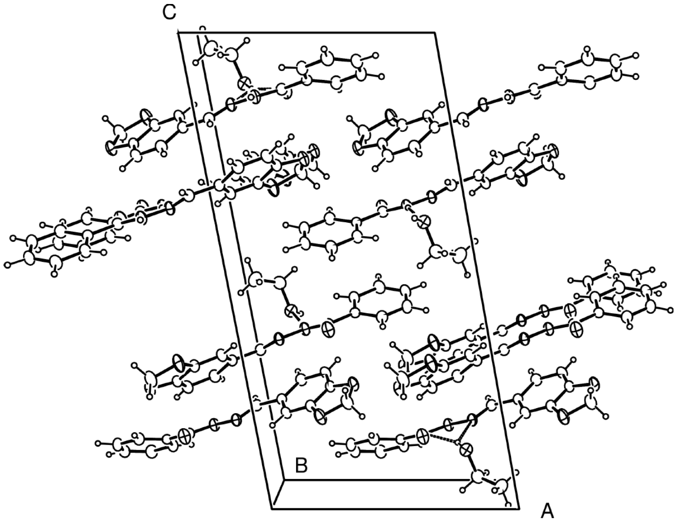


Figure 2
The crystal packing, viewed approximately along the *b* axis.

C15 bonds [1.373 (3) and 1.374 (3) Å]. This is similar to the situation in 3,4-(methylenedioxy)benzaldehyde semicarbazone (Wang, Jia & Yu, 2004). The N1—C1 bond length is 1.277 (3) Å, which is close to the value of 1.280 (5) Å found for the imine bond length in *p*-dimethylenedioxybenzaldehyde 2,4-dinitrobenzoylhydrazone (Wang, Jia, Miao & Li, 2004) and shorter than the value of 1.337 (2) Å found for the C—N single bond in the 1:1 complex of 1-phenyl-3-methyl-4-benzoyl-5-pyrazolone and nicotinoylhydrazine (Liu *et al.*, 2001). The N1—N2 bond length is 1.385 (3) Å, which is close to 1.3794 (19) and 1.388 (2) Å in *p*-dimethylenedioxybenzaldehyde benzoylhydrazone (Fun *et al.*, 1997), indicating that a partially conjugated system operates in this hydrazone. The C2—O1, N2—C2 and C2—C21 bond lengths in the amide group are 1.228 (3), 1.347 (3) and 1.488 (3) Å, respectively, close to the values of 1.241 (2), 1.334 (2) and 1.462 (2) Å found for 3,4-(methylenedioxy)benzaldehyde semicarbazone.

The distances between adjacent 3,4-methylenedioxybenzene planes and between phenyl planes are 3.163 (5) and 3.567 (5) Å, respectively, indicating that aromatic stacking interactions are present. This is similar to the situation in compounds such as salicylaldehyde 4-nitrophenylhydrazone (Shan *et al.*, 2003). In addition, hydrogen bonds are observed between 3,4-(methylenedioxy)benzaldehyde benzoylhydrazone molecules and ethanol molecules (Table 1 and Fig. 2).

Experimental

Benzoylhydrazine (0.01 mol) in anhydrous ethanol (20 ml) was slowly added to 3,4-(methylenedioxy)benzaldehyde (0.01 mol) in anhydrous ethanol (20 ml). The solution was stirred and refluxed for about 2 h at 343–353 K. When the solution was cooled to room temperature, some white needles separated out. These were filtered off, washed several times with cold anhydrous ethanol and dried in a vacuum over $CaCl_2$. Analysis found: C 67.26, H 4.46, N 10.52%; calculated for $C_{15}H_{12}N_2O_3$: C 67.16, H 4.51, N 10.44%. M.p. 455 K. A few crystals were dissolved in anhydrous ethanol (10 ml). Colourless single crystals suitable for X-ray analysis were obtained by slow evaporation at room temperature after 3 d.

Crystal data

$C_{15}H_{12}N_2O_3{\cdot}C_2H_6O$
$M_r = 314.33$
Monoclinic, $P2_1/c$
$a = 11.163\ (4)$ Å
$b = 6.651\ (2)$ Å
$c = 21.519\ (7)$ Å
$\beta = 100.015\ (6)^\circ$
$V = 1573.3\ (9)$ Å^3
$Z = 4$
$D_x = 1.327$ Mg m^{-3}
Mo $K\alpha$ radiation
Cell parameters from 881 reflections
$\theta = 3.2$–22.7°
$\mu = 0.10$ mm^{-1}
$T = 297\ (2)$ K
Block, colourless
0.22 × 0.18 × 0.14 mm

Data collection

Bruker SMART 1000 CCD area-detector diffractometer
φ and ω scans
Absorption correction: none
7720 measured reflections
2762 independent reflections
1746 reflections with $I > 2\sigma(I)$
$R_{\rm int} = 0.039$
$\theta_{\rm max} = 25.0^\circ$
$h = -11 \rightarrow 13$
$k = -7 \rightarrow 5$
$l = -25 \rightarrow 25$

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)] = 0.045$
$wR(F^2) = 0.136$
$S = 1.06$
2762 reflections
281 parameters
All H-atom parameters refined
$w = 1/[\sigma^2(F_o^2) + (0.0713P)^2]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{\rm max} = 0.001$
$\Delta\rho_{\rm max} = 0.19$ e Å^{-3}
$\Delta\rho_{\rm min} = -0.17$ e Å^{-3}
Extinction correction: *SHELXL97*
Extinction coefficient: 0.0047 (16)

Table 1
Hydrogen-bond geometry (Å, °).

D—H···A	D—H	H···A	D···A	D—H···A
N2—H2···O2	0.88 (2)	2.11 (2)	2.959 (3)	159 (2)
O2—H02···N1$^{\rm i}$	0.81 (4)	2.64 (4)	3.265 (3)	135 (3)
O2—H02···O1$^{\rm i}$	0.81 (4)	2.09 (4)	2.844 (3)	155 (4)

Symmetry code: (i) $x, y + 1, z$.

All H atoms were located in difference Fourier maps and refined isotropically. C—H bond lengths are in the range 0.91 (3)–1.02 (3) Å.

Data collection: *SMART* (Bruker, 1999); cell refinement: *SAINT* (Bruker, 1999); data reduction: *SAINT*; program(s) used to solve structure: *SHELXS97* (Sheldrick, 1997); program(s) used to refine structure: *SHELXL97* (Sheldrick, 1997); molecular graphics: *ORTEP-3* for Windows (Farrugia, 1997); software used to prepare material for publication: *WinGX* (Farrugia, 1999).

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