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(*E*,*E*)-4-Hydroxy-3-methoxybenzaldehyde azine

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Key indicators

Single-crystal X-ray study
T = 292 K
Mean σ(C–C) = 0.002 Å
R factor = 0.042
wR factor = 0.117
Data-to-parameter ratio = 14.3

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

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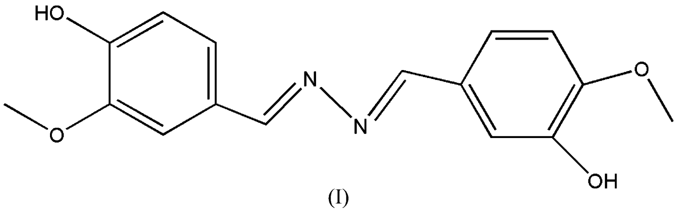
The title compound, $C_{16}H_{16}N_2O_4$, contains two vanillin aromatic rings, which are bridged by a C═N—N═C unit. There are two half-molecules in the asymmetric unit; the mid-points of the N—N bonds lie on centres of symmetry. The molecular structure is stabilized by two intramolecular O—H···O and intermolecular O—H···O and O—H···N hydrogen bonds.

Comment

Much work has been devoted to the physicochemical characterization of substituted aromatic Schiff bases, because these compounds show remarkable photochromic properties. Photochromism arises from intramolecular H-atom transfer, together with a change in the π-electron system. The effect of intermolecular interactions, such as π–π charge transfer or hydrogen bonding, on H-atom transfer processes has been investigated in the solid state (Hadjoudis *et al.*, 1987; Puranik *et al.*, 1992).


(I)

The hydrogen bond is important in a vast number of chemical, biological and materials systems (Steiner & Saenger, 1993). The weak hydrogen bond of the C—H···X type (X = O, N and π acceptors) has been well established in structural, supramolecular and biological chemistry, and it has been widely used as a tool for the crystal engineering of organic and organometallic solids (Desiraju, 1996; Braga & Grepioni, 2000). In the literature, there are only a few structural reports of Schiff bases derived from vanillin (4-hydroxy-3-methoxybenzaldehyde) (Kaitner & Pavlovic, 1995; Lin *et al.*, 2005).

The crystal structure of the title compound, (I) (Fig. 1), has two half-molecules in the asymmetric unit; the mid-points of the N—N bonds lie on centres of symmetry. The molecules contain two vanillin aromatic rings, which are bridged by C═N—N═C units. As expected, the non-H atoms are nearly coplanar, forming an extended conjugated system. The molecules crystallize in the (E, E) configuration, with the two 4-hydroxy-3-methoxyphenyl groups *trans* to each other. This configuration agrees with those commonly found in similar compounds (Glaser *et al.*, 1995; Hunig *et al.*, 2000).

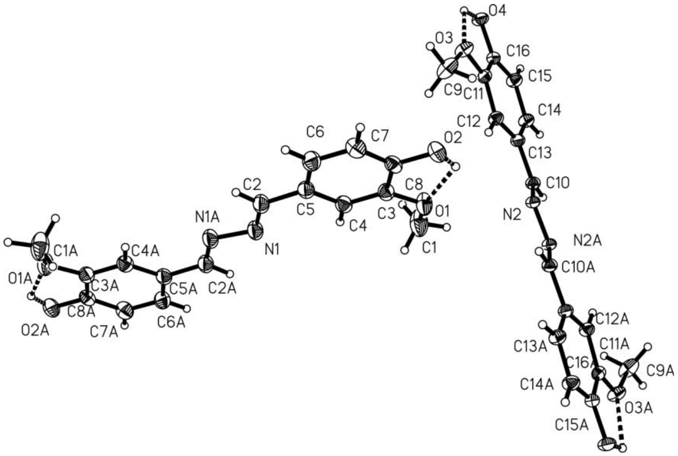


Figure 1
A drawing of (I), with the atom-numbering scheme. Displacement ellipsoids are drawn at the 50% probability level. Dashed lines indicate intramolecular hydrogen bonds. [Symmetry codes: (A) $2 - x, 2 - y, 1 - z$ (left-hand molecule); $1 - x, -y, 1 - z$ (right-hand molecule).]

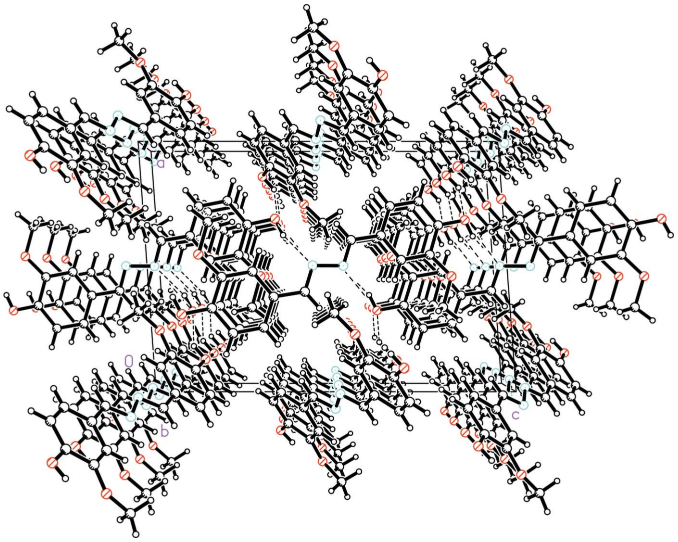


Figure 2
A packing diagram of (I); O—H···O and N—H···O hydrogen bonds are shown as dashed lines.

The bond lengths and angles (Table 1) are within the expected ranges (Allen *et al.*, 1987). The molecules are stacked along the *b* axis in a parallel fashion (Fig. 2). The centroid-to-centroid and perpendicular distances between the aromatic rings are 4.0886 (9) and 3.503 Å, respectively, which may indicate weak π–π interactions. The molecular structure is stabilized by two intramolecular O—H···O and intermolecular O—H···O and O—H···N hydrogen bonds (Table 2), which cause the formation of an infinite two-dimensional network by linking the neighbouring molecules in the *ac* plane (Fig. 2).

Experimental

The title compound was prepared by the condensation reaction of vanillin (1.52 g) in ethanol (20 ml) and hydrazine hydrochloride (0.34 g) in ethanol (20 ml). The reaction mixture was refluxed and stirred for 2 h. The resulting clear solution was kept in air and, after slow evaporation of the solvent over a period of a week, yellow crystals were formed at the bottom of the vessel. The crystals were isolated, washed three times with ethanol and dried in a vacuum desiccator using anhydrous $CaCl_2$ (yield 68%). Analysis calculated for $C_{16}H_{16}N_2O_4$: C 63.93, H 5.37, N 9.33%; found: C 63.66, H 5.48, N 9.31%.

Crystal data

$C_{16}H_{16}N_2O_4$
$M_r = 300.31$
Monoclinic, $P2_1/c$
$a = 11.6941$ (8) Å
$b = 7.5492$ (5) Å
$c = 16.7683$ (12) Å
$\beta = 94.169$ (1)°
$V = 1476.41$ (18) Å^3
$Z = 4$
$D_x = 1.351$ Mg m^{-3}
Mo $K\alpha$ radiation
Cell parameters from 3000 reflections
$\theta = 2.0$–$28.4°$
$\mu = 0.10$ mm^{-1}
$T = 292$ (2) K
Block, yellow
0.36 × 0.33 × 0.30 mm

Data collection

Bruker SMART CCD area-detector diffractometer
φ and ω scans
Absorption correction: multi-scan (*SADABS*; Sheldrick, 1996)
$T_{\min} = 0.971$, $T_{\max} = 0.971$
12220 measured reflections
2905 independent reflections
2573 reflections with $I > 2\sigma(I)$
$R_{\rm int} = 0.047$
$\theta_{\max} = 26.0°$
$h = -14 \rightarrow 14$
$k = -8 \rightarrow 9$
$l = -20 \rightarrow 20$

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)] = 0.042$
$wR(F^2) = 0.117$
$S = 1.06$
2905 reflections
203 parameters
H-atom parameters constrained
$w = 1/[\sigma^2(F_o^2) + (0.0725P)^2 + 0.1651P]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{\max} < 0.001$
$\Delta\rho_{\max} = 0.18$ e Å^{-3}
$\Delta\rho_{\min} = -0.22$ e Å^{-3}

Table 1
Selected geometric parameters (Å, °).

C2—N1	1.274 (2)	C10—C13	1.4572 (15)
C2—C5	1.463 (2)	N1—N1$^{\rm i}$	1.413 (2)
C10—N2	1.2817 (16)	N2—N2$^{\rm ii}$	1.4094 (17)
N1—C2—C5	122.71 (14)	C2—N1—N1$^{\rm i}$	111.66 (17)
N2—C10—C13	122.03 (10)	C10—N2—N2$^{\rm ii}$	112.95 (11)
N1—C2—C5—C6	−175.98 (14)	N2—C10—C13—C14	167.39 (11)
N1—C2—C5—C4	2.7 (2)	N2—C10—C13—C12	−9.42 (18)
O1—C3—C8—O2	−0.65 (18)	O3—C11—C16—O4	−4.44 (17)
C4—C3—C8—O2	178.27 (12)	C12—C11—C16—O4	175.05 (11)
O1—C3—C8—C7	178.43 (12)	C5—C2—N1—N1$^{\rm i}$	178.95 (13)
O3—C11—C12—C13	−177.08 (11)	C13—C10—N2—N2$^{\rm ii}$	179.90 (11)

Symmetry codes: (i) $-x + 2, -y + 2, -z + 1$; (ii) $-x + 1, -y, -z + 1$.

Table 2
Hydrogen-bond geometry (Å, °).

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
O2—H2*A*···O1	0.82	2.22	2.6658 (14)	114
O4—H4*A*···O3	0.82	2.24	2.6731 (12)	114
O2—H2*A*···O4$^{\rm iii}$	0.82	2.34	2.9973 (14)	138
O4—H4*A*···N2$^{\rm iv}$	0.82	2.05	2.8346 (12)	159

Symmetry codes: (iii) $-x + 1, y - \frac{1}{2}, -z + \frac{1}{2}$; (iv) $-x + 1, y + \frac{1}{2}, -z + \frac{1}{2}$.

Hydroxy H atoms were located in a difference map, while the remaining H atoms were positioned geometrically [0.82 (OH), 0.93 (CH) and 0.96 Å (CH_3)] and constrained to ride on their parent atoms with U_{iso}(H) values of 1.2 (1.5 for methyl and hydroxy H atoms) times U_{eq}(C,O).

Data collection: *SMART* (Siemens, 1996); cell refinement: *SAINT* (Siemens, 1996); data reduction: *SAINT*; program(s) used to solve structure: *SHELXS97* (Sheldrick, 1997*a*); program(s) used to refine structure: *SHELXL97* (Sheldrick, 1997*a*); molecular graphics: *SHELXTL* (Sheldrick, 1997*b*); software used to prepare material for publication: *SHELXTL*.

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