N,N'-Bis(3-hydroxy-4-methoxybenzylidene)-hydrazine

Xue-Min Duan,[a] Peng-Wu Zheng[a]* and Bin Zhou[b]

[a]School of Pharmacy, Jiangxi Science and Technology Normal University, Nanchang 330013, People's Republic of China, and [b]College of Pharmaceuticals and Biotechnology, Tianjin University, Tianjin 300072, People's Republic of China

Correspondence e-mail: zhengpw@sohu.com

The title compound, $C_{16}H_{16}N_2O_4$, was synthesized by the reaction of 3-hydroxy-4-methoxybenzaldehyde with hydrazine hydrate. The molecule possesses a crystallographically imposed centre of symmetry. An intramolecular O—H···O hydrogen bond promotes planarity of the molecular backbone. In the crystal structure, intermolecular O—H···N and C—H···O hydrogen bonds stabilize the crystal packing.

Comment

Recently, a number of azine compounds containing both a diimine linkage and N—N bonding have been investigated in terms of their crystallography and coordination chemistry (Zheng *et al.*, 2005; Kundu *et al.*, 2005; Kesslen & Euler, 1999; Armstrong *et al.*, 1998; Xu *et al.*, 1997). We report here the crystal structure of the title compound, (I), where two 3-hydroxy-4-methoxybenzylidene units are directly linked through the imine N atoms.

(I)
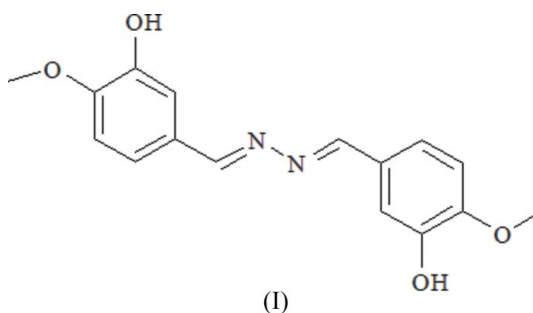
The molecule of (I) possesses a crystallographically imposed centre of symmetry at the mid-point of the N—N bond (Fig. 1). The N—N bond length of 1.412 (2) Å (Table 1) is somewhat longer than that observed in related azine compounds (Xu *et al.*, 2005; Liu *et al.*, 2004; Şengül *et al.*, 2004).
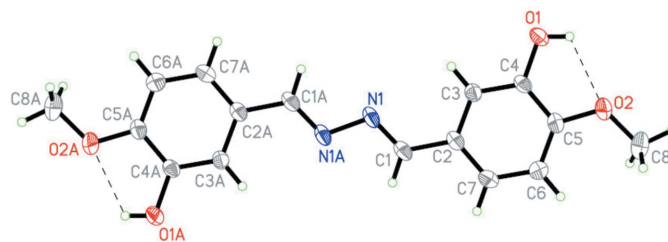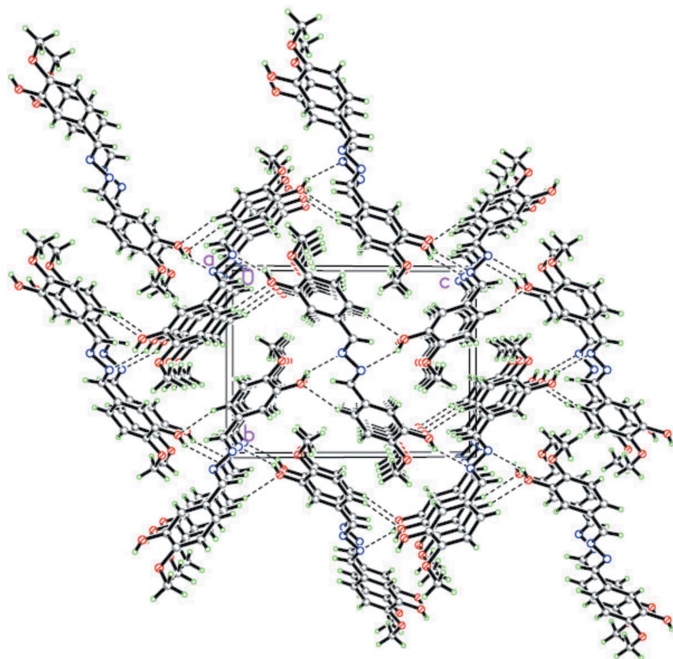

Figure 1
A view of (I), showing the intramolecular hydrogen bonds (dashed lines) and with displacement ellipsoids at the 30% probability level. The suffix A corresponds to symmetry operation ($-1 - x, 1 - y, 1 - z$).

Figure 2
The crystal packing of (I), viewed approximately down the *a* axis, showing the intermolecular hydrogen-bonded (dashed lines) extended network.

The C=N—N angle [112.82 (15)°] is similar to that in N,N'-bis(4-chlorobenzylidene)hydrazine (Zheng *et al.*, 2005) but significantly smaller than the ideal sp^2 value of 120°, as a consequence of repulsion between the nitrogen lone pairs and the adjacent C=N bond. An intramolecular O—H···O hydrogen bond (Table 2) provides planarity of the molecular backbone, with an r.m.s deviation of 0.034 (6) Å.

In the crystal structure, intermolecular O—H···N and C—H···O hydrogen bonds (Table 2) stabilize the crystal packing (Fig. 2).

Experimental

The title compound was synthesized by the reaction of 3-hydroxy-4-methoxybenzaldehyde with hydrazine hydrate in refluxing ethanol (Liu *et al.*, 2004). Single crystals suitable for X-ray analysis were obtained by slow evaporation of a tetrahydrofuran solution.

Crystal data

$C_{16}H_{16}N_2O_4$	$D_x = 1.339$ Mg m^{-3}
$M_r = 300.31$	Mo $K\alpha$ radiation
Monoclinic, $P2_1/c$	Cell parameters from 1595
$a = 5.6858$ (12) Å	reflections
$b = 10.018$ (2) Å	$\theta = 2.6$–26.1°
$c = 13.902$ (3) Å	$\mu = 0.10$ mm^{-1}
$\beta = 109.841$ (4)°	$T = 294$ (2) K
$V = 744.8$ (3) Å^3	Block, light yellow
$Z = 2$	$0.30 \times 0.22 \times 0.16$ mm

Data collection

Bruker SMART CCD area-detector diffractometer	1523 independent reflections
	1065 reflections with $I > 2\sigma(I)$
φ and ω scans	$R_{int} = 0.026$
Absorption correction: multi-scan (*SADABS*; Sheldrick, 1996)	$\theta_{max} = 26.4°$
$T_{min} = 0.946$, $T_{max} = 0.980$	$h = -5 \rightarrow 7$
4096 measured reflections	$k = -12 \rightarrow 10$
	$l = -17 \rightarrow 15$

Refinement

Refinement on F^2	$w = 1/[\sigma^2(F_o^2) + (0.0456P)^2$
$R[F^2 > 2\sigma(F^2)] = 0.037$	$+ 0.1273P]$
$wR(F^2) = 0.103$	where $P = (F_o^2 + 2F_c^2)/3$
$S = 1.03$	$(\Delta/\sigma)_{max} < 0.001$
1523 reflections	$\Delta\rho_{max} = 0.12$ e Å^{-3}
111 parameters	$\Delta\rho_{min} = -0.18$ e Å^{-3}
H atoms treated by a mixture of independent and constrained refinement	

Table 1
Selected geometric parameters (Å, °).

O1—C4	1.3659 (17)	N1—C1	1.269 (2)
O2—C5	1.3631 (17)	N1—N1^i	1.413 (2)
O2—C8	1.417 (2)		
C5—O2—C8	118.80 (13)	N1—C1—C2	122.33 (13)
C1—N1—N1^i	112.82 (15)	O1—C4—C3	119.14 (14)
N1^i—N1—C1—C2	177.44 (14)	N1—C1—C2—C3	−9.5 (2)
N1—C1—C2—C7	171.79 (14)		

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

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

D—H···A	D—H	H···A	D···A	D—H···A
O1—H1···O2	0.86 (1)	2.20 (2)	2.6405 (17)	112 (2)
O1—H1···N1^{ii}	0.86 (1)	2.18 (2)	2.9365 (16)	147 (2)
C7—H7···O1^{iii}	0.93	2.49	3.391 (2)	164

Symmetry codes: (ii) $-x, y + \frac{1}{2}, -z + \frac{3}{2}$; (iii) $x, -y + \frac{3}{2}, z - \frac{1}{2}$.

All H atoms were located in a difference Fourier map. C-bound H atoms were refined as riding, with C—H = 0.93–0.96 Å and $U_{iso}(H) = 1.2U_{eq}(C)$. Atom H1 was refined isotropically, with O1—H1 = 0.85 (1) Å.

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

References

Armstrong, J. A., Barnes, J. C. & Weakley, T. J. R. (1998). *Acta Cryst.* C**54**, 1923–1925.
Bruker (1997). *SMART, SAINT* and *SHELXTL*. Bruker AXS Inc., Madison, Wisconsin, USA.
Kesslen, E. C. & Euler, W. B. (1999). *Chem. Mater.* **11**, 336–340.
Kundu, N., Chatterjee, P. B., Chaudhury, M. & Tiekink, E. R. T. (2005). *Acta Cryst.* E**61**, m1583–m1585.
Liu, S. L., Chen, Y., Dai, J. F. & Liu, H. W. (2004). *Chin. J. Synth. Chem.* **12**, 219–221.
Şengül, A., Karadayi, N. & Büyükgüngör, O. (2004). *Acta Cryst.* C**60**, o507–o508.
Sheldrick, G. M. (1996). *SADABS*. University of Göttingen, Germany.

Sheldrick, G. M. (1997). *SHELXS97* and *SHELXL97*. University of Göttingen, Germany.

Xu, L. Z., Xu, H. Z., Yang, S. H., Li, C. L & Zhou, K. (2005). *Acta Cryst.* E**61**, o31–o32.

Xu, Z., Thompson, L. K. & Miller, D. O. (1997). *Inorg. Chem.* **36**, 3985–3995.

Zheng, P.-W., Wang, W. & Duan, X.-M. (2005). *Acta Cryst.* E**61**, o3020–o3021.