

Hydrogen bonding in *meso*-4,5-diphenyl-3,6-diazaoctane-1,8-diol: the formation of one-dimensional linear chains of edge-fused rings

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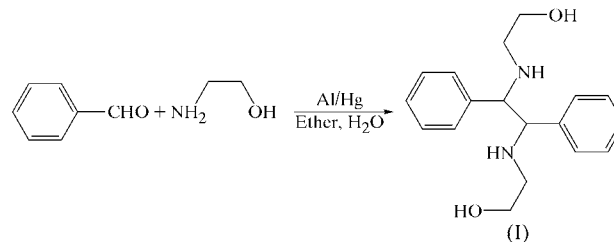
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The molecule of the title compound, C₁₈H₂₄N₂O₂, resides on a crystallographic inversion centre. The molecule adopts a *transoid* conformation with respect to the central C—C single bond and is in the *meso* form. A polarimetric study of the compound did not show any optical activity, indicating that the compound is a racemic mixture entirely consistent with the centrosymmetric *C2/c* space group. In the molecule, there is one intramolecular N—H···O interaction, resulting in the formation of a five-membered ring. In the crystal structure, intermolecular O—H···N and C—H···O interactions are also observed. These interactions form an *R*₂²(9) ring and one-dimensional linear chains of edge-fused rings running parallel to the [010] direction, which stabilize the crystal packing.

Comment

The generation of different types of packing motifs in crystalline lattices has been illustrated in detail in the literature. Hydrogen-bonding motifs play an important role in the interaction, recognition and conformation of both small and large molecules, *e.g.* in biologically active molecules (Watson & Crick, 1953; Zeng *et al.*, 2000). These motifs have frequently been used as supramolecular synthons to direct solid-state structures (crystal engineering; Steiner, 2002; Aakerøy & Seddon, 1993) and for the synthesis of complex supramolecules. Molecular assembly in a crystal is predominantly governed by intermolecular forces, conventionally described by strong and directional N—H···O, O—H···O and O—H···N hydrogen bonds (Desiraju, 2002). In molecules lacking these hydrogen-bond donors and acceptors, other types of weak and less directional forces, such as C—H···O, C—H···π and π—π interactions, become important in generating supramolecular architectures (Desiraju & Steiner, 1999; Hunder & Sanders, 1990; Nishio *et al.*, 1998; Umezawa *et al.*, 1998;

Calhorda, 2000). The title compound, (I), is the intermediate product obtained during the synthesis of macromolecular metal complexes, which is an ongoing project in our laboratory. We present here the complete geometric characterization of (I) in the solid state, together with an analysis of the intermolecular interactions in the crystal structure.



The structure of (I), together with the atom-labelling scheme, is shown in Fig. 1. Both phenyl and 2-hydroxyethylamino moieties adopt a *transoid* configuration with respect to the C7—C7ⁱ bond [symmetry code: (i) $-x + \frac{1}{2}, -y + \frac{1}{2}, -z$], which minimizes the steric hindrance in the molecule. The compound crystallizes in space group *C2/c* and the molecules reside on inversion centres located at the mid-point of the C7—C7ⁱ bond. Selected bond lengths and angles are listed in Table 1. The N1—C7 and N1—C8 imine bond lengths are 1.478 (2) and 1.473 (2) Å, respectively, and are in the accepted range for covalent single bonds (Allen *et al.*, 1987). The Φ_{NC} torsion angle (C7—N1—C8—C9) is 171.2 (2)°, which shows that the conformation about the N1—C8 bond is (+)anti-periplanar. However, the Φ_{CC} torsion angle (N1—C8—C9—O1) is -54.5 (2)°, indicating that the conformation about the C8—C9 bond is (-)synclinal. As can be seen from these torsion angles, the C7—N1—C8—C9 backbone is in a zigzag chain form. However, the hydroxy O atom does not lie in the plane of this chain and is oriented towards the imine N atom. This orientation leads to an intramolecular N—H···O hydrogen bond, resulting in the formation of a five-membered ring.

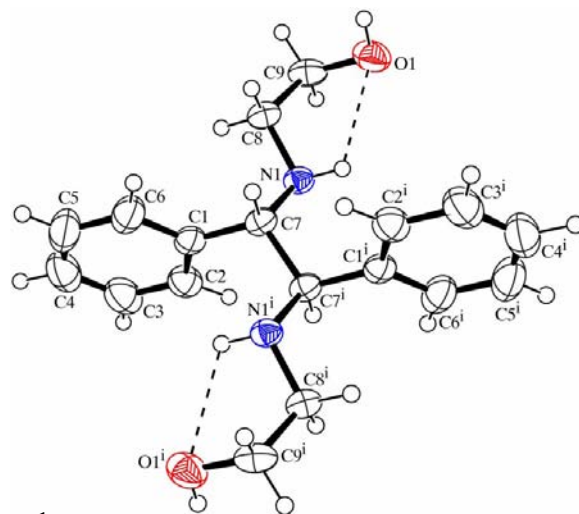


Figure 1
The molecule of (I), showing the atom-labelling scheme and 50% probability displacement ellipsoids. The intramolecular N—H···O hydrogen bonds are represented by dashed lines. [Symmetry code: (i) $-x + \frac{1}{2}, -y + \frac{1}{2}, -z$.]

In the crystal lattice of (I), the 1,2-diphenylethane part of the molecule resides along the *a* axis of the unit cell, while two symmetry-related 2-(methylamino)ethanol parts of the molecule lie along the *c* axis of the unit cell. In this arrangement, the molecules are connected to each other by means of hydrogen bonds between 2-hydroxyethylamino tails. There are no intermolecular interactions between molecules in the *a* or *c* direction. In the construction of this connection, two 2-hydroxyethylamino tails of the 2_1 screw symmetry-related molecules, which interdigitate with the 2-hydroxyethylamino tails of two neighbouring molecules translated linearly along the *b* axis of the unit cell, play an active role, in which each tail is connected to neighbouring tails by means of one O—H...N hydrogen bond and one leading C—H...O hydrogen bond (Taylor & Kennard, 1982; Steiner & Desiraju, 1998), so the hydroxy O atom acts as both donor and acceptor in two different hydrogen bonds. Atom O1 acts as a hydrogen-bond donor, *via* atom H1O, to atom N1 at $(-x + \frac{3}{2}, y - \frac{1}{2}, -z + \frac{3}{2})$, while atom C8 acts as a hydrogen-bond donor, *via* atom H8B, to atom O1 at $(-x + \frac{3}{2}, y - \frac{1}{2}, -z + \frac{3}{2})$, so generating a centrosymmetric $R_2^2(9)$ ring (Bernstein *et al.*, 1995). The geometric parameters of the C—H...O bonds are well within the accepted ranges (Desiraju, 1991, 1996) and the C—H...O angle is close to the optimally observed value of 160°. A similar type of graph set produced by intermolecular interactions of these types is also observed in the literature (Howie *et al.*, 2003). Propagation of this hydrogen-bonding motif generates one-dimensional linear chains of edge-fused rings running parallel to the [010] direction (Fig. 2). In addition, as can be seen in Fig. 2, this arrangement forms another one-dimensional linear chain of edge-fused rings running parallel to the [010] direction between two chains of fused $R_2^2(9)$ rings [graph set $R_4^4(16)$]. The detailed geometry of the intra- and intermolecular interactions is given in Table 2. There are no

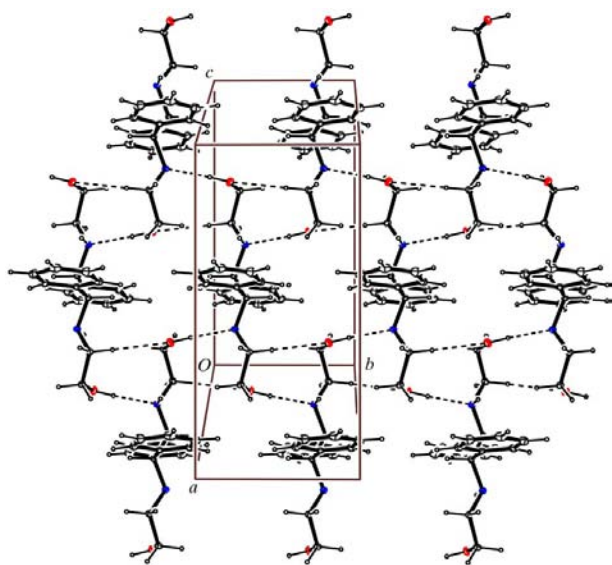


Figure 2
Part of the crystal structure of (I), showing the formation of one-dimensional linear chains of edge-fused rings along [010].

other significant interactions, such as π – π and C—H... π interactions, in the crystal structure of (I).

Experimental

The synthesis of the title compound was performed with minor modifications of the literature method of Koçak (2000). Benzaldehyde (10.6 g, 0.1 mol) and 2-aminoethanol (6.1 g, 0.1 mol) were mixed and heated in a water bath for 1 h. The mixture was then cooled to room temperature and a mixture of aluminium amalgam (7.0 g) in ether (400 ml) and distilled water (7.5 ml) was added slowly. The reaction mixture was kept at room temperature for 24 h with stirring and the precipitate which formed was removed by suction filtration. The compound was extracted from the precipitate with benzene by the use of a Soxhlet extractor. The white crystalline compound which formed was removed by filtration and recrystallized from ethanol [yield: 6.90 g, 46%; m.p. 362 K, literature value 363 K (Jeyaprabha *et al.*, 2005)]. $^1\text{H NMR}$ (DMSO- d_6): δ 7.16 (*m*, 10H, Ar-H), 4.19 (*t*, 2H, OH, D₂O exchangeable), 3.77 (*s*, 2H, >CH—), 3.31 (*t*, 4H, O—CH₂—), 2.31 (*t*, 4H, N—CH₂), 1.92 (*s*, 2H, —NH—, D₂O exchangeable); characteristic IR bands (cm^{−1}) are: 3410 [ν (O—H)], 2800–3000 [aliphatic ν (C—H)], 3000–3100 [aromatic ν (C—H)], 3292 [ν (N—H)], 1619 [ν (C=N)]. The compound is soluble in common organic solvents such as benzene, toluene, ethanol and methanol.

Crystal data

$\text{C}_{18}\text{H}_{24}\text{N}_2\text{O}_2$	$D_x = 1.217 \text{ Mg m}^{-3}$
$M_r = 300.39$	Mo $K\alpha$ radiation
Monoclinic, $C2/c$	Cell parameters from 4012 reflections
$a = 17.585 (2) \text{ \AA}$	$\theta = 2.4\text{--}27.9^\circ$
$b = 6.8615 (5) \text{ \AA}$	$\mu = 0.08 \text{ mm}^{-1}$
$c = 13.9453 (16) \text{ \AA}$	$T = 296 \text{ K}$
$\beta = 103.035 (10)^\circ$	Prism, colourless
$V = 1639.3 (3) \text{ \AA}^3$	$0.57 \times 0.51 \times 0.44 \text{ mm}$
$Z = 4$	

Data collection

Stoe IPDS-2 diffractometer	$R_{\text{int}} = 0.096$
ω scans	$\theta_{\text{max}} = 26.0^\circ$
4103 measured reflections	$h = -16 \rightarrow 21$
1597 independent reflections	$k = -8 \rightarrow 8$
1388 reflections with $I > 2\sigma(I)$	$l = -17 \rightarrow 17$

Refinement

Refinement on F^2	$w = 1/[\sigma^2(F_o^2) + (0.0838P)^2 + 0.3333P]$
$R[F^2 > 2\sigma(F^2)] = 0.053$	where $P = (F_o^2 + 2F_c^2)/3$
$wR(F^2) = 0.148$	$(\Delta/\sigma)_{\text{max}} < 0.001$
$S = 1.09$	$\Delta\rho_{\text{max}} = 0.31 \text{ e \AA}^{-3}$
1597 reflections	$\Delta\rho_{\text{min}} = -0.20 \text{ e \AA}^{-3}$
148 parameters	
All H-atom parameters refined	

Table 1

Selected geometric parameters (\AA , $^\circ$).

O1—C9	1.408 (2)	C1—C7	1.512 (2)
N1—C8	1.4732 (16)	C7—C7 ⁱ	1.541 (2)
N1—C7	1.4781 (16)	C8—C9	1.512 (2)
C8—N1—C7	111.95 (10)	N1—C7—C7 ⁱ	109.20 (13)
C6—C1—C7	120.45 (14)	C1—C7—C7 ⁱ	112.29 (13)
C2—C1—C7	121.35 (13)	N1—C8—C9	110.78 (12)
N1—C7—C1	109.75 (10)	O1—C9—C8	113.23 (12)

Symmetry code: (i) $-x + \frac{3}{2}, -y + \frac{1}{2}, -z + 1$.

The H atoms were located in a difference map and refined isotropically, with C—H = 0.93 (3)–1.036 (16) \AA , O—H = 0.91 (3) \AA and N—H = 0.922 (16) \AA .

Table 2

Hydrogen-bond geometry (Å, °).

$D-H\cdots A$	$D-H$	$H\cdots A$	$D\cdots A$	$D-H\cdots A$
$N1-H1\cdots O1$	0.92 (2)	2.48 (2)	2.901 (2)	107.8 (11)
$O1-H1O\cdots N1^{ii}$	0.91 (3)	1.95 (3)	2.859 (2)	175 (2)
$C8-H8B\cdots O1^{ii}$	0.99 (2)	2.60 (2)	3.548 (2)	161 (2)

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

Data collection: *X-AREA* (Stoe & Cie, 2002); cell refinement: *X-AREA*; data reduction: *X-RED32* (Stoe & Cie, 2002); 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) and *PLATON* (Spek, 2003).

Supplementary data for this paper are available from the IUCr electronic archives (Reference: GZ1025). Services for accessing these data are described at the back of the journal.

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