

1,4-Bis{2-hydroxy-3-[N-(2-hydroxyethyl)imino]-5-methylbenzyl}piperazine

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Key indicators

Single-crystal X-ray study

$T = 293\text{ K}$

Mean $\sigma(\text{C}-\text{C}) = 0.003\text{ \AA}$

R factor = 0.060

w R factor = 0.190

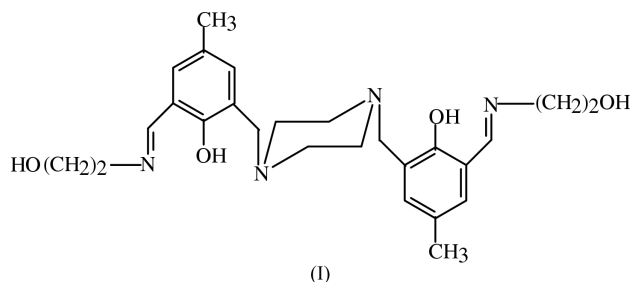
Data-to-parameter ratio = 20.0

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

In the title compound, $\text{C}_{26}\text{H}_{36}\text{N}_4\text{O}_4$, the two phenyl rings are almost parallel, making a dihedral angle of $2.54(6)^\circ$. The piperazine ring adopts a chair conformation, with the substituents on the N atoms both lying in equatorial positions so that the molecule has an extended conformation and is pseudocentrosymmetric. The crystal structure is stabilized by intramolecular and intermolecular $\text{O}-\text{H}\cdots\text{N}$ hydrogen bonds and weak $\text{C}-\text{H}\cdots\text{O}$ interactions.

Comment

An X-ray study of the title compound, (I), was of interest because some piperazine-containing derivatives constitute a novel class of mixed $D2/D4$ receptor antagonists (Zhao *et al.*, 2002), and N,N' -disubstituted piperazine derivatives are anti-filarial, antiamebic and spermicidal agents (Sonurlikar *et al.*, 1977). In addition, piperazine derivatives are useful precursors of mixed-ligand dithiolenes of interest for non-linear optics (Bigoli *et al.*, 2001).



The molecular structure of (I) and the atom-numbering scheme are shown in Fig. 1. The bond lengths observed in the two phenyl rings agree with each other and are comparable with average values reported in the literature (Domenicano *et al.*, 1975; Allen *et al.*, 1987). The bond lengths and angles observed in the piperazine ring are comparable with those of related reported structures (Gunasekaran *et al.*, 1996; Thirumurugan *et al.*, 1998). The bond lengths $\text{C}23-\text{N}24$ and $\text{C}30-$

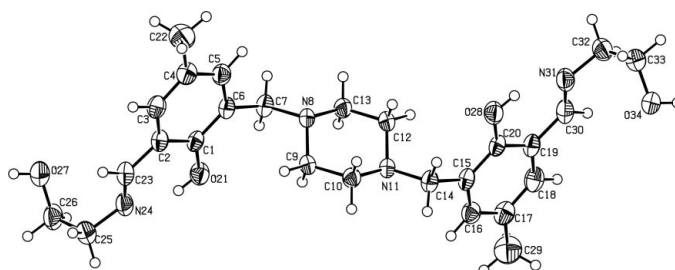


Figure 1

The molecular structure of (I), showing ellipsoids at the 50% probability level.

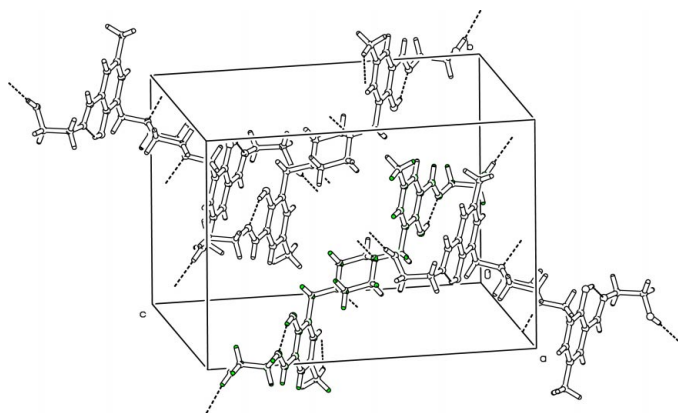


Figure 2
The packing of the molecules, viewed down the *a* axis.

N31, which are 1.260 (2) and 1.252 (3) Å, respectively, indicate a double-bond character (Thirumurugan *et al.*, 1998).

The sum of the bond angles around atoms N8 [329.2 (1)°] and N11 [330.3 (1)°] indicate that these atoms are sp^3 -hybridized (Perales *et al.*, 1977; Gunasekaran *et al.*, 1996).

The conformation of the attachment of the two phenyl rings to the piperazine ring is described by the torsion angles C13–N8–C7–C6 [–170.9 (2)°] and C10–N11–C14–C15 [170.7 (2)°], *i.e.* they adopt –antiperiplanar and +anti-periplanar conformations with respect to one another. The torsion angles C5–C6–C7–N8 and C16–C15–C14–N11 [76.6 (2) and –78.3 (2)°, respectively] define the orientation of the substituents on the piperazine ring; both are in equatorial positions and the molecule thus has an extended conformation and is pseudocentrosymmetric (Thirumurugan *et al.*, 1998).

The two phenyl rings (C1–C6 and C15–C20) are planar and almost parallel, with a maximum deviation of 0.01 (3) Å for atom C5. The dihedral angle between these two phenyl rings is 2.54 (6)°.

The piperazine ring adopts a chair conformation. This is confirmed by the puckering parameters $q_2 = 0.010$ (4), $q_3 = -0.577$ (4), $Q_T = 0.577$ (4) and $\theta = 178.9$ (4) (Cremer & Pople, 1975).

The structure is stabilized by C–H···O and O–H···N intramolecular interactions. In the crystal, symmetry-related molecules are linked by O–H···N and C–H···O intermolecular hydrogen bonds. The hydrogen bonds observed in the structure are listed in Table 2.

Experimental

Paraformaldehyde (2 mol) and piperazine (1 mol) were stirred in 15 ml of acetic acid for 1 h. 4-Methyl-2-formylphenol (2 mol), dissolved in 20 ml of acetic acid, was added and the solution was stirred for 6 h. This 2:2:1 mixture was subjected to Schiff base condensation with two moles of 2-aminoethanol to yield the title compound. The resulting mixture was neutralized with saturated Na_2CO_3 and the title compound was extracted with chloroform. The crude sample was purified on a silica-gel column using an *n*-hexane–chloroform (30:70 *v/v*) solvent mixture as eluant. The compound was crystallized by slow evaporation from an ether/ CHCl_3 mixture.

Crystal data

$\text{C}_{26}\text{H}_{36}\text{N}_4\text{O}_4$
 $M_r = 468.62$
Monoclinic, $P2_1/c$
 $a = 9.7575$ (2) Å
 $b = 13.8091$ (3) Å
 $c = 18.4270$ (1) Å
 $\beta = 99.964$ (1)°
 $V = 2445.45$ (7) Å³
 $Z = 4$

$D_x = 1.273$ Mg m⁻³
Mo $K\alpha$ radiation
Cell parameters from 6435 reflections
 $\theta = 1.9$ – 28.6 °
 $\mu = 0.09$ mm⁻¹
 $T = 293$ (2) K
Block, colourless
0.48 × 0.38 × 0.22 mm

Data collection

Siemens SMART CCD area-detector diffractometer
 ω scans
17 219 measured reflections
6219 independent reflections
3721 reflections with $I > 2\sigma(I)$

$R_{\text{int}} = 0.043$
 $\theta_{\text{max}} = 28.7$ °
 $h = -13 \rightarrow 11$
 $k = -17 \rightarrow 18$
 $l = -23 \rightarrow 24$

Refinement

Refinement on F^2
 $R[F^2 > 2\sigma(F^2)] = 0.060$
 $wR(F^2) = 0.190$
 $S = 1.06$
6219 reflections
311 parameters

H-atom parameters constrained
 $w = 1/[\sigma^2(F_o^2) + (0.1P)^2]$
where $P = (F_o^2 + 2F_c^2)/3$
 $(\Delta/\sigma)_{\text{max}} < 0.001$
 $\Delta\rho_{\text{max}} = 0.28$ e Å⁻³
 $\Delta\rho_{\text{min}} = -0.21$ e Å⁻³

Table 1

Selected geometric parameters (Å, °).

C1–O21	1.345 (2)	N11–C12	1.451 (2)
C4–C22	1.493 (3)	N11–C14	1.464 (2)
C7–N8	1.469 (2)	C12–C13	1.504 (2)
N8–C9	1.453 (2)	C17–C29	1.496 (3)
N8–C13	1.455 (2)	C20–O28	1.346 (2)
C9–C10	1.506 (2)	C23–N24	1.260 (2)
C10–N11	1.454 (2)	C30–N31	1.252 (3)
O21–C1–C6	119.4 (2)	C12–N11–C14	111.2 (1)
O21–C1–C2	120.5 (2)	C10–N11–C14	110.1 (1)
C3–C4–C22	121.3 (2)	C18–C17–C29	121.4 (2)
C5–C4–C22	121.1 (2)	C16–C17–C29	121.0 (2)
C9–N8–C13	109.3 (1)	O28–C20–C15	119.3 (2)
C9–N8–C7	110.8 (1)	O28–C20–C19	120.5 (2)
C13–N8–C7	109.1 (1)	C23–N24–C25	117.3 (2)
C12–N11–C10	109.0 (1)	C30–N31–C32	117.0 (2)
N8–C7–C6–C5	76.6 (2)	C10–N11–C14–C15	170.7 (2)
C6–C7–N8–C13	–170.9 (2)	N11–C14–C15–C16	–78.3 (2)

Table 2

Hydrogen-bonding geometry (Å, °).

<i>D</i> – <i>H</i> ··· <i>A</i>	<i>D</i> – <i>H</i>	<i>H</i> ··· <i>A</i>	<i>D</i> ··· <i>A</i>	<i>D</i> – <i>H</i> ··· <i>A</i>
O21–H21···N24	0.82	1.88	2.609 (2)	148
O28–H28···N31	0.82	1.88	2.608 (2)	148
C7–H7A···O21	0.97	2.43	2.848 (2)	105
C14–H14B···O28	0.97	2.43	2.837 (2)	105
O27–H27···N11 ⁱ	0.82	2.00	2.811 (2)	171
O34–H34···N8 ⁱⁱ	0.82	2.04	2.852 (2)	172
C29–H29A···O27 ⁱⁱⁱ	0.96	2.50	3.388 (3)	154

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

The H atoms were positioned geometrically and were treated as riding on their parent C and O atoms, with aromatic C–H distances of 0.93 Å, other C–H distances of 0.97 Å and O–H distances of 0.82 Å.

Data collection: *SMART* (Siemens, 1996); cell refinement: *SAINT* (Siemens, 1996); 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* (Farrugia, 1997) and *PLATON* (Spek, 2003); software used to prepare material for publication: *SHELXL97* and *PARST* (Nardelli, 1995).

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