Preliminary examination and data collection were performed employing Siemens CCD and *P*4 automated single-crystal X-ray diffractometers using graphite-monochromated Mo $K\alpha$ radiation (λ = 0.71073 Å). In the case of the CCD data set [for (3*a*)], ω-scan and double-pass methods were used for data collection. The first 50 frames were recollected at the end of the data collection to monitor crystal decay. *SADABS* correction (Blessing, 1995) was applied to the raw data. The structures were solved by direct methods. The non-H atoms were refined (least squares) anisotropically to convergence. H atoms were treated using an appropriate riding model [C—H = 0.94 Å and U(H) = 1.2U_{eq}(carrier), or, for methyl H atoms, C—H = 0.98 Å and U(H) = 1.5U_{eq}(carrier)].

Data collection: *SMART* (Siemens, 1996*b*) for (3*a*); *XSCANS* (Siemens, 1996*c*) for (4*b*). Cell refinement: *SAINT* (Siemens, 1996*a*) for (3*a*); *XSCANS* for (4*b*). Data reduction: *SAINT* for (3*a*); *SHELXTL-Plus* (Sheldrick, 1995) for (4*b*). For both compounds, program(s) used to solve structures: *SHELXTL-Plus*; program(s) used to refine structures: *SHELXTL-Plus*; molecular graphics: *SHELXTL-Plus*; software used to prepare material for publication: *SHELXTL-Plus*.

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N,N'-Methylenebis[(*S*)-5-phenyloxazolidine]

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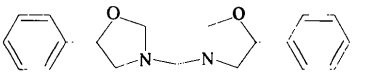
Abstract

The 2:3 condensation product, $C_{19}H_{22}N_2O_2$, formed by reaction of (*S*)-(+)-2-amino-1-phenylethanol with formaldehyde has an *N,N'*-methylenebis(oxazolidine) heterocyclic skeleton. The two oxazolidine rings have envelope-type conformations, with their N atoms displaced by 0.604 (5) and 0.537 (6) Å from the mean planes formed by the remaining four atoms of each ring. The dihedral angle between the two planes is 110.2 (2)°.

Comment

The condensation product of (±)-2-amino-1-phenylethanol with formaldehyde in a respective molar ratio of 2:3 was first described as a synthetic reagent in 1994 and was assumed to have an *N,N'*-methylenebis(oxazolidine) heterocyclic skeleton (Pevarello *et al.*, 1994). While it has been shown that a similar aminoalcohol, (–)-norephedrine, with formaldehyde gives a 2:3 adduct which has this type of structure (Engel *et al.*, 1982), another related aminoalcohol, (–)-2-amino-2-phenylethanol, gives a quite different isomeric 2:3 adduct, having a 1,6-diaza-3,8-dioxabicyclo[4.4.1]undecane skeleton (Aitken *et al.*, 1991). This latter observation casts some doubt on the assumption of the structure of the title compound, (I), and since firm knowledge of its molecular structure is necessary in order to understand its chemical reactivity, confirmation was sought by an X-ray diffraction study.



(I)

The title compound consists of two 5-phenyloxazolidine rings with an –NCH_2N– bridge. Corresponding bond angles and distances in each of the

two ring systems are very similar and are comparable with those observed for *N,N'*-methylenebis(4-methyl-5-phenyloxazolidine) (Engel *et al.*, 1982). The molecules have almost perfect C_2 symmetry about the C6 atom. The conformation of the oxazolidine rings is of the envelope type, with atom N1 0.604 (5) Å from plane *P*3 (defined by atoms C2, O3, C4 and C5) and atom N7 0.537 (6) Å from plane *P*4 (defined by atoms C8, O9, C10 and C11). *P*3 makes a dihedral angle of 110.2 (2)° with *P*4. The endocyclic torsion angles N1—C2—O3—C4 and N7—C8—O9—C10 are −26.3 (4) and −15.6 (4)°, respectively. The dihedral angles (Ito & Sugawara, 1983) between planes *P*1 (C12–C17) and *P*3 on the one hand, and planes *P*2 (C18–C23) and *P*4 on the other hand, are 79.7 (2) and 18.8 (2)°, respectively.

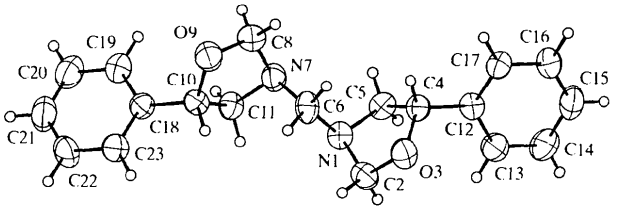


Fig. 1. Perspective view of *N,N'*-methylenebis(5-phenyloxazolidine). Displacement ellipsoids have been scaled to enclose regions of 50% probability.

In the crystal structure, the packing is due mainly to van der Waals interactions [the smallest value is 3.440 (5) Å for N1···C5^{i}; symmetry code: (i) $x-\frac{1}{2}$, $\frac{1}{2}-y$, $1-z$] and all intermolecular contacts agree with those predicted from radii-sum rules.

Experimental

The title compound was prepared according to the literature procedure of Pevarello *et al.* (1994), except that (*S*)-(+)-2-amino-1-phenylethanol (Meyers & Slade, 1980) was used as starting material instead of the racemic compound. A single crystal was grown by slow evaporation of a heptane–benzene solution.

Crystal data

$C_{19}H_{22}N_2O_2$
M_r = 310.39
Orthorhombic
$P2_12_12_1$
a = 5.960 (3) Å
b = 8.312 (5) Å
c = 32.80 (3) Å
V = 1624.7 (18) Å^3
Z = 4
D_x = 1.269 Mg m^{-3}
D_m = 1.24 Mg m^{-3}
D_m measured by flotation in CH_2Cl_2 and C_6H_{12}
Mo $K\alpha$ radiation
λ = 0.71069 Å
Cell parameters from 28 reflections
θ = 6.16–16.25°
μ = 0.083 mm^{-1}
T = 293 (2) K
Parallelepiped
0.43 × 0.38 × 0.25 mm
White

Data collection

Siemens *P*3 diffractometer
ω–2θ scans
Absorption correction: none
3092 measured reflections
2858 independent reflections
1573 reflections with $I > 2\sigma(I)$
R_{int} = 0.066
θ_{max} = 25.04°
$h = -7 \rightarrow 7$
$k = 0 \rightarrow 9$
$l = 0 \rightarrow 38$
3 standard reflections every 60 reflections
intensity decay: 2%

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)]$ = 0.058
$wR(F^2)$ = 0.200
S = 1.078
2808 reflections
228 parameters
H atoms: see below
$w = 1/[\sigma^2(F_o^2) + (0.0386P)^2 + 0.0232P]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{max}$ = 0.004
$\Delta\rho_{max}$ = 0.124 e Å^{-3}
$\Delta\rho_{min}$ = −0.175 e Å^{-3}
Extinction correction: none
Scattering factors from *International Tables for Crystallography* (Vol. C)

Table 1. *Selected geometric parameters (Å, °)*

N1—C2	1.438 (5)	C6—N7	1.460 (5)
N1—C5	1.458 (4)	N7—C8	1.438 (5)
N1—C6	1.464 (4)	N7—C11	1.461 (4)
C2—O3	1.444 (4)	C8—O9	1.431 (5)
O3—C4	1.441 (4)	O9—C10	1.432 (4)
C4—C12	1.501 (5)	C10—C18	1.503 (5)
C4—C5	1.551 (5)	C10—C11	1.543 (5)
C2—N1—C5	99.8 (3)	C8—N7—C6	113.2 (3)
C2—N1—C6	112.8 (3)	C8—N7—C11	101.6 (3)
C5—N1—C6	111.8 (3)	C6—N7—C11	110.7 (3)
N1—C2—O3	107.4 (3)	O9—C8—N7	109.0 (3)
C4—O3—C2	107.1 (3)	C8—O9—C10	107.6 (3)
O3—C4—C12	112.2 (3)	O9—C10—C18	111.9 (3)
O3—C4—C5	102.9 (3)	O9—C10—C11	103.8 (3)
C12—C4—C5	115.7 (3)	C18—C10—C11	115.0 (3)
N1—C5—C4	104.6 (3)	N7—C11—C10	103.6 (3)
N7—C6—N1	109.6 (3)		
C4—O3—C2—N1	−26.3 (4)	N7—C8—O9—C10	−15.6 (4)
O3—C2—N1—C5	41.4 (4)	O9—C10—C11—N7	28.9 (4)
C2—N1—C5—C4	−39.9 (4)	C8—O9—C10—C11	−8.5 (4)
N1—C5—C4—O3	25.2 (4)	C8—N7—C11—C10	−37.3 (4)
C5—C4—O3—C2	0.2 (4)	O9—C8—N7—C11	33.7 (4)

Refinement was on F^2 for all reflections except for 50 with very negative F^2 or flagged for potential systematic errors. The H atoms were placed in geometrically calculated positions and their positions were allowed to refine with C—H distance restraints. A common isotropic displacement parameter was refined for each type of H atom (*i.e.* CH, CH_2, *etc.*). The choice of the *S,S* enantiomer for the refinement was based on the known absolute configuration of the synthetic precursor, (*S*)-(+)-2-amino-1-phenylethanol.

Data collection: *P*3 (Siemens, 1990). Cell refinement: *P*3. Data reduction: *P*3. Program(s) used to solve structure: *SHELXS*86 (Sheldrick, 1985). Program(s) used to refine structure: *SHELXL*93 (Sheldrick, 1993). Molecular graphics: *CAMERON* (Watkin *et al.*, 1996). Software used to prepare material for publication: *SHELXL*93.

Supplementary data for this paper are available from the IUCr electronic archives (Reference: LN1028). Services for accessing these data are described at the back of the journal.

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2′-[1-(4-Fluorophenyl)ethyl]isonicotino-κN-hydrazide–Cyanoborane

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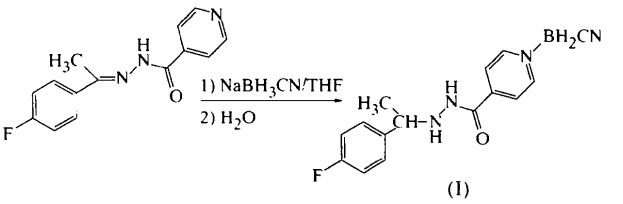
Abstract

The title compound, $C_{15}H_{16}BFN_4O$, is a cyanoborane adduct showing a butterfly-like conformation, with the fluorophenyl moiety disordered.

Comment

Interest in cyanoborane adducts stems from their extraordinary biological activities, *i.e.* anti-inflammatory (Hall *et al.*, 1980), anti-neoplastic (Sood *et al.*, 1991) and hypolipidemic (Hall *et al.*, 1981). By using known procedures (Calabretta *et al.*, 1991), an extensive series of cyanoborane adducts of aliphatic and aromatic isonicotinohydrazides has recently been prepared (Vigorita *et al.*, 1998). In particular, cyanoboranes derived from the primary antitubercular agent isoniazid (INH) should be of interest as potential anti-infective agents, in view of the current re-emergence of tuberculosis and other AIDS-associated microbacterial diseases. The synthesis of the title isonicotinohydrazide–cyanoborane adduct, (I), was carried out by the treatment of 4′-fluoroacetophenone isonicotinoylhydrazone with an excess of sodium cyanoborohydride at pH 3–5 in anhydrous tetrahydrofuran.



(I)

However, since the title compound possesses at least two basic N atoms that might be in competition as sites coordinating BH_2CN (Calabretta *et al.*, 1991), the present X-ray diffraction analysis was required to determine the structure. This analysis shows that the packing is mainly determined by normal van der Waals interactions and intermolecular hydrogen bonds involving O1 and N1 (Table 2). Intermolecular hydrogen-bond interactions also occur in the disordered part of the molecule and involve the F atom (H···F distances range from 2.43 to 2.98 Å, whereas C—H···F angles range from 105 to 164°).

The cyanoborane group shows the typical tetrahedral geometry for the B atom [C1—B1—N2 = 109.2 (3)°], and distances and angles are in good agreement with those reported in the literature (Ferguson *et al.*, 1990). The angle between the N2—B1—C1—N1 moiety and the pyridine ring is 55.9 (2)°. The interplanar angle between the pyridine system and the O1—C7—N3—N4 moiety is 11.1 (1)°, showing that π-electron delocalization is not wide-ranging, since it is confined to the pyridine ring. The mutual orientation of the aromatic ring and the hydrazonic moiety is due to the presence of a weak intramolecular hydrogen-bond interaction involving C5 and O1 (Table 2).

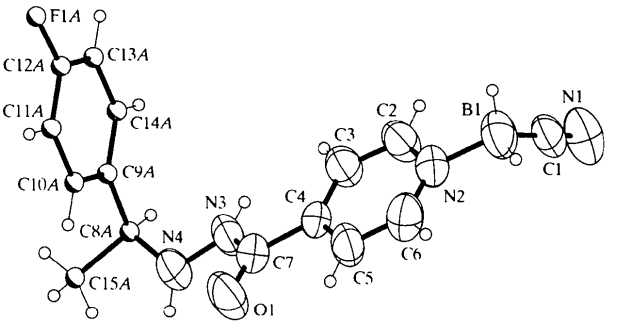


Fig. 1. View of the title compound showing the atomic numbering scheme, with displacement ellipsoids at 50% probability for non-H atoms. For clarity, only one position of the disordered moiety has been labelled.