organic compounds

1-(2,3-Dimethoxybenzyl)-*N*-nitro-imidazolidin-2-imine

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In the title compound, $C_{12}H_{16}N_4O_4$, the *N*-nitroimidazolidin-2-imine ring and benzene ring make a dihedral angle of 86.98 (2)°. Intermolecular N—H···O hydrogen bonds link the molecules into hydrogen-bonded dimers. C—H···O hydrogen bonds are also present.

Related literature

For the crystal structures of related compounds, see: Nordenson (1981); Ravikumar *et al.* (2006). For details of the biological activities of nicotinic compounds, see: Kagabu (1997).
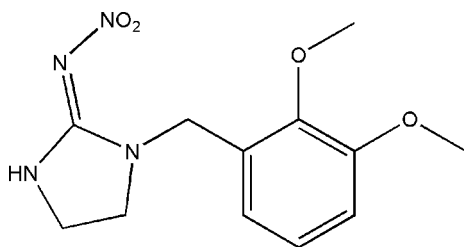
Experimental

Crystal data

$C_{12}H_{16}N_4O_4$	V = 1346.1 (6) Å^3
M_r = 280.29	Z = 4
Monoclinic, $P2_1/n$	Mo $K\alpha$ radiation
a = 11.949 (3) Å	μ = 0.11 mm^{-1}
b = 9.152 (2) Å	T = 294 (2) K
c = 12.312 (3) Å	0.42 × 0.26 × 0.10 mm
β = 91.102 (5)°	

Data collection

Bruker SMART CCD area-detector diffractometer	7385 measured reflections
	2752 independent reflections
Absorption correction: multi-scan (*SADABS*; Sheldrick, 2004)	1384 reflections with $I > 2\sigma(I)$
T_{min} = 0.957, T_{max} = 0.990	R_{int} = 0.050

Refinement

$R[F^2 > 2\sigma(F^2)]$ = 0.047	183 parameters
$wR(F^2)$ = 0.136	H-atom parameters constrained
S = 1.00	$\Delta\rho_{max}$ = 0.18 e Å^{-3}
2752 reflections	$\Delta\rho_{min}$ = −0.18 e Å^{-3}

Table 1
Hydrogen-bond geometry (Å, °).

D—H···A	D—H	H···A	D···A	D—H···A
N2—H2A···O2^i	0.86	2.18	2.908 (3)	143
C10—H10B···O3^{ii}	0.97	2.56	3.296 (3)	133

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

Data collection: *SMART* (Bruker, 2001); cell refinement: *SAINT* (Bruker, 2001); data reduction: *SAINT*; program(s) used to solve structure: *SHELXTL* (Sheldrick, 2001); program(s) used to refine structure: *SHELXTL*; molecular graphics: *SHELXTL*; software used to prepare material for publication: *SHELXTL*.

References

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supplementary materials

1-(2,3-Dimethoxybenzyl)-*N*-nitroimidazolidin-2-imine

L.-Z. Xu, Z.-J. Xu and W.-Z. Bi

Comment

Nicotinic insecticide containing *N*-nitroimidazolidin-2-imine group is given more attention because its characteristics of high-efficient, low toxicity, broad-spectrum, security and environmental protection (Kagabu, 1997). We report here the crystal structure of (I).

Bond lengths and angles of the *N*-nitroimidazolidin-2-imine group are in agreement with those in previous reports (Nordenson, 1981; Ravikumar *et al.*, 2006). The *N*-nitroimidazolidin-2-imine ring (N1/N2/C10/C11/C12) and benzene rings (C1–C6) make a dihedral angle of 86.98 (2)°. Intermolecular N—H·O hydrogen bonds link the two molecules into hydrogen-bonded dimers.

Experimental

To a 50-ml flask, 3.5 mmol of *N*-nitroimidazolidin-2-imine, 3.5 mmol of 1-(chloromethyl)-2,3-dimethoxybenzene, and 3.5 mmol of potassium carbonate in 30 ml of acetonitrile was refluxed for 3–4 h. The reaction mixture was filtered and an excess of acetonitrile was removed under reduced pressure to give title compound. Single crystals suitable for X-ray measurements were obtained by recrystallization from methanol at room temperature.

Refinement

All H atoms were placed in calculated positions, with N—H = 0.86 and C—H = 0.93–0.97 Å, and included in the final cycles of refinement using a riding model, with $U_{iso}(H) = 1.2(1.5$ for methyl groups) times $U_{eq}(C,N)$.
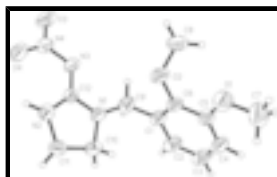
Figures

Fig. 1. View of the title compound (I), with displacement ellipsoids drawn at the 40% probability level.
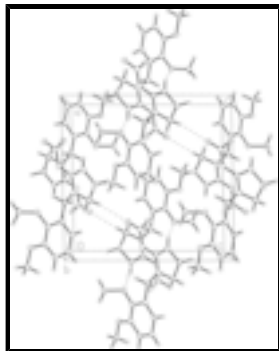
Fig. 2. A packing diagram of the molecule of the title compound, view down *b* axis. Hydrogen bonds are shown as dashed lines.

1-(2,3-Dimethoxybenzyl)-*N*-nitroimidazolidin-2-imine

Crystal data

$C_{12}H_{16}N_4O_4$	$F_{000} = 592$
$M_r = 280.29$	$D_x = 1.383$ Mg m^{-3}
Monoclinic, $P2_1/n$	Mo $K\alpha$ radiation $\lambda = 0.71073$ Å
Hall symbol: -P 2yn	Cell parameters from 2652 reflections
$a = 11.949$ (3) Å	$\theta = 2.6–25.6°$
$b = 9.152$ (2) Å	$\mu = 0.11$ mm^{-1}
$c = 12.312$ (3) Å	$T = 294$ (2) K
$\beta = 91.102$ (5)°	Block, yellow
$V = 1346.1$ (6) Å^3	$0.42 \times 0.26 \times 0.10$ mm
$Z = 4$	

Data collection

Bruker SMART CCD area-detector diffractometer	2752 independent reflections
Radiation source: fine-focus sealed tube	1384 reflections with $I > 2\sigma(I)$
Monochromator: graphite	$R_{int} = 0.050$
$T = 294(2)$ K	$\theta_{max} = 26.5°$
φ and ω scans	$\theta_{min} = 2.4°$
Absorption correction: multi-scan (SADABS; Sheldrick, 2004)	$h = -14 \rightarrow 14$
$T_{min} = 0.957$, $T_{max} = 0.990$	$k = -9 \rightarrow 11$
7385 measured reflections	$l = -15 \rightarrow 10$

Refinement

Refinement on F^2	Secondary atom site location: difference Fourier map
Least-squares matrix: full	Hydrogen site location: inferred from neighbouring sites
$R[F^2 > 2\sigma(F^2)] = 0.047$	H-atom parameters constrained
$wR(F^2) = 0.136$	$w = 1/[\sigma^2(F_o^2) + (0.0577P)^2 + 0.143P]$

$$\text{where } P = (F_o{}^2 + 2F_c{}^2)/3$$

$S = 1.00$	$(\Delta/\sigma)_{max} = 0.004$
2752 reflections	$\Delta\rho_{max} = 0.18$ e Å^{-3}
183 parameters	$\Delta\rho_{min} = -0.18$ e Å^{-3}
Primary atom site location: structure-invariant direct methods	Extinction correction: none

Special details

Geometry. All e.s.d.'s (except the e.s.d. in the dihedral angle between two l.s. planes) are estimated using the full covariance matrix. The cell e.s.d.'s are taken into account individually in the estimation of e.s.d.'s in distances, angles and torsion angles; correlations between e.s.d.'s in cell parameters are only used when they are defined by crystal symmetry. An approximate (isotropic) treatment of cell e.s.d.'s is used for estimating e.s.d.'s involving l.s. planes.

Refinement. Refinement of F^2 against ALL reflections. The weighted *R*-factor *wR* and goodness of fit S are based on F^2, conventional *R*-factors *R* are based on F, with F set to zero for negative F^2. The threshold expression of F$^2 > 2$sigma(F^2) is used only for calculating *R*-factors(gt) *etc*. and is not relevant to the choice of reflections for refinement. *R*-factors based on F^2 are statistically about twice as large as those based on F, and R– factors based on ALL data will be even larger.

Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters (Å^2)

	x	y	z	U_{iso}*/U_{eq}
O1	0.92476 (15)	0.0235 (2)	0.19671 (15)	0.0798 (6)
O2	0.71309 (12)	0.11934 (17)	0.16349 (13)	0.0639 (5)
O3	0.27319 (15)	0.3031 (2)	0.12564 (17)	0.0969 (7)
O4	0.27201 (14)	0.1400 (2)	0.00015 (16)	0.0796 (6)
N1	0.59195 (15)	0.2334 (2)	−0.05226 (16)	0.0539 (5)
N2	0.45011 (16)	0.1084 (2)	−0.11518 (16)	0.0612 (6)
H2A	0.3830	0.0748	−0.1202	0.073*
N3	0.42989 (15)	0.2705 (2)	0.04020 (16)	0.0552 (5)
N4	0.32161 (17)	0.2346 (2)	0.05378 (17)	0.0599 (6)
C1	0.77387 (18)	0.2918 (2)	0.0326 (2)	0.0531 (6)
C2	0.8612 (2)	0.3547 (3)	−0.0246 (2)	0.0676 (8)
H2	0.8461	0.4283	−0.0749	0.081*
C3	0.9694 (2)	0.3082 (3)	−0.0071 (2)	0.0717 (8)
H3	1.0270	0.3526	−0.0446	0.086*
C4	0.9941 (2)	0.1975 (3)	0.0650 (2)	0.0663 (8)
H4	1.0676	0.1657	0.0744	0.080*
C5	0.9091 (2)	0.1331 (3)	0.1238 (2)	0.0555 (7)
C6	0.79974 (19)	0.1834 (3)	0.1064 (2)	0.0510 (6)
C7	1.0357 (2)	−0.0281 (3)	0.2161 (3)	0.0920 (10)
H7A	1.0663	−0.0625	0.1493	0.138*
H7B	1.0345	−0.1067	0.2677	0.138*
H7C	1.0812	0.0500	0.2445	0.138*
C8	0.7121 (2)	0.1560 (4)	0.2768 (2)	0.0865 (10)
H8A	0.7832	0.1322	0.3097	0.130*
H8B	0.6541	0.1017	0.3116	0.130*
H8C	0.6981	0.2587	0.2848	0.130*

C9	0.65535 (19)	0.3400 (3)	0.0119 (2)	0.0637 (7)
H9A	0.6190	0.3546	0.0808	0.076*
H9B	0.6555	0.4327	−0.0262	0.076*
C10	0.6384 (2)	0.1580 (3)	−0.1451 (2)	0.0600 (7)
H10A	0.7005	0.0958	−0.1231	0.072*
H10B	0.6636	0.2268	−0.1993	0.072*
C11	0.53989 (19)	0.0677 (3)	−0.18826 (18)	0.0574 (7)
H11A	0.5211	0.0932	−0.2629	0.069*
H11B	0.5558	−0.0361	−0.1840	0.069*
C12	0.48429 (19)	0.2028 (2)	−0.04067 (18)	0.0444 (6)

Atomic displacement parameters (Å^2)

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
O1	0.0706 (13)	0.0912 (14)	0.0777 (14)	0.0060 (11)	0.0037 (11)	0.0097 (12)
O2	0.0538 (11)	0.0780 (11)	0.0606 (11)	−0.0225 (9)	0.0199 (9)	−0.0092 (9)
O3	0.0641 (13)	0.1346 (17)	0.0934 (15)	−0.0125 (12)	0.0356 (12)	−0.0383 (14)
O4	0.0525 (12)	0.0985 (14)	0.0885 (14)	−0.0204 (10)	0.0165 (10)	−0.0211 (12)
N1	0.0413 (12)	0.0620 (12)	0.0587 (13)	−0.0056 (10)	0.0107 (10)	−0.0074 (11)
N2	0.0475 (13)	0.0761 (14)	0.0601 (13)	−0.0105 (11)	0.0066 (10)	−0.0102 (12)
N3	0.0396 (11)	0.0679 (13)	0.0587 (13)	−0.0067 (10)	0.0129 (10)	−0.0067 (11)
N4	0.0490 (14)	0.0748 (14)	0.0564 (14)	−0.0022 (11)	0.0120 (11)	0.0025 (12)
C1	0.0434 (15)	0.0527 (14)	0.0636 (16)	−0.0074 (11)	0.0102 (12)	−0.0085 (13)
C2	0.0563 (18)	0.0683 (17)	0.079 (2)	−0.0121 (14)	0.0198 (15)	0.0039 (15)
C3	0.0551 (19)	0.0798 (19)	0.081 (2)	−0.0219 (15)	0.0291 (15)	−0.0060 (17)
C4	0.0424 (16)	0.0772 (18)	0.080 (2)	−0.0057 (13)	0.0120 (14)	−0.0198 (17)
C5	0.0460 (15)	0.0623 (16)	0.0583 (16)	−0.0095 (12)	0.0059 (13)	−0.0131 (14)
C6	0.0419 (15)	0.0575 (14)	0.0541 (15)	−0.0154 (11)	0.0141 (12)	−0.0132 (12)
C7	0.081 (2)	0.095 (2)	0.100 (2)	0.0179 (17)	−0.0197 (19)	−0.0086 (19)
C8	0.081 (2)	0.119 (2)	0.0606 (19)	−0.0196 (17)	0.0261 (16)	−0.0154 (17)
C9	0.0505 (16)	0.0576 (15)	0.083 (2)	−0.0061 (12)	0.0090 (14)	−0.0047 (14)
C10	0.0578 (16)	0.0682 (16)	0.0546 (16)	0.0017 (13)	0.0156 (13)	0.0006 (13)
C11	0.0594 (16)	0.0677 (15)	0.0453 (14)	0.0070 (13)	0.0073 (12)	0.0020 (12)
C12	0.0437 (14)	0.0469 (13)	0.0428 (13)	0.0000 (11)	0.0022 (11)	0.0082 (12)

Geometric parameters (Å, °)

O1—C5	1.357 (3)	C3—C4	1.376 (4)
O1—C7	1.423 (3)	C3—H3	0.9300
O2—C6	1.392 (2)	C4—C5	1.390 (3)
O2—C8	1.435 (3)	C4—H4	0.9300
O3—N4	1.237 (2)	C5—C6	1.397 (3)
O4—N4	1.233 (2)	C7—H7A	0.9600
N1—C12	1.327 (3)	C7—H7B	0.9600
N1—C10	1.454 (3)	C7—H7C	0.9600
N1—C9	1.458 (3)	C8—H8A	0.9600
N2—C12	1.319 (3)	C8—H8B	0.9600
N2—C11	1.462 (3)	C8—H8C	0.9600
N2—H2A	0.8600	C9—H9A	0.9700

N3—N4	1.348 (3)	C9—H9B	0.9700
N3—C12	1.350 (3)	C10—C11	1.525 (3)
C1—C6	1.377 (3)	C10—H10A	0.9700
C1—C2	1.394 (3)	C10—H10B	0.9700
C1—C9	1.500 (3)	C11—H11A	0.9700
C2—C3	1.374 (3)	C11—H11B	0.9700
C2—H2	0.9300		
C5—O1—C7	118.2 (2)	H7A—C7—H7B	109.5
C6—O2—C8	114.36 (18)	O1—C7—H7C	109.5
C12—N1—C10	111.68 (19)	H7A—C7—H7C	109.5
C12—N1—C9	125.30 (19)	H7B—C7—H7C	109.5
C10—N1—C9	122.76 (19)	O2—C8—H8A	109.5
C12—N2—C11	111.95 (19)	O2—C8—H8B	109.5
C12—N2—H2A	124.0	H8A—C8—H8B	109.5
C11—N2—H2A	124.0	O2—C8—H8C	109.5
N4—N3—C12	117.2 (2)	H8A—C8—H8C	109.5
O4—N4—O3	120.8 (2)	H8B—C8—H8C	109.5
O4—N4—N3	123.8 (2)	N1—C9—C1	112.00 (19)
O3—N4—N3	115.4 (2)	N1—C9—H9A	109.2
C6—C1—C2	118.0 (2)	C1—C9—H9A	109.2
C6—C1—C9	121.5 (2)	N1—C9—H9B	109.2
C2—C1—C9	120.4 (2)	C1—C9—H9B	109.2
C3—C2—C1	120.3 (3)	H9A—C9—H9B	107.9
C3—C2—H2	119.9	N1—C10—C11	103.16 (18)
C1—C2—H2	119.9	N1—C10—H10A	111.1
C2—C3—C4	121.3 (2)	C11—C10—H10A	111.1
C2—C3—H3	119.4	N1—C10—H10B	111.1
C4—C3—H3	119.4	C11—C10—H10B	111.1
C3—C4—C5	119.9 (2)	H10A—C10—H10B	109.1
C3—C4—H4	120.1	N2—C11—C10	102.59 (18)
C5—C4—H4	120.1	N2—C11—H11A	111.2
O1—C5—C4	124.4 (2)	C10—C11—H11A	111.2
O1—C5—C6	117.5 (2)	N2—C11—H11B	111.2
C4—C5—C6	118.1 (3)	C10—C11—H11B	111.2
C1—C6—O2	118.5 (2)	H11A—C11—H11B	109.2
C1—C6—C5	122.4 (2)	N2—C12—N1	110.6 (2)
O2—C6—C5	119.1 (2)	N2—C12—N3	131.8 (2)
O1—C7—H7A	109.5	N1—C12—N3	117.6 (2)
O1—C7—H7B	109.5		
C12—N3—N4—O4	3.3 (3)	O1—C5—C6—O2	−0.2 (3)
C12—N3—N4—O3	−177.1 (2)	C4—C5—C6—O2	179.2 (2)
C6—C1—C2—C3	0.2 (4)	C12—N1—C9—C1	144.6 (2)
C9—C1—C2—C3	−178.7 (2)	C10—N1—C9—C1	−41.7 (3)
C1—C2—C3—C4	1.4 (4)	C6—C1—C9—N1	−74.4 (3)
C2—C3—C4—C5	−1.8 (4)	C2—C1—C9—N1	104.4 (2)
C7—O1—C5—C4	1.1 (3)	C12—N1—C10—C11	−2.2 (3)
C7—O1—C5—C6	−179.4 (2)	C9—N1—C10—C11	−176.6 (2)
C3—C4—C5—O1	−180.0 (2)	C12—N2—C11—C10	−0.7 (3)

C3—C4—C5—C6	0.6 (4)	N1—C10—C11—N2	1.6 (2)
C2—C1—C6—O2	−179.6 (2)	C11—N2—C12—N1	−0.7 (3)
C9—C1—C6—O2	−0.8 (3)	C11—N2—C12—N3	178.5 (2)
C2—C1—C6—C5	−1.3 (3)	C10—N1—C12—N2	1.9 (3)
C9—C1—C6—C5	177.5 (2)	C9—N1—C12—N2	176.1 (2)
C8—O2—C6—C1	−109.7 (3)	C10—N1—C12—N3	−177.48 (19)
C8—O2—C6—C5	71.9 (3)	C9—N1—C12—N3	−3.3 (3)
O1—C5—C6—C1	−178.5 (2)	N4—N3—C12—N2	3.1 (4)
C4—C5—C6—C1	1.0 (3)	N4—N3—C12—N1	−177.63 (19)

Hydrogen-bond geometry (Å, °)

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
N2—H2A···O2[i]	0.86	2.18	2.908 (3)	143
C10—H10B···O3[ii]	0.97	2.56	3.296 (3)	133

Symmetry codes: (i) −*x*+1, −*y*, −*z*; (ii) *x*+1/2, −*y*+1/2, *z*−1/2.

Fig. 1

Fig. 2