

Triclinic
 $P\bar{1}$
 $a = 11.369 (3) \text{ \AA}$
 $b = 12.175 (3) \text{ \AA}$
 $c = 12.268 (3) \text{ \AA}$
 $\alpha = 64.42 (2)^\circ$
 $\beta = 72.89 (2)^\circ$
 $\gamma = 73.37 (2)^\circ$
 $V = 1438.7 (6) \text{ \AA}^3$
 $Z = 1$
 $D_r = 1.245 \text{ Mg m}^{-3}$
 D_m not measured

Data collection

Siemens SMART CCD with
 an LT-2 device for low-
 temperature collection
 Frame scans
 Absorption correction: none
 13 318 measured reflections
 5063 independent reflections

Cell parameters from 65
 reflections
 $\theta = 0-23^\circ$
 $\mu = 0.256 \text{ mm}^{-1}$
 $T = 245 (2) \text{ K}$
 Prism
 $0.33 \times 0.20 \times 0.20 \text{ mm}$
 Pale yellow

Refinement

Refinement on F^2
 $R[F^2 > 2\sigma(F^2)] = 0.036$
 $wR(F^2) = 0.084$
 $S = 1.073$
 5061 reflections
 502 parameters
 H atoms of CH_2Cl_2 riding;
 coordinates of remaining
 H atoms refined, with U_{iso}
 restrained

$w = 1/[\sigma^2(F_o^2) + (0.0533P)^2]$
 where $P = (F_o^2 + 2F_c^2)/3$
 $(\Delta/\sigma)_{\text{max}} = -0.094$
 $\Delta\rho_{\text{max}} = 0.164 \text{ e \AA}^{-3}$
 $\Delta\rho_{\text{min}} = -0.277 \text{ e \AA}^{-3}$
 Extinction correction: none
 Scattering factors from
International Tables for
Crystallography (Vol. C)

Table 2. Selected torsion angles ($^\circ$)

N1—C2—C3—O4	37.7 (2)	C6—N7—C8—C9	-86.0 (2)
C5—C6—N7—C8	160.67 (13)		

Data collection for compound (II) was carried out using a Siemens SMART CCD area detector and standard reflections were, therefore, not measured. The first 50 frames of data were recollected at the end of data collection. No crystal decay was observed.

Data collection: XSCANS (Fait, 1991) for (I); SMART (Siemens, 1994) for (II). Cell refinement: XSCANS for (I); SAINT (Siemens, 1994) for (II). Data reduction: XSCANS for (I); SAINT for (II). For both compounds, program(s) used to solve structures: SIR92 (Altomare *et al.*, 1994); program(s) used to refine structures: SHELXL93 (Sheldrick, 1993); molecular graphics: ZORTEP (Zsolnai & Pritzkow, 1995); software used to prepare material for publication: SHELXL93, PARST (Nardelli, 1983, 1995) and PARST-CIF (Nardelli, 1991).

Helpful discussions with Dr Tullio Pilati are gratefully acknowledged. The author thanks Dr Piero Macchi for his assistance in performing data collection on a Siemens SMART CCD device.

Supplementary data for this paper are available from the IUCr electronic archives (Reference: NA1370). Services for accessing these data are described at the back of the journal.

References

- Altomare, A., Cascarano, G., Giacovazzo, C., Guagliardi, A., Burla, M. C., Polidori, G. & Camalli, M. (1994). *J. Appl. Cryst.* **27**, 435.
 Fait, J. (1991). *XSCANS User's Manual*. Siemens Analytical X-ray Instruments Inc., Madison, Wisconsin, USA.
 Lehn, J.-M. (1995). In *Supramolecular Chemistry*. Weinheim: VCH.
 Méndez, L., Singleton, R., Slawin, A. M. Z., Stoddart, J. F., Williams, D. J. & Williams, M. K. (1992). *Angew. Chem. Int. Ed. Engl.* **31**, 478–480.
 Nardelli, M. (1983). *Comput. Chem.* **7**, 95–98.
 Nardelli, M. (1991). *PARSTCIF. Program for Creating a CIF from the Output of PARST*. University of Parma, Italy.
 Nardelli, M. (1995). *J. Appl. Cryst.* **28**, 659.
 Quici, S., Manfredi, A. & Buttafava, M. (1996). *J. Org. Chem.* **61**, 3870–3873.
 Quici, S. *et al.* (1998). In preparation.
 Sheldrick, G. M. (1993). *Program for the Refinement of Crystal Structures*. University of Göttingen, Germany.
 Siemens (1994). *SMART and SAINT. Area Detector Control and Integration Software*. Siemens Analytical X-ray Instruments Inc., Madison, Wisconsin, USA.
 Zsolnai, L. & Pritzkow, H. (1995). *ZORTEP. An Interactive ORTEP Program*. University of Heidelberg, Germany.

Acta Cryst. (1998). **C54**, 1923–1925

1,2-Bis(2-pyridylformimidoyl)hydrazine†

JENNIFER A. ARMSTRONG,^a JOHN C. BARNES^a AND
 TIMOTHY J. R. WEAKLEY^b

^aDepartment of Chemistry, University of Dundee, Perth Road, Dundee DD1 4HN, Scotland, and ^bDepartment of Chemistry, University of Oregon, Eugene OR 97403-1253, USA. E-mail: j.c.barnes@dundee.ac.uk

(Received 12 March 1998; accepted 8 June 1998)

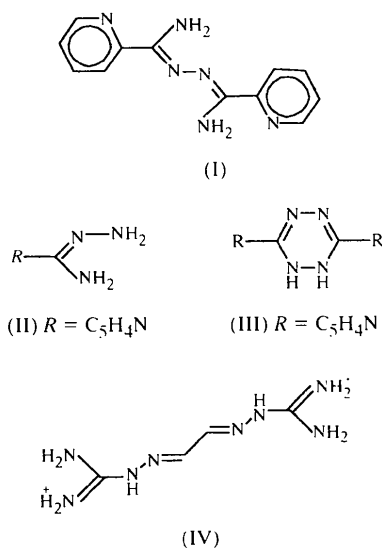
Abstract

The title compound, $\text{C}_{12}\text{H}_{12}\text{N}_6$, is the minor *trans* product of the preparation of a dihydrotetrazine. Extensive electron delocalization is shown by the planarity of the molecule, except for one pyridyl group twisted out of plane by $14.1 (1)^\circ$. The $\text{C}=\text{N}-\text{N}=\text{C}$ fragment has a C—N distance of $1.297 (2) \text{ \AA}$ and an N—N distance of $1.404 (2) \text{ \AA}$. All other C—N distances are close to 1.34 \AA , typical of C=N bonds. The NH_2 groups form hydrogen bonds to pyridyl and hydrazine N atoms of adjacent molecules.

Comment

The title compound, (I), can be regarded as a dihydrazidine or an amide azine (Watson & Neilson, 1975). It is formed as the minor product from refluxing picro-

† Alternative name: 1,2-bis[amino(2-pyridyl)methylene]diazane.



linic acid amidrazone, (II), in ethanol under nitrogen (Kubota *et al.*, 1972). The major product is 3,6-di(2-pyridyl)-1,2-dihydro-1,2,4,5-tetrazine, (III), in which two molecules of (II) are joined in a *cis* pattern. Compound (I) has now been shown to have *trans* geometry (Fig. 1), with all atoms essentially coplanar (r.m.s. fit 0.038 Å), except for the N15-containing pyridyl ring, which is twisted by 14.1(1)° from this plane. Hence, there is not a genuine molecular inversion centre at the midpoint of N1—N11. The twist allows the formation of two independent intermolecular N—H···N hydrogen bonds (Table 2) which assemble the molecules into puckered chains along the *a* direction (Fig. 2). The possible intermolecular interactions N2—H2A···N5, N2—H2B···N11, N12—H12A···N1 and N12—H12B···N15 cannot be regarded as conventional hydrogen bonds. The angles at hydrogen lie in the range 99–109° and the angles at the acceptor N atom are 80–84°.

Two H atoms each were found on N2 and N12, coplanar with the main molecule and with H—N—H *ca* 120°. There is no evidence of H atoms attached to N1, N5, N11 or N15. Accepting 1.47 and 1.27 Å as typical

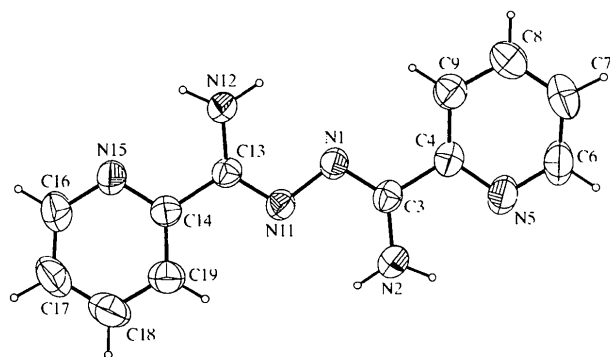


Fig. 1. The structure of (I) showing 50% probability displacement ellipsoids.

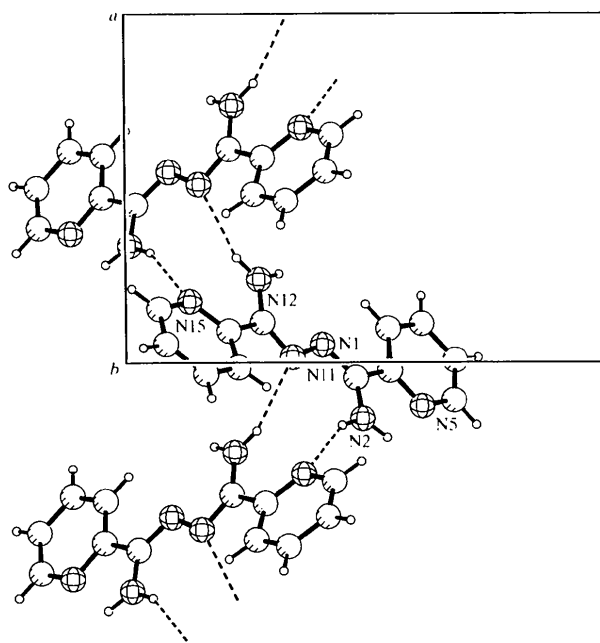


Fig. 2. The packing diagram for (I) showing chains in the *a* direction formed by intermolecular hydrogen bonds.

C—N and C=N distances (Hamilton & La Placa, 1968), C3—N2 and C13—N12, along with C4—N5, C6—N5, C14—N15 and C16—N15 in the pyridine residues, have approximately 50% double-bond character. These C=N bonds are some 20σ longer than C3—N1 and C13—N11 [1.296(2) and 1.298(2) Å, respectively], but the latter are still significantly longer than full double bonds. The N1—N11 bond [1.404(2) Å] is at the short end of the range of N—N single bonds taken from *International Tables for Crystallography* (1994, Vol. C). The corresponding distance in hydrazine is 1.46 Å (Baglen *et al.*, 1967).

The pattern of electron delocalization is broadly similar to that in the series of glyoxal(bisamidino)hydrazone salts, (IV), for which leukemia-inhibition properties have been reported (Hamilton & La Placa, 1968; Mutikainen *et al.*, 1986; Lumme *et al.*, 1986). The presence of an H atom on one of the hydrazine N atoms in (IV) does not greatly alter the heavy-atom framework. In the most precise determination, the C—N(H)N bond [1.339(2) Å] is longer than in (I), and the N—N bond shorter [1.364(2) Å].

Experimental

Crystals were prepared by K. M. Watson according to the method of Kubota *et al.* (1972).

Crystal data

C₁₂H₁₂N₆
M_r = 240.28

Mo *K*_α radiation
 $\lambda = 0.71073 \text{ \AA}$

Orthorhombic
Pbca
 $a = 9.4810(9) \text{ \AA}$
 $b = 19.8360(19) \text{ \AA}$
 $c = 13.1780(14) \text{ \AA}$
 $V = 2478.3(4) \text{ \AA}^3$
 $Z = 8$
 $D_x = 1.288 \text{ Mg m}^{-3}$
 D_m not measured

Cell parameters from 25 reflections
 $\theta = 13\text{--}15^\circ$
 $\mu = 0.085 \text{ mm}^{-1}$
 $T = 296 \text{ K}$
 Prism
 $0.50 \times 0.25 \times 0.22 \text{ mm}$
 Yellow

Data collection

Enraf–Nonius CAD-4 diffractometer
 ω - 2θ scans
 Absorption correction: none
 2306 measured reflections
 2174 independent reflections
 1528 reflections with $I > 2\sigma(I)$

$R_{\text{int}} = 0.012$
 $\theta_{\text{max}} = 24.97^\circ$
 $h = 0 \rightarrow 11$
 $k = -23 \rightarrow 1$
 $l = 0 \rightarrow 15$
 3 standard reflections
 frequency: 60 min
 intensity decay: 0.5%

Refinement

Refinement on F^2
 $R[F^2 > 2\sigma(F^2)] = 0.036$
 $wR(F^2) = 0.135$
 $S = 0.951$
 2174 reflections
 176 parameters
 H atoms treated by a mixture of independent and constrained refinement
 $w = 1/[\sigma^2(F_o^2) + (0.1P)^2]$
 where $P = (F_o^2 + 2F_c^2)/3$

$(\Delta/\sigma)_{\text{max}} = 0.011$
 $\Delta\rho_{\text{max}} = 0.139 \text{ e \AA}^{-3}$
 $\Delta\rho_{\text{min}} = -0.166 \text{ e \AA}^{-3}$
 Extinction correction: *SHELXL97*
 Extinction coefficient: 0.0073 (16)
 Scattering factors from *International Tables for Crystallography* (Vol. C)

Table 1. Selected geometric parameters (\AA , $^\circ$)

N1—N11	1.4045 (18)	N12—C13	1.345 (2)
N1—C3	1.2956 (19)	C13—C14	1.486 (2)
N2—C3	1.343 (2)	C14—C19	1.383 (2)
N11—C13	1.2982 (19)		
N11—N1—C3	111.55 (12)	N1—C3—N2	125.94 (15)
N1—N11—C13	112.02 (12)	N11—C13—N12	124.92 (15)
C3—N1—N11—C13	173.53 (13)	N12—C13—C14—N15	16.6 (2)
N2—C3—C4—N5	2.9 (2)		

Table 2. Hydrogen-bonding geometry (\AA , $^\circ$)

$D\cdots H\cdots A$	$D\cdots H$	$H\cdots A$	$D\cdots A$	$D\cdots H\cdots A$
N2—H2B \cdots N15 ⁱ	0.89 (2)	2.24 (2)	3.073 (2)	154.9 (17)
N12—H12B \cdots N11 ⁱⁱ	0.89 (2)	2.262 (19)	3.0918 (19)	154.8 (17)

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

Data collection: *CAD-4/PC* (Enraf–Nonius, 1993). Cell refinement: *CAD-4/PC*. Data reduction: *XCAD4* (Harms & Wocadlo, 1996). Program(s) used to solve structure: *SHELXS97* (Sheldrick, 1997b). Program(s) used to refine structure: *SHELXL97* (Sheldrick, 1997a). Molecular graphics: *PLUTON92* and *PLATON92* (Spek, 1992). Software used to prepare material for publication: *SHELXL97*.

Thanks are extended to Dr K. M. Watson for the gift of the title compound.

Supplementary data for this paper are available from the IUCr electronic archives (Reference: FG1463). Services for accessing these data are described at the back of the journal.

References

- Baglen, F. G., Bush, S. F. & Durig, J. R. (1967). *J. Chem. Phys.* **47**, 2104–2109.
- Enraf–Nonius (1993). *CAD-4/PC*. Version 1.2. Enraf–Nonius, Delft, The Netherlands.
- Hamilton, W. C. & La Placa, S. J. (1968). *Acta Cryst.* **B24**, 1147–1156.
- Harms, K. & Wocadlo, S. (1996). *XCAD4. Program for the Lp Correction of Enraf–Nonius Four-Circle Diffractometer Data*. University of Marburg, Germany.
- Kubota, S., Kirino, O., Koida, Y. & Miyake, K. (1972). *J. Pharm. Soc. Jpn.* **92**, 275–279.
- Lumme, P. O., Mutikainen, I. & Elo, H. O. (1986). *Acta Cryst.* **C42**, 1209–1211.
- Mutikainen, I., Elo, H. O. & Lumme, P. O. (1986). *J. Chem. Soc. Perkin Trans. 2*, pp. 291–293.
- Sheldrick, G. M. (1997a). *SHELXL97. Program for the Refinement of Crystal Structures*. University of Göttingen, Germany.
- Sheldrick, G. M. (1997b). *SHELXS97. Program for the Solution of Crystal Structures*. University of Göttingen, Germany.
- Spek, A. L. (1992). *PLATON92 and PLUTON92. Programs for Molecular Geometry Calculations*. University of Utrecht, The Netherlands.
- Watson, K. M. & Neilson, D. G. (1975). *The Chemistry of the Amidines and Imidates*, edited by S. Patai, p. 494. London: Wiley.

Acta Cryst. (1998). **C54**, 1925–1927

1,9a-Epoxy-1,2,3,4,4a,4b,5,8,8a,9a-decahydro-2-methyl-4-nitro-5,8-methano-9H-fluoren-9-one

HOONG-KUN FUN,^a KANDASAMY CHINNAKALI,^{a†} NIRMAL KUMAR HAZRA,^b DIPAKRANJAN MAL,^b GUR DAYAL NIGAM^c AND IBRAHIM ABDUL RAZAK^a

^a*X-ray Crystallography Unit, School of Physics, Universiti Sains Malaysia, 11800 USM, Penang, Malaysia*, ^b*Department of Chemistry, Indian Institute of Technology, Kharagpur 721 302, West Bengal, India*, and ^c*Department of Physics and Meteorology, Indian Institute of Technology, Kharagpur 721 302, West Bengal, India*. E-mail: hkfun@usm.my

(Received 13 March 1998; accepted 28 May 1998)

Abstract

The six-membered ring of the title compound, $\text{C}_{15}\text{H}_{17}\text{NO}_4$, adopts a half-chair conformation, and the

[†] On leave from: Department of Physics, Anna University, Chennai 600 025, India.