This work was supported in part by a grant from the Israeli Ministry of Science and Technology.

Lists of structure factors, anisotropic displacement parameters, H-atom coordinates and complete geometry have been deposited with the IUCr (Reference: CF1092). Copies may be obtained through The Managing Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

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Acta Cryst. (1996). C**52**, 2880–2882

3-(4-Nitrobenzyloximino)hexahydroazepin-2-one

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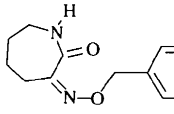
(*Received 19 February 1996; accepted 10 June 1996*)

Abstract

The title compound, $C_{13}H_{15}N_3O_4$, occurs as two stereoisomers. The *Z* and *E* isomers both display potent relaxant activity. Crystal structure analysis clearly demonstrates that the studied isomer, the less active form, adopts the *Z* conformation. The molecules are held together by N—H···O hydrogen bonds and intermolecular ring-to-ring interactions forming head-to-tail hydrogen-bonded dimers and a layered π-stacking arrangement.

Comment

This work is part of a general program aiming to synthesize new potential relaxant molecules. One derivative of the oximinocaprolactame family, 3-[*O*-(4-nitrobenzyl)oximino ether]hexahydroazepine-2-one (NOHA), displays promising anti-convulsive activity (Elfrom, 1995) of almost the same order as that of the reference relaxant agents theophilline and cromakalim. Cross-reference tests on NOHA showed that its activity depends strongly on the nature and/or the position of the different functional groups of the molecule. Thus, changing the position of NO_2 on the phenyl group in NOHA or substitution of the caprolactame, a seven-membered heterocycle (C_6NO), by a benzene ring seriously affects its activity.



Z-NOHA

Thin-layer chromatography and NMR studies showed that two stereoisomers, the *Z* and *E* forms, coexist in NOHA. Subsequent pharmacological tests showed that the *E* isomer is 1.5 times more active than the *Z* form. For structure–activity-correlation purposes, the geometrical properties of both isomers are of interest. Unfortunately, only single crystals of *Z* are available as efforts to grow the *E* isomers have so far been unsuccessful. However, it was decided to perform a crystal structure determination of the *Z* isomer with the assumption that the molecular structures of both isomers should be similar; the molecular geometry of the *Z* form could also be helpful in solving the structure of the *E* isomer by powder methods. The crystal structure solution confirms the *Z* conformation of the title compound. The molecules are packed in an extensive benzene–benzene π-stacking arrangement (ring–ring distance 3.4 Å) and as head-to-tail hydrogen-bonded dimers [N3···O4($1-x, -y, 1-z$) = 2.977 (6) Å].

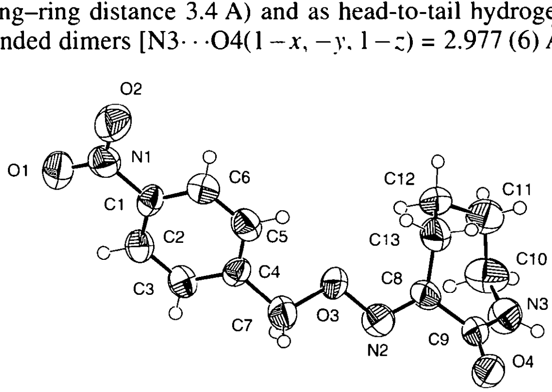


Fig. 1. The molecular structure of *Z*-NOHA showing the atom-labelling scheme with 50% probability displacement ellipsoids.

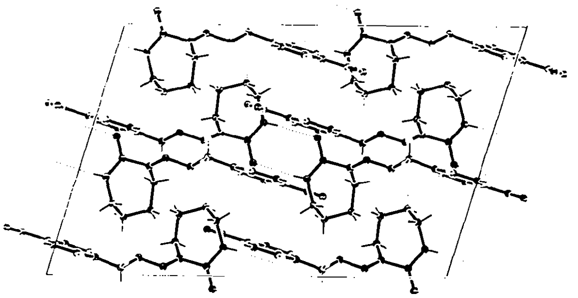


Fig. 2. View of the unit cell down the *b* axis of *Z*-NOHA; hydrogen bonds are represented by dashed lines.

Experimental

Crystals suitable for X-ray analysis were prepared by condensation of hexahydroazepine-2,3-dione with benzylhydroxylamine (Elfrom, 1995). The compound was recrystallized from methyl acetyl solution.

Crystal data

$C_{13}H_{15}N_3O_4$
$M_r = 277.28$
Monoclinic
$C2/c$
$a = 21.141\ (5)$ Å
$b = 9.610\ (3)$ Å
$c = 13.980\ (4)$ Å
$\beta = 107.33\ (8)°$
$V = 2711.3\ (13)$ Å^3
$Z = 8$
$D_x = 1.359$ Mg m^{-3}
D_m not measured

Cu $K\alpha$ radiation
$\lambda = 1.54180$ Å
Cell parameters from 23 reflections
$\theta = 10–24°$
$\mu = 0.860$ mm^{-1}
$T = 293\ (2)$ K
Needle
0.18 × 0.08 × 0.06 mm
Colourless

Data collection

CAD-4 diffractometer
ω scans
Absorption correction: none
4443 measured reflections
962 independent reflections
603 observed reflections $[I > 2\sigma(I)]$
$R_{\rm int} = 0.04$

$\theta_{\rm max} = 64.99°$
$h = -24 \rightarrow 22$
$k = 0 \rightarrow 11$
$l = 0 \rightarrow 16$
3 standard reflections monitored every 40 reflections frequency: 60 min intensity decay: 12%

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)] = 0.0406$
$wR(F^2) = 0.1077$
$S = 0.438$
962 reflections
182 parameters
H atoms riding
$w = 1/[\sigma^2(F_o^2) + (0.1706P)^2 + 1.9204P]$ where $P = (F_o^2 + 2F_c^2)/3$

$(\Delta/\sigma)_{\rm max} < 0.001$
$\Delta\rho_{\rm max} = 0.130$ e Å^{-3}
$\Delta\rho_{\rm min} = -0.152$ e Å^{-3}
Extinction correction: none
Atomic scattering factors from *International Tables for Crystallography* (1992, Vol. C, Tables 4.2.6.8 and 6.1.1.4)

Table 1. *Fractional atomic coordinates and equivalent isotropic displacement parameters (Å^2)*

$$U_{\rm eq} = (1/3)\Sigma_i\Sigma_j U_{ij}a_i^*a_j^*\mathbf{a}_i.\mathbf{a}_j.$$

	x	y	z	$U_{\rm eq}$
O1	−0.1296 (2)	−0.0324 (4)	0.3175 (3)	0.0834 (12)
O2	−0.1035 (2)	0.1815 (5)	0.3265 (4)	0.0940 (13)
O3	0.2229 (2)	0.0128 (4)	0.4318 (3)	0.0721 (10)
O4	0.4306 (2)	0.0298 (4)	0.5503 (3)	0.0695 (10)
N1	−0.0881 (2)	0.0613 (6)	0.3355 (3)	0.0661 (12)
N2	0.2906 (2)	−0.0249 (4)	0.4644 (3)	0.0617 (12)
N3	0.4237 (2)	0.0182 (4)	0.3871 (3)	0.0675 (12)
C1	−0.0175 (2)	0.0228 (5)	0.3681 (3)	0.0503 (11)
C2	0.0002 (2)	−0.1129 (5)	0.3859 (4)	0.0572 (13)
C3	0.0657 (2)	−0.1476 (5)	0.4167 (3)	0.0559 (12)
C4	0.1145 (2)	−0.0450 (5)	0.4316 (3)	0.0496 (12)
C5	0.0952 (3)	0.0907 (5)	0.4121 (4)	0.0613 (14)
C6	0.0291 (3)	0.1265 (5)	0.3810 (3)	0.0573 (13)
C7	0.1859 (2)	−0.0846 (6)	0.4697 (4)	0.0651 (15)
C8	0.3252 (2)	0.0633 (5)	0.4322 (3)	0.0573 (13)
C9	0.3983 (2)	0.0329 (5)	0.4619 (4)	0.0546 (13)
C10	0.3863 (3)	−0.0005 (7)	0.2816 (4)	0.085 (2)
C11	0.3494 (3)	0.1221 (7)	0.2298 (4)	0.076 (2)
C12	0.2868 (2)	0.1581 (6)	0.2573 (4)	0.0706 (15)
C13	0.3004 (3)	0.1883 (5)	0.3688 (4)	0.0682 (14)

Table 2. *Selected geometric parameters (Å, °)*

O1—N1	1.230 (5)	C2—C3	1.364 (6)
O2—N1	1.197 (5)	C3—C4	1.398 (6)
O3—N2	1.414 (4)	C4—C5	1.370 (6)
O3—C7	1.419 (5)	C4—C7	1.492 (6)
O4—C9	1.222 (5)	C5—C6	1.377 (7)
N1—C1	1.473 (6)	C8—C9	1.503 (7)
N2—C8	1.286 (6)	C8—C13	1.492 (6)
N3—C9	1.318 (6)	C10—C11	1.478 (8)
N3—C10	1.463 (7)	C11—C12	1.523 (7)
C1—C2	1.359 (7)	C12—C13	1.527 (7)
C1—C6	1.375 (7)		
N2—O3—C7	109.2 (3)	O1—N1—C1	118.3 (5)
O2—N1—O1	122.0 (5)	C8—N2—O3	110.2 (4)
O2—N1—C1	119.7 (5)	C9—N3—C10	125.9 (4)

Refinement was based on the set of 962 reflections with $I > 1.5\sigma(I)$ in view of the weak reflectivity and instability of the available crystal. H atoms were refined riding on their carrier atoms with one common isotropic displacement parameter.

Data collection and cell refinement: *CAD*-4 *Software* (Enraf–Nonius, 1989). Data reduction: *SDP* (Frenz, 1980). Programs used to solve the structure: *SHELXS*86 (Sheldrick, 1990) and *SHELXL*93 (Sheldrick, 1993). Molecular graphics: *Xtal-GX* (Hall & du Boulay, 1995) and *PLUTON* (Spek, 1992).

Lists of structure factors, anisotropic displacement parameters, H-atom coordinates and complete geometry have been deposited with the IUCr (Reference: SK1016). Copies may be obtained through The Managing Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

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Acta Cryst. (1996). C**52**, 2882–2883

1,1′-Diacetyl-2,2′-biimidazole

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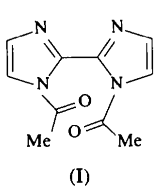
(*Received 29 May 1996; accepted 17 June 1996*)

Abstract

A crystallographic twofold rotation axis passes through the C—C bond joining the imidazole rings of the title compound, $C_{10}H_{10}N_4O_2$. The molecule crystallizes in a *cis* disposition. The planar acetyl group is twisted by 5.0 (3)° with respect to the imidazole ring and the two imidazole rings are tilted by 60.53 (5)° in relation to one another.

Comment

First studied for its antiprotozoal activity (Melloni *et al.*, 1975) and later incorporated into hexaaza macrocyclic metal complexes (Kandil & Collier, 1988), 1,1′-diacetyl-2,2′-biimidazole, (I), exhibits a *cis* conformation in the solid state, unlike all previously reported 1,1′-disubstituted 2,2′-biimidazole derivatives (Secondo, Barnett, Collier & Baughman, 1996; Sokal, Baikalova, Domnina & Poria-Koshits, 1992) which are *trans* oriented.



(I)

The bond lengths and angles of the 2,2′-biimidazole skeleton do not deviate significantly from those in the free molecule (Cromer, Ryan & Storm, 1987). The unfavorable *cis* conformation [dihedral angle 60.53 (5)°] is stabilized by intermolecular interactions between (*a*) the methyl group and an imidazole N atom[C5—H···N1(x, $-y$, $-\frac{1}{2}+z$), H···N = 2.521 (2) Å] and (*b*) an imidazole C—H and an O atom [C3—H···O($-\frac{1}{2}+x$, $\frac{1}{2}-y$, $-\frac{1}{2}+z$), H···O = 2.415 (2) Å]. The non-H atoms of each half molecule are nearly coplanar, with O1 showing the maximum deviation from the eight-atom least-squares plane [0.056 (1) Å].

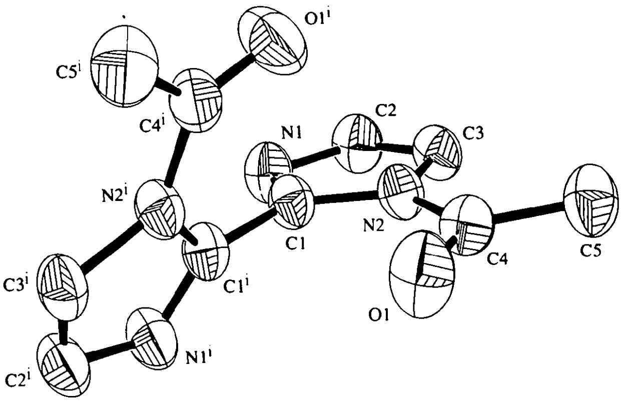


Fig. 1. View of the title molecule with displacement ellipsoids plotted at the 50% probability level.

Experimental

The preparation of the title compound has been reported previously (Melloni *et al.*, 1975). Crystals suitable for diffraction were grown by slow cooling of a hot, saturated acetic anhydride solution. Though the title material is susceptible to hydrolysis both as a solid and in acetic anhydride, none was noted during data collection as evidenced by the low variation of the check-reflection intensities.

Crystal data

$C_{10}H_{10}N_4O_2$
$M_r = 109.11$
Monoclinic
$C2/c$
$a = 11.987\,(2)$ Å
$b = 9.7183\,(15)$ Å
$c = 10.2177\,(12)$ Å
$\beta = 119.889\,(9)°$
$V = 1032.0\,(2)$ Å^3
$Z = 4$
$D_x = 1.404$ Mg m^{-3}
D_m not measured

Mo $K\alpha$ radiation
$\lambda = 0.71073$ Å
Cell parameters from 50 reflections
$\theta = 5.5$–$18.7°$
$\mu = 0.103$ mm^{-1}
$T = 293\,(2)$ K
Block cut from larger crystal
0.50 × 0.38 × 0.31 mm
Colorless

Data collection

Siemens $P3$ diffractometer
$\theta/2\theta$ scans
Absorption correction: none
1090 measured reflections
914 independent reflections
778 observed reflections [$I > 2\sigma(I)$]
$R_{\rm int} = 0.0077$

$\theta_{\rm max} = 25.07°$
$h = 0 \rightarrow 14$
$k = -3 \rightarrow 11$
$l = -12 \rightarrow 10$
3 standard reflections monitored every 50 reflections
intensity decay: 1.1%