O28B—C22B—C23B—C24B	59 (1)
O29B—C22B—C23B—O30B	23 (1)
O29B—C22B—C23B—C24B	−109 (1)
C22B—C23B—C24B—C25B	−164.6 (8)
O30B—C23B—C24B—C25B	60 (1)
C23B—C24B—C25B—O26B	−148.0 (9)
C23B—C24B—C25B—O27B	39 (1)

Data were collected using a fixed scan speed of 1.2° $\min^{-1}$ and a scan width of 1.2°. Structure solved by Patterson and direct methods for the difference structure. Refinement with full-matrix least squares on F^2 for all reflections. H atoms (except the carboxyl H atom, which was obtained from a difference Fourier synthesis) were calculated at geometrical positions and riding on their parent atom. The hydrogen malate anion was disordered. Both enantiomers are present at the same position and were refined with geometrical and similar U_{ij} restraints (*SADI* and *SIMU* in *SHELXL*93). The sum of the occupancy factors was constrained to 1.0. The occupancy factor for molecule *A* was 0.688 (2).

Data collection: *Y*290 (Hilger & Watts, 1969). Cell refinement: *XRAY*76 (Stewart *et al.*, 1976). Data reduction: *REDU*4 (Stoe & Cie, 1992). Program(s) used to solve structure: *DIRDIF* (Beurskens *et al.*, 1992). Program(s) used to refine structure: *SHELXL*93 (Sheldrick, 1993). Molecular graphics: *ORTEX*2.1 (McArdle, 1994). Software used to prepare material for publication: *PARST* (Nardelli, 1983*a*).

The authors thank Dr J. Tollenaere of Janssen Research Foundation (Beerse, Belgium) for providing a sample of the pizotifen malate.

Lists of structure factors, anisotropic displacement parameters, H-atom coordinates and complete geometry have been deposited with the IUCr (Reference: NA1139). 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. (1995). C**51**, 1438–1440

1,3,4,10b-Tetrahydro-4-phenylpyrido[2,1-*a*]-isoindole-2,6-dione

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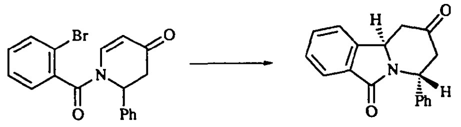
(*Received* 18 *October* 1994; *accepted* 24 *January* 1995)

Abstract

The piperidone ring in the title compound, $C_{18}H_{15}NO_2$, is in a chair conformation. Coordination at the N atom is planar. The phenyl substituent and the bridgehead H atom at the ring junction are *cis* and in axial orientations with respect to the piperidone ring.

Comment

In the course of studies aimed at the diastereoselective synthesis of alkaloids containing the indolizidine nucleus, the substituted dihydropyridone (I) was treated with tributylstannane in benzene to afford (II) as the sole cyclized product (Beckwith, Joseph & Mayadunne, 1993).



(I) (II)

The structure and stereochemistry of (II) were predicted from ^1H and ^{13}C NMR spectra, and molecular-mechanics calculations on the transition structure. These predictions have now been validated by the present single-crystal X-ray analysis.

Cremer & Pople (1975) puckering parameters for the piperidone ring, calculated using *RING*88 (Cremer, 1988), indicate that it is in a chair conformation ($q_2 = 0.059$, $q_3 = 0.493$ Å, $\varphi_2 = 83.2°$ and $\theta_2 = 6.8°$). The phenyl substituent at C12 and the H atom at the ring junction (C8) are *cis* and in axial orientations with respect to the piperidone ring, while the C7—C8 bond formed during the cyclization is equatorial. The five-membered ring has a shallow twist conformation

(q_2 = 0.027 Å and φ_2 = 339.9°) (Cremer, 1980). All atoms of the isoindolone unit with the exception of C8 are coplanar with a maximum deviation of these atoms from their least-squares plane of 0.007 (4) Å; C8 itself lies only 0.044 (2) Å from this plane. The angles around the N atom sum to 360.0°, a feature also observed in 13b-phenyl-7*H*-benz[*de*]isoindolo-[1,2-*a*]isoquinolin-9(13*H*)-one (Hitchings, Helliwell & Vernon, 1990) which, like the present compound, has an additional six-membered ring fused at N and C8. In 6,7-benzo-3-benzyl-5-hydroxy-4-phenyl-1,3-diazabicyclo[3.3.0]octan-8-one (Coyle, Smart, Challiner & Haws, 1985), where the additional ring is five-membered, the angles sum to only 339.3°. $\theta_{\rm C}$ and $\theta_{\rm N}$, which measure the pyramidalization at C8 and N, respectively (Winkler & Dunitz, 1971; Cieplak, 1994) are 0.6 and 1.1°, respectively, for (II). Bond lengths and angles closely parallel the corresponding dimensions in 3-hydroxyphthalimidine (Ohrt, Tsoucaris-Kupfer & Lechat, 1978), 2-(3-methyl-1-oxoisoindolin-2-yl)-butyramide (Carlström, Hacksell, Jönsson & Söderholm, 1983) and 1,2-dihydro-1-hydroxy-(2-pyrazinyl)-1*H*-3-isoindolone (Rodier, Martin, Miocque, Mettey & Vierfond, 1988).

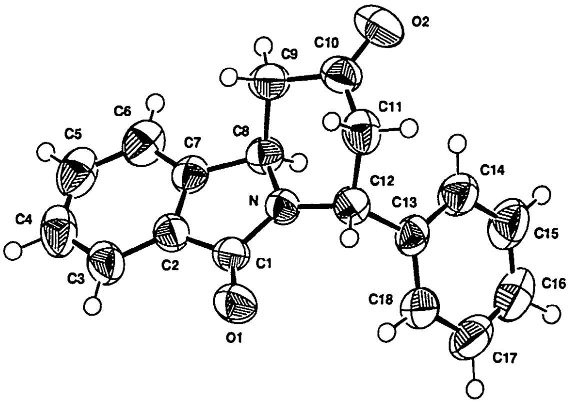


Fig. 1. View of $C_{18}H_{15}NO_2$ showing the labelling of the non-H atoms. Displacement ellipsoids are drawn at the 50% probability level; H atoms are drawn as small spheres of arbitrary radii.

Experimental

Crystal data

$C_{18}H_{15}NO_2$
M_r = 277.33
Triclinic
$P\bar{1}$
a = 8.531 (2) Å
b = 9.406 (1) Å
c = 9.759 (2) Å
α = 75.92 (1)°
β = 78.44 (2)°
γ = 68.71 (1)°
V = 702.3 (2) Å^3
Z = 2
D_x = 1.311 Mg m^{-3}

Cu $K\alpha$ radiation
λ = 1.5418 Å
Cell parameters from 25 reflections
θ = 46.5–49.7°
μ = 0.65 mm^{-1}
T = 293 K
Multifacetted
0.30 × 0.16 × 0.11 mm
Colourless

Data collection

Rigaku AFC-6*R* diffractometer
$\theta/2\theta$ scans
Absorption correction: by integration from crystal shape
$T_{\rm min}$ = 0.88, $T_{\rm max}$ = 0.93
2509 measured reflections
2079 independent reflections
1714 observed reflections [$I > 3\sigma(I)$]

$R_{\rm int}$ = 0.031
$\theta_{\rm max}$ = 60.0°
$h = -8 \rightarrow 9$
$k = 0 \rightarrow 10$
$l = -10 \rightarrow 10$
3 standard reflections monitored every 150 reflections intensity decay: 10%

Refinement

Refinement on F
R = 0.047
wR = 0.069
S = 1.789
1714 reflections
235 parameters
Only coordinates of H atoms refined
$w = 1/[\sigma^2(F) + 0.0009F^2]$

$(\Delta/\sigma)_{\rm max}$ = 0.031
$\Delta\rho_{\rm max}$ = 0.18 e Å^{-3}
$\Delta\rho_{\rm min}$ = −0.28 e Å^{-3}
Extinction correction: none
Atomic scattering factors from *International Tables for X-ray Crystallography* (1974), Vol. IV, Tables 2.2*B* and 2.3.1

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	0.7576 (2)	0.3675 (2)	0.8653 (2)	0.060 (2)
O2	0.2570 (2)	0.9943 (2)	0.6244 (2)	0.078 (2)
N	0.6367 (2)	0.6084 (2)	0.7334 (2)	0.040 (1)
C1	0.7346 (3)	0.4565 (2)	0.7505 (2)	0.042 (2)
C2	0.8027 (2)	0.4233 (2)	0.6053 (2)	0.043 (2)
C3	0.9092 (3)	0.2868 (3)	0.5618 (3)	0.056 (2)
C4	0.9530 (3)	0.2891 (3)	0.4164 (3)	0.064 (3)
C5	0.8929 (3)	0.4248 (4)	0.3187 (3)	0.064 (3)
C6	0.7882 (3)	0.5617 (3)	0.3624 (3)	0.056 (2)
C7	0.7436 (2)	0.5590 (2)	0.5073 (2)	0.043 (2)
C8	0.6359 (3)	0.6889 (2)	0.5847 (2)	0.041 (2)
C9	0.4535 (3)	0.7639 (3)	0.5498 (2)	0.050 (2)
C10	0.3468 (3)	0.8615 (3)	0.6582 (3)	0.050 (2)
C11	0.3584 (3)	0.7809 (3)	0.8120 (2)	0.051 (2)
C12	0.5396 (3)	0.6931 (2)	0.8479 (2)	0.043 (2)
C13	0.6337 (3)	0.7931 (2)	0.8735 (2)	0.043 (2)
C14	0.5904 (3)	0.9509 (3)	0.8214 (3)	0.056 (2)
C15	0.6879 (4)	1.0340 (3)	0.8389 (3)	0.067 (3)
C16	0.8260 (4)	0.9624 (4)	0.9099 (3)	0.068 (3)
C17	0.8679 (3)	0.8063 (4)	0.9679 (3)	0.069 (3)
C18	0.7720 (3)	0.7213 (3)	0.9494 (3)	0.055 (2)

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

O1—C1	1.227 (2)	C7—C8	1.504 (3)
O2—C10	1.209 (3)	C8—C9	1.528 (3)
N—C1	1.357 (2)	C9—C10	1.507 (3)
N—C8	1.465 (2)	C10—C11	1.513 (3)
N—C12	1.467 (3)	C11—C12	1.531 (3)
C1—C2	1.481 (3)	C12—C13	1.528 (4)
C2—C3	1.386 (3)	C13—C14	1.381 (3)
C2—C7	1.389 (3)	C13—C18	1.388 (3)
C3—C4	1.390 (4)	C14—C15	1.388 (5)
C4—C5	1.389 (4)	C15—C16	1.358 (4)
C5—C6	1.385 (4)	C16—C17	1.379 (5)
C6—C7	1.385 (3)	C17—C18	1.395 (5)
C1—N—C8	113.8 (2)	C7—C8—C9	114.8 (2)
C1—N—C12	126.0 (2)	C8—C9—C10	110.1 (2)
C8—N—C12	120.2 (1)	O2—C10—C9	122.3 (2)
O1—C1—N	125.3 (2)	O2—C10—C11	122.5 (2)
O1—C1—C2	128.5 (2)	C9—C10—C11	115.3 (2)

N—C1—C2	106.2 (2)	C10—C11—C12	114.4 (2)
C1—C2—C3	130.2 (2)	N—C12—C11	108.7 (2)
C1—C2—C7	108.6 (2)	N—C12—C13	109.7 (2)
C3—C2—C7	121.3 (2)	C11—C12—C13	115.4 (2)
C2—C3—C4	117.8 (2)	C12—C13—C14	123.1 (2)
C3—C4—C5	120.8 (3)	C12—C13—C18	118.4 (2)
C4—C5—C6	121.4 (3)	C14—C13—C18	118.5 (3)
C5—C6—C7	117.8 (2)	C13—C14—C15	120.6 (2)
C2—C7—C6	121.0 (2)	C14—C15—C16	120.7 (3)
C2—C7—C8	109.6 (2)	C15—C16—C17	119.8 (4)
C6—C7—C8	129.5 (2)	C16—C17—C18	120.0 (3)
N—C8—C7	101.7 (1)	C13—C18—C17	120.4 (2)
N—C8—C9	109.6 (2)		

C8—N—C1—O1	178.1 (2)	C1—C2—C7—C8	1.1 (3)
C8—N—C1—C2	−2.5 (3)	C2—C7—C8—N	−2.4 (3)
C12—N—C1—O1	−0.7 (4)	N—C8—C9—C10	51.8 (3)
C1—N—C8—C7	3.1 (3)	C8—C9—C10—O2	129.9 (3)
C12—N—C8—C9	−56.1 (3)	C8—C9—C10—C11	−50.9 (3)
C8—N—C12—C11	50.4 (3)	O2—C10—C11—C12	−133.2 (3)
O1—C1—C2—C7	−179.9 (3)	C9—C10—C11—C12	47.5 (3)
N—C1—C2—C7	0.8 (3)	C10—C11—C12—N	−43.1 (3)

The θ-scan width was $(1.5 + 0.3\tan\theta)^\circ$, with θ-scan rate $16^\circ\ \mathrm{min}^{-1}$ with up to four scans on weak reflections and background counts of one quarter of scan time on each end of every scan.

Data collection: *MSC/AFC Diffractometer Control Software* (Molecular Structure Corporation, 1992). Cell refinement: *MSC/AFC Diffractometer Control Software*. Data reduction: *Xtal*3.0 *DIFDAT*, *ABSORB*, *SORTRF*, *ADDREF* (Hall & Stewart, 1990). Program(s) used to solve structure: *SHELXS*86 (Sheldrick, 1985). Program(s) used to refine structure: *Xtal*3.0 *CRYLSQ*. Molecular graphics: *Xtal*3.0. Software used to prepare material for publication: *Xtal*3.0 *BONDLA*, *CIFIO*.

Lists of structure factors, anisotropic displacement parameters, H-atom coordinates and complete geometry have been deposited with the IUCr (Reference: KH1018). 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. (1995). C**51**, 1440–1442

4-Méthyl-1,2,4-triazolidine-3,5-dione (Méthylurazole)

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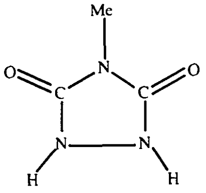
(*Reçu le* 4 *octobre* 1994, *accepté le* 3 *janvier* 1995)

Abstract

The title structure, 4-methyl-1,2,4-triazolidine-3,5-dione, (methylurazole), $C_3H_5N_3O_2$, has been solved in order to understand the influence of intermolecular hydrogen bonds on the conformation and packing mode. In the crystal, the molecules exist in the diketo form. The ring adopts a geometry between an envelope and a half-chair conformation. The two adjacent H atoms of the ring are in a *trans* position. The molecules are stacked in parallel planes and each molecule is linked to two others by two N—H···O hydrogen bonds which determine the formation of the eight-membered pseudocycle.

Commentaire

Les dérivés de l'urazole peuvent présenter un polymorphisme moléculaire lié à la possibilité d'existence de plusieurs conformations (enveloppe, demi-chaise, plane) et de formes tautomères (dicétone, cétone–énol, énol–énol). L'un des objectifs de notre laboratoire étant d'isoler et d'identifier ces espèces que nous voulons générer photochimiquement, nous nous sommes proposés tout d'abord d'effectuer la détermination structurale du méthylurazole (I). Ce composé sera comparé essentiellement à l'urazole (II) (Belaj, 1992).



(I)

En ce qui concerne les longueurs de liaison, les valeurs sont concordantes entre les composés (I) et (II) sauf pour N2—C3 où cette distance est plus petite dans le composé (I) avec un écart de 0,02 Å. Quant à la longueur N4—C4, égale à 1,457 (4) Å, elle correspond à celle entre un carbone sp^3 et un azote sp^2 et est aussi compatible à celles qu'on trouve dans des composés

Acta Crystallographica Section C
ISSN 0108-2701 ©1995