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8-Methyl-8-azabicyclo[4.3.1]decane-10-spiro-5′-hydantoin*†

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Abstract. $C_{12}H_{19}N_3O_2$, monoclinic, $P2_1/n$, $a = 13{\cdot}092\,(1)$, $b = 13{\cdot}453\,(1)$, $c = 6{\cdot}986\,(1)$ Å, $\beta = 90{\cdot}80\,(1)^\circ$, $Z = 4$, $d_x = 1{\cdot}273$ Mg m^{-3}, μ(Mo $K\alpha$) = 0·098 mm^{-1}. Final $R = 0{\cdot}079$. The cyclohexane ring has a chair conformation and the cycloheptane ring a conformation intermediate between a twist chair (C_2) and a chair (C_s).

Introduction. The structure of the title compound was determined as the sixth in a series of heterocyclic spiro hydantoin derivatives with interesting pharmacological properties (Gonzalez-Trigo, Avendaño & Martinez-Moreno, 1974). The results of the five other structures are available (Smith-Verdier, Florencio & García-Blanco, 1977, 1979; Florencio, Smith-Verdier & García-Blanco, 1978*a*,*b*; P. Smith-Verdier, F. Florencio & S. García-Blanco, unpublished). In the present compound the substituents and their positions in the bicyclic portion have been changed to study the effect of these changes on the conformation. Crystals were supplied by Professor Galvez (Facultad de Farmacia, Universidad Complutense de Madrid).

The space group was assigned uniquely from the systematic absences $h0l$, $h + l = 2n + 1$; $0k0$, $k = 2n + 1$. Intensities were collected on a Philips PW 1100 four-circle diffractometer by the ω–2θ scan technique between 2 and 30° in θ with a crystal 0·30 × 0·45 × 0·32 mm and graphite-monochromated Mo $K\alpha$ radiation. Of the 3701 reflexions measured, 2626 were classed as observed [$I > 2\sigma(I)$, where $\sigma(I)$ was calculated from counting statistics]. Intensities were corrected for Lorentz and polarization factors but not for absorption.

The structure was solved with *MULTAN* (Main, Woolfson, Lessinger, Germain & Declercq, 1974), with the 200 largest E values, and the subsequent electron density synthesis. All H atoms were located in a difference map. Least-squares refinement converged to $R = 0{\cdot}079$. Unit weight was assigned to each reflexion. A final difference synthesis showed no peaks >0·03 e Å^{-3}. Scattering factors were taken from *International Tables for X-ray Crystallography* (1974). Positional parameters are listed in Table 1. The atomic numbering is shown in Fig. 1. Bond lengths and angles are listed in Table 2.‡

Table 1. *Fractional atomic coordinates* (×10^4, *for* H × 10^3)

	x	y	z
C(1)	6218 (1)	8594 (1)	3790 (3)
C(2)	6116 (1)	7765 (1)	2303 (3)
C(3,5′)	5281 (1)	7027 (1)	2903 (3)
C(4)	5452 (1)	6655 (1)	4972 (3)
C(5)	5646 (2)	7525 (2)	6341 (3)
N(6)	6450 (1)	8212 (1)	5714 (2)
C(7)	7167 (2)	7303 (2)	1811 (4)
C(8)	7517 (2)	6312 (2)	2553 (6)
C(9)	7313 (2)	5965 (2)	4476 (6)
C(10)	6242 (2)	5808 (2)	5149 (5)
C(11)	6490 (2)	9063 (2)	7043 (4)
N(1′)	5145 (1)	6217 (3)	1514 (3)
C(2′)	4203 (2)	6180 (2)	743 (4)
N(3′)	3649 (1)	6957 (1)	1489 (3)
C(4′)	4210 (1)	7515 (1)	2739 (3)
O(1)	3865 (1)	5581 (1)	−415 (4)
O(2)	3908 (1)	8244 (1)	3552 (2)
H(11)	549 (2)	858 (2)	831 (4)
H(12)	676 (3)	904 (1)	340 (3)
H(2)	592 (3)	811 (4)	114 (3)
H(4)	484 (2)	636 (2)	541 (3)
H(51)	593 (3)	722 (2)	763 (2)
H(52)	499 (2)	790 (2)	650 (3)
H(71)	767 (4)	784 (1)	239 (3)
H(72)	733 (5)	736 (3)	49 (1)
H(81)	838 (4)	630 (4)	241 (1)
H(82)	717 (1)	572 (5)	153 (2)
H(91)	764 (2)	530 (4)	470 (1)
H(92)	762 (2)	662 (3)	552 (4)
H(101)	623 (1)	560 (1)	636 (4)
H(102)	611 (2)	519 (3)	417 (1)
H(111)	716 (2)	954 (3)	664 (3)
H(112)	592 (2)	944 (4)	707 (3)
H(113)	670 (2)	883 (3)	833 (4)
H(1′)	556 (3)	580 (2)	135 (2)
H(3′)	292 (2)	703 (3)	123 (2)

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* The Conformation of Heterocyclic Spiro Compounds. VI.

† Note that the atomic numbering used in this paper is arbitrary and does not correspond to that used in naming the compound.

‡ Lists of structure factors and anisotropic thermal parameters have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 34518 (27 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

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Table 2. *Bond lengths* (Å) *and angles* (°) *for non-hydrogen atoms with e.s.d.'s in parentheses*

C(1)–C(2)	1·529 (4)
C(2)–C(3,5′)	1·539 (3)
C(3,5′)–C(4)	1·543 (4)
C(4)–C(5)	1·530 (4)
C(5)–N(6)	1·473 (3)
N(6)–C(1)	1·467 (3)
N(6)–C(11)	1·474 (4)
C(2)–C(7)	1·552 (4)
C(7)–C(8)	1·501 (4)
C(8)–C(9)	1·450 (6)
C(9)–C(10)	1·500 (5)
C(10)–C(4)	1·543 (4)
C(3,5′)–N(1′)	1·468 (3)
N(1′)–C(2′)	1·340 (4)
C(2′)–N(3′)	1·379 (4)
N(3′)–C(4′)	1·360 (3)
C(4′)–C(3,5′)	1·551 (3)
C(2′)–O(1)	1·220 (4)
C(4′)–O(2)	1·203 (3)

C(2)–C(1)–N(6)	112·4 (2)
C(1)–C(2)–C(3,5′)	109·8 (2)
C(1)–C(2)–C(7)	112·0 (2)
C(3,5′)–C(2)–C(7)	116·0 (2)
C(2)–C(3,5′)–C(4)	111·7 (2)
C(2)–C(3,5′)–C(4′)	110·6 (2)
C(2)–C(3,5′)–N(1′)	112·1 (2)
C(4)–C(3,5′)–C(4′)	108·9 (2)
C(4)–C(3,5′)–N(1′)	113·1 (2)
C(3,5′)–C(4)–C(5)	111·0 (2)
C(3,5′)–C(4)–C(10)	113·7 (2)
C(5)–C(4)–C(10)	114·1 (2)
C(4)–C(5)–N(6)	114·1 (2)
C(5)–N(6)–C(1)	110·7 (2)
C(5)–N(6)–C(11)	108·6 (2)
C(1)–N(6)–C(11)	108·1 (2)
C(2)–C(7)–C(8)	123·1 (3)
C(7)–C(8)–C(9)	123·2 (3)
C(8)–C(9)–C(10)	121·3 (3)
C(9)–C(10)–C(4)	120·0 (3)
C(3,5′)–N(1′)–C(2′)	113·3 (3)
N(1′)–C(2′)–N(3′)	107·8 (3)
N(1′)–C(2′)–O(1)	127·9 (3)
N(3′)–C(2′)–O(1)	124·4 (3)
C(2′)–N(3′)–C(4′)	112·3 (2)
N(3′)–C(4′)–C(3,5′)	107·1 (2)
N(3′)–C(4′)–O(2)	125·3 (3)
C(3,5′)–C(4′)–O(2)	127·8 (2)
N(1′)–C(3,5′)–C(4′)	99·5 (2)

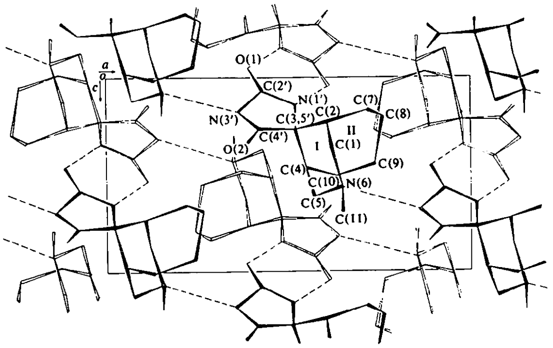

Fig. 1. Projection of the structure along *b*.

Discussion. The molecule contains a six-membered (I) and a seven-membered ring (II), joined by a common C(2)–C(3,5′)–C(4) bridge which has a hydantoin ring attached to its central spiranic C(3,5′).

Ring (I) has a chair conformation. The assymetry parameters (Duax & Norton, 1975) show that the rotational symmetry is dominant [C_2 axis intersecting C(3,5′)–C(4) and C(1)–N(6), with $\Delta C_2^{(3,4)} = 1{\cdot}12$, $\Delta C_2^{(1,2)} = 2{\cdot}61$ and $\Delta C_s^{(2)} = 7{\cdot}01$ °]. The deviations of C(3,5′) and N(6) from the least-squares plane through C(1), C(2), C(4) and C(5) are 0·634 (2) and –0·688 (2) Å respectively.

The seven-membered ring has a very distorted conformation. Signs for the torsion angles (Table 3) show that the ring does not adopt the boat conformation. According to Hendrickson (1961) both the chair and boat forms of cycloheptane are flexible and may undergo pseudorotation, and in each family the twist form, with an axis of symmetry C_2, is the most stable. Inspection of molecular models reveals that the seven-membered ring is relatively rigid, mainly in the part corresponding to the bridge, the opposite part of the molecule being the more distorted. In an ideal molecule with the chair–chair conformation the non-bonded C(8)···N(6) and C(9)···N(6) contacts would be 2·29 Å and in the present compound these distances are 3·667 (5) and 3·345 (4) Å respectively; this increase shows the strong interaction between these non-bonded atoms, which could be due to the bulky hydantoin ring attached to the spiranic C atom. Thus ring (II) adopts a conformation intermediate between the symmetrical forms. The lowest asymmetry parameter is $\Delta C_s^{10} = 24$ °.

The valence angles in ring (I) are slightly larger than tetrahedral, and are similar to those in other cyclohexane rings in the compounds already mentioned. The valency angles in ring (II) are larger than tetrahedral. The largest values correspond to the more flattened part and, as a result, C(8)–C(9) is significantly short.

The hydantoin ring is planar as in the other compounds of this series.

A projection along *b* showing the hydrogen bonds is given in Fig. 1. The methyl group attached to N(6) is equatorial. The configuration of N(6) is pyramidal. N(6) accepts a proton from N(3′) of the hydantoin ring to form a hydrogen bond. A second hydrogen bond is N(1′)–H···O(1), which forms around a symmetry center. The geometries of these hydrogen bonds are shown in Table 4. Other intermolecular contacts correspond to normal van der Waals interactions.

Table 3. *Torsion angles* (°) *involving non-hydrogen atoms*

The sign convention is as defined by Klyne & Prelog (1960).

C(1)–C(2)–C(3,5′)–C(4)	–52·1 (2)
C(2)–C(3,5′)–C(4)–C(5)	49·1 (2)
C(3,5′)–C(4)–C(5)–N(6)	–51·3 (2)
C(4)–C(5)–N(6)–C(1)	55·9 (2)
C(5)–N(6)–C(1)–C(2)	–58·9 (2)
N(6)–C(1)–C(2)–C(3,5′)	57·5 (2)
C(2)–C(7)–C(8)–C(9)	–38·3 (4)
C(7)–C(8)–C(9)–C(10)	62·2 (4)
C(8)–C(9)–C(10)–C(4)	–53·5 (4)
C(9)–C(10)–C(4)–C(3,5′)	58·3 (3)
C(10)–C(4)–C(3,5′)–C(2)	–81·3 (2)
C(4)–C(3,5′)–C(2)–C(7)	76·0 (2)
C(3,5′)–C(2)–C(7)–C(8)	23·3 (3)

Table 4. *Interatomic distances* (Å) *and angles* (°) *in the hydrogen bonds*

a b c	*ab*	*bc*	*ac*	∠*abc*
N(1′)—H···O(1) $(-x+1, -y+1, -z)$	0·792 (2)	2·108 (2)	2·854 (3)	157·3 (2)
N(3′)—H···N(6) $(x-\frac{1}{2}, -y+\frac{3}{2}, z-\frac{1}{2})$	0·981 (2)	1·974 (2)	2·930 (3)	164·4 (2)

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Structure of 3a,5-Diethyl-4-oxo-6,6a-diphenyl-3-(2,4,6-trimethylphenyl)-3a,4-dihydrocyclopenta[2,3-*d*]isoxazoline (TPDCI)

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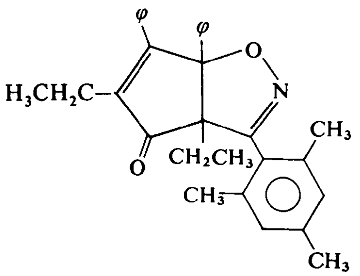
Abstract. $C_{31}H_{31}NO_2$, $M_r = 449·566$, monoclinic, $P2_1/n$, $a = 19·440\ (2)$, $b = 14·681\ (1)$, $c = 8·900\ (1)$ Å, $\beta = 95·02\ (1)°$, $Z = 4$, $V = 2530·16$ Å^3, λ(Mo $K\alpha$) = 0·71069 Å, $F(000) = 960$, $D_c = 1·178$, $D_m = 1·164$ Mg m^{-3}, m.p. 468–469 K, $\mu = 0·078$ mm^{-1}; 1223 independent non-zero reflexions. The structure was solved by direct phase determination with *MULTAN*. The positional and vibrational parameters, with anisotropic temperature factors for the non-hydrogen atoms, were refined by full-matrix least-squares calculations to a final $R = 0·068$. The molecule of the compound has a cyclopentenone–isoxazoline structure. To the two central five-membered isoxazoline and cyclopentenone rings, at an angle of 113·20°, are linked three benzene rings and two ethyl groups with different orientations. Unlike other isoxazoline derivatives, the isoxazoline ring in this case is almost perpendicular to the trimethyl-substituted benzene ring, instead of forming a roughly coplanar system with it.

Introduction. In two previous papers (Rodiou, Kokkou & Rentzeperis, 1978; Stergiou, Kokkou & Rentzeperis, 1978) the structures of two new isoxazoline derivatives, prepared at the Laboratory of Organic Chemistry of the Aristotle University of Thessaloniki (Alexandrou & Argyropoulos, 1977), were investigated. The present structure determination of TPDCI is the third of a series under investigation.



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