atoms to one carboxylic oxygen O(2) and to three water oxygen atoms, 2O(*W*2) and O(*W*3). On the other hand, all the hydrogen atoms of water molecules participate in O—H···O hydrogen bonds. As given in Table 2, the geometries of the hydrogen bonds are reasonable, except for the H(7)···H(7^{iii}) distance (1·4 Å), which is too short to be accepted. Such an undesirable approach of two H(7) atoms suggests that the water molecule *W*2 must be orientationally disordered with occupancy 0·5 for H(7). The so-called half-hydrogen model was adopted in the final calculations of least-squares refinement and Fourier syntheses, although the remaining half-hydrogen atom could not be located certainly.

The rather large *R* value in the present study may be attributed mainly to the relatively large amplitude of thermal motion of the chlorine atoms. In fact, no ^{35}Cl NQR signal was detected in the wide temperature region 100–300 K, indicating that all the CCl_3 groups are rapidly reorienting about their threefold axes (Hashimoto, 1986). Additionally, molecular motion of the ammonium ion has been confirmed from the ^{1}H NMR results.

References

Golič, L. & Lazarini, F. (1975). *Cryst. Struct. Commun.* **4**, 487–490.
Hashimoto, M. (1986). Private communication.
International Tables for X-ray Crystallography (1974). Vol. IV, pp. 71–151. Birmingham: Kynoch Press. (Present distributor D. Reidel, Dordrecht.)
Johnson, C. K. (1976). *ORTEP*II. Report ORNL-5138. Oak Ridge National Laboratory, Tennessee.
Kiriyama, H. (1962). *Bull. Chem. Soc. Jpn*, **35**, 1199–1205.
Main, P., Hull, S. E., Lessinger, L., Germain, G., Declercq, J.-P. & Woolfson, M. M. (1978). *MULTAN78. A System of Computer Programs for the Automatic Solution of Crystal Structures from X-ray Diffraction Data.* Univs. of York, England, and Louvain, Belgium.
The Universal Crystallographic Computing System–Osaka (1979). The Computation Center, Osaka Univ.
Wei, K.-T. & Ward, D. L. (1977). *Acta Cryst.* **B33**, 522–526.

Acta Cryst. (1987). C**43**, 1177–1179

Structure of the *E* Isomer of 5-Benzylidene-1-methyl-2,4-imidazolidinedione

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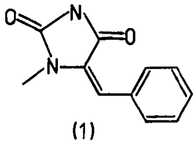
Abstract. $C_{11}H_{10}N_2O_2$, $M_r = 202\cdot22$, orthorhombic, *Pcan*, $a = 7\cdot184\,(7)$, $b = 20\cdot330\,(9)$, $c = 13\cdot532\,(9)$ Å, $V = 1976\cdot4$ Å^3, $Z = 8$, $D_m = 1\cdot33\,(3)$, $D_x = 1\cdot35$ g cm^{-3}, $\lambda(\text{Mo } K\alpha) = 0\cdot7107$ Å, $\mu = 0\cdot58$ cm^{-1}, $F(000) = 848$, room temperature, final $R = 0\cdot069$ for 868 $[I > 2\sigma(I)]$ independent observed reflections. The molecule, also known as 5-benzylidene-1-methylhydantoin, is essentially flat with the two planes of non-H atoms (the 2,4-imidazolidinedione and the benzene rings) at an angle of 5·7 (3)°. As is usual with most hydantoins containing an imine H and two O atoms, hydrogen bonds are observed with N(3)—H(31)···O(14) and H(31)···O(14) at 2·81 (1) and 1·82 (6) Å respectively.

Introduction. Compounds of 2,4-imidazolidinedione (also known as hydantoin) are well known (*Kirk-Othmer Encyclopedia of Chemical Technology*, 1980). However, only a few such compounds contain a methylene linkage at the 5-position. In previous structure determinations (Drew, Mok, Ang & Tan, 1987*a*,*b*) it was reported that the *Z* isomer of 5-[(4-methoxyphenyl)methylene]-2,4-imidazolidinedione was flat within 0·30 Å despite the presence of strain caused by the proximity of two H atoms, while in the *Z* isomer of 5-benzylidene-1-methylhydantoin the five-membered ring was rotated 45·9 (3)° from the plane of the phenyl ring. The structure of the *E* isomer of the latter molecule (1) is now reported.



(1)

Experimental. Compound (1) prepared as described elsewhere (Tan, Ang & Fong, 1986). Density measured by flotation in CCl_4/ligroin. Space group *Pcan* (non-standard setting of *Pbcn* with $a \rightarrow b$, $b \rightarrow a$, $c \rightarrow \bar{c}$).

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Precession photograph established preliminary cell constants. Crystal of approximate size 0·3 × 0·3 × 0·5 mm mounted on Stoe Stadi-2 diffractometer to rotate about the *a* axis. Cell dimensions by measurement of about 20 high-angle axial reflections (θ range 30 to 40°). Intensity data collected *via* variable-width ω scan, background counts 20 s, step-scan rate 0·033° s^{-1}, width (°) (2·0 + $\sin\mu/\tan\theta$). Absorption and extinction corrections not applied. Standard reflections *h*34 measured every 20 measurements for each layer: about 50% degradation in intensity observed. 1906 data measured with $2\theta_{\max}$ 50° ($0 \le h \le 8$, $0 \le k \le 23$, $0 \le l \le 16$). 868 data with $I > 2\sigma(I)$ used in subsequent calculations. Structure solved by direct methods using *SHELX*76 (Sheldrick, 1976). Each layer given separate scaling factor. These were refined together with atoms given isotropic thermal parameters and then fixed. Non-H atoms were then refined anisotropically before placing H atoms in tetrahedral or trigonal positions. Methyl group refined as rigid group. Thermal parameters of H atoms on N(3) and C(6) allowed to refine individually; those on phenyl ring and methyl group constrained to refine with common parameters. Weighting scheme $w = 1/[\sigma^2(F) + 0{\cdot}003F^2]$, $\sigma(F)$ taken from counting statistics. Final *R*(*F*) 0·069 (*wR* 0·075, *S* 1·69) and final shift/e.s.d. ratio <0·05. In final difference map, max. and min. peaks 0·16, −0·17 e Å^{-3}. Scattering factors from *International Tables for X-ray Crystallography* (1974). Atomic coordinates are given in Table 1, interatomic distances and angles in Table 2.*

Discussion. It has been noted that the imidazolidine ring is planar in most hydantoins which are mono- or disubstituted at the 5-position, with the N(1)—C(5) and C(4)—C(5) distances and N(1)—C(5)—C(4) bond angle in the ranges 1·45 to 1·48, 1·51 to 1·55 Å and 99 to 101° respectively (Camerman & Camerman, 1971; Florencio, Smith-Verdier & García-Blanco, 1978*a*, *b*; Fujiwara & Van der Veen, 1979; Koch, Germain, Declercq & Dusausoy, 1975; Smith-Verdier, Florencio & García-Blanco, 1977, 1979). However, these corresponding values shift to about 1·38, 1·47 Å and 105° respectively when there is a methylene linkage at the 5-position (Drew *et al.*, 1987*a*,*b*; Thieme & Haedicke, 1978). In (1), which is an *E* isomer, the five-membered ring is also planar and the corresponding values are 1·399(6), 1·471(6) Å and 105·0(4)° respectively. Otherwise the bond lengths and angles in both the imidazolidine and the phenyl rings are normal. The C(6)—C(7) bond length at 1·449 (7) Å is shorter than expected for a single bond but much longer than that for a double bond, which indicates partial double-bond character.

In the *Z* isomer of a 5-benzylidene-1-methylhydantoin molecule the large methyl group at the N(1) position makes it impossible to have a planar structure and as a result the benzene ring is rotated 45·9 (3)° with respect to the imidazolidine ring (Drew *et al.*, 1987*b*). However, in (1), which is the *E* isomer, the *N*-methyl group is no longer in direct contact with an H atom at an *ortho* position of the phenyl ring and this angle is considerably reduced to 5·7 (3)° (see Table 3). The molecule is shown in Fig. 1. Such a relatively flat conformation was also observed in the *Z* isomer of 5-[(4-methoxyphenyl)methylene]hydantoin (Drew *et al.*, 1987*a*) in which there was little steric interaction between the N(1) imine H and the phenyl ring H atoms.

Another interesting feature in the structure of (1) is the values of the angles around the C(5) atom. The N(1)—C(5)—C(4) angle in the five-membered ring is around 105° in the 5-arylmethylenehydantoins as mentioned earlier and is 105·0 (4)° in this molecule.

Table 1. *Atomic coordinates* (×10^4) *and isotropic thermal parameters* (×10^3) *with e.s.d.'s in parentheses*

	x	*y*	*z*	$\bar{U}$*(Å^2)
N(1)	1411 (6)	1154 (2)	4232 (3)	60 (5)
C(2)	1348 (9)	656 (2)	3590 (4)	62 (6)
N(3)	1204 (7)	913 (2)	2661 (3)	62 (5)
C(4)	1218 (7)	1587 (2)	2683 (3)	54 (6)
C(5)	1351 (7)	1758 (2)	3736 (3)	50 (5)
C(6)	1422 (8)	2327 (2)	4223 (4)	55 (6)
C(7)	1334 (7)	3010 (2)	3921 (3)	50 (5)
C(8)	1604 (8)	3465 (2)	4666 (4)	65 (7)
C(9)	1544 (9)	4125 (3)	4463 (5)	78 (9)
C(10)	1234 (10)	4335 (3)	3530 (5)	88 (9)
C(11)	987 (8)	3897 (3)	2784 (5)	87 (7)
C(12)	1075 (8)	3234 (2)	2975 (4)	71 (6)
C(13)	1586 (10)	1068 (3)	5285 (4)	86 (7)
O(14)	1389 (6)	74 (2)	3789 (3)	82 (5)
O(15)	1190 (5)	1927 (2)	1956 (2)	69 (4)

* $\bar{U} = \frac{1}{3}\sum_i\sum_j U_{ij}\mathbf{a}_i.\mathbf{a}_j$.

Table 2. *Bond distances* (Å) *and angles* (°)

N(1)—C(2)	1·335 (6)	C(5)—C(6)	1·333 (7)
N(1)—C(5)	1·399 (6)	C(6)—C(7)	1·449 (7)
N(1)—C(13)	1·441 (6)	C(7)—C(8)	1·382 (7)
C(2)—N(3)	1·365 (7)	C(7)—C(12)	1·371 (7)
C(2)—O(14)	1·214 (6)	C(8)—C(9)	1·366 (7)
N(3)—C(4)	1·371 (6)	C(9)—C(10)	1·350 (10)
C(4)—C(5)	1·471 (6)	C(10)—C(11)	1·358 (9)
C(4)—O(15)	1·203 (6)	C(11)—C(12)	1·374 (7)
C(2)—N(1)—C(5)	110·6 (4)	N(1)—C(5)—C(6)	121·6 (4)
C(2)—N(1)—C(13)	123·7 (4)	C(4)—C(5)—C(6)	133·4 (4)
C(5)—N(1)—C(13)	125·6 (4)	C(5)—C(6)—C(7)	133·7 (5)
N(1)—C(2)—N(3)	108·2 (4)	C(6)—C(7)—C(8)	115·4 (4)
N(1)—C(2)—O(14)	126·5 (5)	C(6)—C(7)—C(12)	126·0 (4)
N(3)—C(2)—O(14)	125·3 (5)	C(8)—C(7)—C(12)	118·5 (4)
C(2)—N(3)—C(4)	111·2 (4)	C(7)—C(8)—C(9)	120·3 (5)
N(3)—C(4)—C(5)	104·9 (4)	C(8)—C(9)—C(10)	120·1 (6)
N(3)—C(4)—O(15)	123·9 (4)	C(9)—C(10)—C(11)	120·7 (5)
C(5)—C(4)—O(15)	131·2 (4)	C(10)—C(11)—C(12)	119·8 (6)
N(1)—C(5)—C(4)	105·0 (4)	C(7)—C(12)—C(11)	120·5 (5)

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* Lists of structure factors, anisotropic thermal parameters and H-atom parameters have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 43786 (7 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Table 3. *Least-squares planes for the molecule*

Distances of atoms from the two planes are given in Å. Atoms not contributing to the plane are marked with an asterisk.

Plane (1): N(1)–0·01 (1), C(2) 0·01 (1), N(3) –0·01 (1), C(4) 0·00 (1), C(5) 0·00 (1), C(6)* 0·01 (1), C(7)* –0·02 (1), C(13)* 0·02 (1), O(14)* –0·02 (1), O(15)* 0·05 (1)

Plane (2): N(1)* –0·02 (1), C(2)* 0·08 (1), C(5)* 0·05 (1), C(6)* –0·02 (1), C(7) –0·01 (1), C(8) 0·00 (1), C(9) 0·00 (1), C(10) 0·00 (1), C(11) –0·01 (1), C(12) 0·02 (1), O(14)* 0·07 (1)

Angle between plane (1) and plane (2): 5·7 (3)°

Plane (1) is that of the imidazolidine ring and (2) that of the benzene ring.

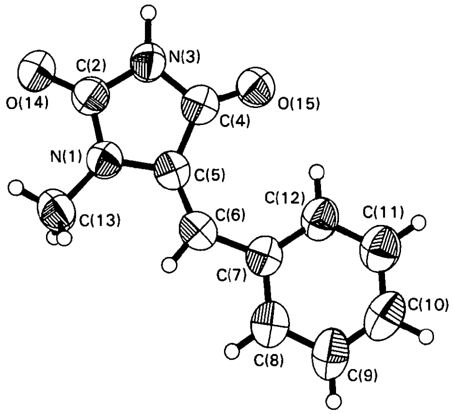

Fig. 1. The structure of (1). Thermal ellipsoids shown as 50% probabilities.

Values of the N(1)—C(5)—C(6) and C(4)—C(5)—C(6) angles in the present *E* isomer are 121·6 (4) and 133·4 (4)° compared to 132·6 (2) and 121·9 (2)° respectively in the *Z* isomer. The sum of these three angles around the C(5) atom is 360·0° to within experimental error in each case.

As is usual with this series of hydantoins, intermolecular hydrogen bonds are observed in the molecule. The N(3)—H(31)···O(14^{i}) and H(31)···O(14^{i}) distances are 2·81 (1) and 1·82 (6) Å respectively while the N(3)—H(31)···O(14^{i}) angle is 165 (5)° [symmetry element: (i) $x, -y, \frac{1}{2}-z$]. Molecules are thus paired by two hydrogen bonds around the twofold axis. It is interesting to note that in the *Z* isomer the hydrogen bond connects N(3) with the O(15) atom of another molecule.

References

Camerman, A. & Camerman, N. (1971). *Acta Cryst.* **B27**, 2205–2211.

Drew, M. G. B., Mok, K. F., Ang, K. P. & Tan, S. F. (1987*a*). *Acta Cryst.* **C43**, 743–745.

Drew, M. G. B., Mok, K. F., Ang, K. P. & Tan, S. F. (1987*b*). *Acta Cryst.* **C43**, 745–748.

Florencio, F., Smith-Verdier, P. & García-Blanco, S. (1978*a*). *Acta Cryst.* **B34**, 1317–1321.

Florencio, F., Smith-Verdier, P. & García-Blanco, S. (1978*b*). *Acta Cryst.* **B34**, 2220–2223.

Fujiwara, H. & Van der Veen, J. M. (1979). *J. Chem. Soc. Perkin Trans.* 2, pp. 659–663.

International Tables for X-ray Crystallography (1974). Vol. IV. Birmingham: Kynoch Press. (Present distributor D. Reidel, Dordrecht.)

Kirk-Othmer Encyclopedia of Chemical Technology (1980). 3rd ed., Vol. 12, p. 693. New York: John Wiley.

Koch, M. H., Germain, G., Declercq, J.-P. & Dusausoy, Y. (1975). *Acta Cryst.* **B31**, 2547–2549.

Sheldrick, G. M. (1976). *SHELX76*. Program for crystal structure determination. Univ. of Cambridge, England.

Smith-Verdier, P., Florencio, F. & García-Blanco, S. (1977). *Acta Cryst.* **B33**, 3381–3385.

Smith-Verdier, P., Florencio, F. & García-Blanco, S. (1979). *Acta Cryst.* **B35**, 216–217.

Tan, S. F., Ang, K. P. & Fong, Y. F. (1986). *J. Chem. Soc. Perkin Trans.* 2, pp. 1941–1944.

Thieme, P. C. & Haedicke, E. (1978). *Justus Liebigs Ann. Chem.* **2**, 227–237.

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Substituted Cyclopropanes. 1. *trans*-1,2,3-Tricyanocyclopropane

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Abstract. *trans*-Cyclopropane-1,2,3-tricarbonitrile, $C_6H_3N_3$, $M_r = 117·11$, orthorhombic, $Pnma$, $a = 11·917\ (3)$, $b = 8·934\ (2)$, $c = 5·456\ (2)$ Å, $V = 580·9$ Å^3, $Z = 4$, $D_x = 1·34$ Mg m^{-3}, $\lambda(\text{Mo } K\alpha) = 0·71069$ Å, $\mu = 0·08$ mm^{-1}, $F(000) = 240$, $T = 293$ K, $R = 0·043$ for 403 unique observed reflections. The

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