

khellactone [Bernotat-Wulf, Niggli, Ulrich & Schmid (1969)]; *trans*-4'-methylkhellactone [Macias, Masanet, Rodriguez-Luis, Salva & Fronczek (1989)].

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Structure of a Glycouril Derivative: Tetrahydro-1,6:3,4-di(methanoxy-methano)-3a,6a-dimethylimidazo[4,5-d]imidazole-2,5(1H,3H)-dione

BY A. SCHOUTEN* AND J. A. KANTERS

Laboratorium voor Kristal- en Structuurchemie, Rijksuniversiteit Utrecht, Transitorium 3, Padualaan 8, 3584 CH Utrecht, The Netherlands

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Abstract. $C_{10}H_{14}N_4O_4$, $M_r = 254.25$, monoclinic, $C2/c$, $a = 13.569$ (3), $b = 6.462$ (1), $c = 12.820$ (2) Å, $\beta = 103.61$ (2)°, $V = 1092.5$ (4) Å³, $Z = 4$, $D_x = 1.546$ g cm⁻³, m.p. 573 K (decomposition), $\lambda(Mo K\alpha) = 0.71073$ Å, $\mu = 1.1$ cm⁻¹, $F(000) = 536$, $T = 295$ K, $R = 0.034$ for 759 observed reflections with $I > 2.5\sigma(I)$. The molecule lies on a twofold rotational axis and contains four fused heterocyclic ring systems. The five-membered imidazolidone ring has a distorted envelope conformation and the

heteroatomic six-membered ring adopts a chair conformation. The least-squares planes of the five-membered rings are at an angle of 76.1 (1)°.

Experimental. Crystals were obtained by heating an aqueous solution of 3a,6a-dimethylglycouril (Himes, Hubbard, Mighell & Fatiadi, 1978) and formaldehyde, adjusting to pH 1 with concentrated hydrochloric acid and recrystallizing from dimethyl sulfoxide (Niele & Nolte, 1988). Colorless, transparent, plate-shaped crystal of dimensions 0.8 × 0.3 × 0.01 mm was used for data collection on an Enraf-

* Author to whom correspondence should be addressed.

Nonius CAD-4 diffractometer with Zr-filtered Mo $K\alpha$ radiation. Lattice parameters were determined from the setting angles of 21 reflections in the range $5.53 < \theta < 15.47^\circ$. The diffracted intensities of 884 unique reflections were collected using the ω - 2θ scan mode, $\Delta\omega = (0.55 + 0.35\tan\theta)^\circ$, $2\theta_{\max} = 52^\circ$ and $0 \leq h \leq 16$, $-7 \leq k \leq 0$, $-15 \leq l \leq 15$, with 759 reflections above the $2.5\sigma(I)$ level. Three standard reflections (020, 004 and 400), measured every hour, showed an average deviation less than 2% during the duration of the data collection. Intensities were corrected for Lp effects, but not for absorption. The structure was solved by the direct methods of *SHELXS86* (Sheldrick, 1986) and Fourier techniques. The H atoms were located on subsequent difference maps. The H atoms of the methyl group were placed at idealized positions (C—H 1.00 Å, H—C—H 109.5°) and refined riding on their bonded atom with an overall isotropic thermal parameter which refined to $0.065(4) \text{ \AA}^2$, whereas the non-methyl H atoms were refined with individual iso-

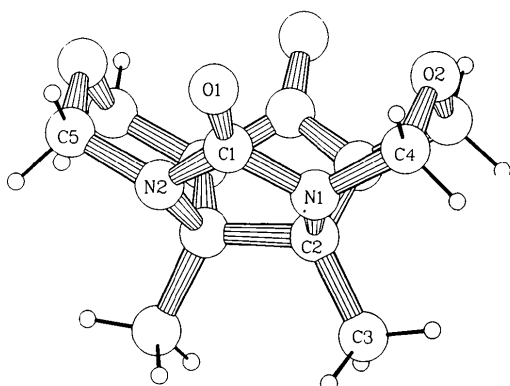


Fig. 1. Perspective view and atomic numbering of the title compound.

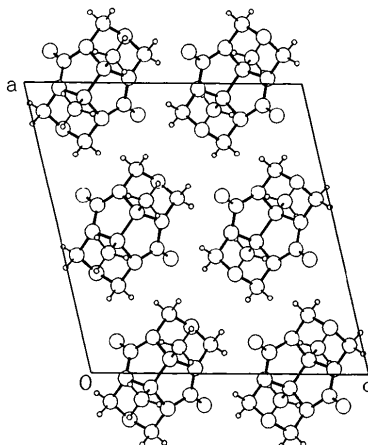


Fig. 2. Projection of the structure along *b*.

Table 1. Fractional coordinates and equivalent isotropic thermal parameters (\AA^2) and their e.s.d.'s in parentheses

$$U_{eq} = \frac{1}{3} \sum_i \sum_j U_{ij} a_i^* a_j^* a_i a_j$$

	<i>x</i>	<i>y</i>	<i>z</i>	U_{eq}
O(1)	0.1085 (1)	0.1647 (2)	0.1181 (1)	0.0479 (5)
O(2)	0.1597 (1)	0.1787 (2)	0.3993 (1)	0.0389 (5)
N(1)	0.1224 (1)	0.4121 (2)	0.2521 (1)	0.0310 (5)
N(2)	-0.0184 (1)	0.4025 (2)	0.1208 (1)	0.0320 (5)
C(1)	0.0735 (1)	0.3072 (3)	0.1599 (2)	0.0323 (6)
C(2)	0.0489 (1)	0.5285 (3)	0.2969 (1)	0.0294 (6)
C(3)	0.0902 (2)	0.7377 (3)	0.3395 (2)	0.0461 (8)
C(4)	0.2015 (2)	0.3082 (3)	0.3304 (2)	0.0382 (7)
C(5)	-0.0993 (2)	0.2978 (4)	0.0455 (2)	0.0399 (7)

Table 2. Bond distances (\AA) and bond angles ($^\circ$) with e.s.d.'s in parentheses

O(1)—C(1)	1.217 (2)	N(2)—C(1)	1.376 (2)
O(2)—C(4)	1.427 (2)	N(2)—C(2)	1.467 (2)
O(2)—C(5)	1.428 (3)	N(2)—C(5)	1.447 (3)
N(1)—C(1)	1.388 (2)	C(2)—C(2')	1.567 (3)
N(1)—C(2)	1.470 (2)	C(2)—C(3)	1.515 (3)
N(1)—C(4)	1.451 (3)		
C(4)—O(2)—C(5)	110.4 (2)	N(1)—C(1)—N(2)	107.6 (2)
C(1)—N(1)—C(2)	110.5 (2)	N(1)—C(2)—N(2)	109.2 (2)
C(1)—N(1)—C(4)	119.4 (2)	N(1)—C(2)—C(2')	103.3 (1)
C(2)—N(1)—C(4)	115.2 (2)	N(1)—C(2)—C(3)	111.7 (2)
C(1)—N(2)—C(2')	111.8 (2)	N(2)—C(2)—C(3)	112.2 (2)
C(1)—N(2)—C(5)	120.9 (2)	C(2)—C(2)—C(3)	116.8 (2)
C(2)—N(2)—C(5)	116.2 (2)	N(2)—C(2)—C(2)	102.8 (1)
O(1)—C(1)—N(1)	125.9 (2)	O(2)—C(4)—N(1)	111.2 (2)
O(1)—C(1)—N(2)	126.3 (2)	O(2)—C(5)—N(2)	110.8 (2)

Symmetry code: (i) $-x, y, \frac{1}{2} - z$.

tropic thermal parameters. Anisotropic full-matrix least-squares refinement on F of 99 parameters converged at $R = 0.034$ and $wR = 0.032$ with $w = 1.9088/\sigma^2(F_o)$, $S = 0.70$, $(\Delta/\sigma)_{av} = 0.040$, $(\Delta/\sigma)_{\max} = 0.584$. Maximum and minimum residual densities in the final difference map were 0.17 and -0.18 e \AA^{-3} respectively. The scattering factors were those of Cromer & Mann (1968) and anomalous-dispersion terms were from Cromer & Liberman (1970). Calculations were performed with *SHELX76* (Sheldrick, 1976) (refinement) and the *EUCLID* package (Spek, 1982) (geometry and illustrations) on a Micro VAX-II computer. A perspective view of the molecule with adopted numbering and a projection of the structure along the *b* axis are shown in Figs. 1 and 2 respectively. Fractional atomic coordinates for non-H atoms are listed in Table 1, bond distances and angles in Table 2.*

* Lists of structure factors, anisotropic thermal parameters, H-atom coordinates and torsion angles have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 53183 (8 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Related literature. The title compound contains four fused heterocyclic ring systems and serves as starting material for new cavitands. The five-membered *cis*-fused imidazolidone rings are distorted envelopes, the puckering parameters (Cremer & Pople, 1975) are $Q = 0.178$ (2) Å and $\varphi = 206.3$ (6)° and the least-squares planes are at an angle of 76.1 (1)°. Each five-membered ring is fused to a six-membered heteroatomic ring that has a chair conformation with puckering parameters $Q = 0.508$ (2) Å, $\theta = 9.9$ (2)° and $\varphi = 3$ (1)°. The molecule has a polar character and the peripheral positions of the O atoms constitute a potential receptor site for e.g. transition metal ammine complexes (Colquhoun, Doughty, Stoddart & Williams, 1984). In the crystal structure there are no intermolecular contacts which are less than the sum of the van der Waals radii.

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Structure of Membranolide, a Diterpene from the Antarctic Sponge *Dendrilla membranosa*

BY V. MANRIQUEZ, A. SAN-MARTIN AND J. ROVIROSA

Departamento de Química, Facultad de Ciencias, Universidad de Chile, Casilla 653, Santiago, Chile

J. DARIAS

Institute of Organic Natural Products CSIC La Laguna, Tenerife, Spain

AND K. PETERS

Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-7000 Stuttgart 80, Federal Republic of Germany

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Abstract. Methyl 2-[1,3-dihydro-3-oxo-5-(2,3,3-trimethylcyclohexyl)isobenzofuran-4-yl]propionate, $C_{21}H_{28}O_4$, $M_r = 344.45$, orthorhombic, $P2_12_12_1$, $a = 7.040$ (2), $b = 14.238$ (4), $c = 18.810$ (5) Å, $V = 1885.4$ Å³, $Z = 4$, $D_x = 1.214$ Mg m⁻³, $\lambda(\text{Mo } K\alpha) = 0.71069$ Å, $\mu = 0.08$ mm⁻¹, $F(000) = 744$, $T = 293$ K, $R = 0.050$ for 1758 unique observed reflections. The structure consists of a cyclohexane ring in the chair conformation which possesses as substituents at C(4) two methyl groups, and at C(10) an equatorial methyl group, and in an axial position an isobenzofuranone ring with an α -(methoxycarbonyl)ethyl side chain. All bond lengths and angles are within the expected ranges.

Experimental. The secondary metabolite membranolide was isolated from an acetonic extract of the Antarctic Sponge *Dendrilla membranosa*. The diter-

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pene was crystallized from ether/hexane solution. A plate-shaped colourless crystal of dimensions $0.3 \times 0.9 \times 0.2$ mm was used on a Huber four-circle diffractometer, graphite-monochromated Mo $K\alpha$ radiation, unit-cell dimensions from 24 centred reflections ($11 < \theta < 16^\circ$). ω scan used for data collection of 1819 unique reflections of which 1758 were observed with $F > 3\sigma(F)$. Three standard reflections ($\bar{3}15$, $1\bar{7}1$, $04\bar{7}$) every 100 measurements show 0.85% variation in intensity. Diffraction intensities were measured up to $\sin \theta/\lambda = 0.70$ Å⁻¹ in the index range $h = 0 \rightarrow 9$, $k = 0 \rightarrow 20$, $l = 0 \rightarrow 26$. Solved by direct phase determination using *SHELXTL-Plus* (Sheldrick, 1987), full-matrix least squares minimized $\sum w(\Delta F)^2$. H-atom positions calculated geometrically and considered isotropically with $U = 1.2U$ of bonded C atom. Positions and thermal parameters of all non-H atoms refined anisotropically giving 227 variables. $R =$