be expected to occur around the diastereometric linkages between the two cages; however, the remainder of the molecules should be equivalent. Molecular mechanics calculations (*MM*2; Allinger & Yuh, 1980) indicate that (5*a*) is about 5·2 kJ mol^{-1} less strained than (5*b*), $\Delta H_f = 53{\cdot}0$ and 58·2 kJ mol^{-1} for (5*a*) and 5(*b*), respectively. While bond-length and bond-angle differences between (5*a*) and 5(*b*) in the calculated structures are not as large as those observed, the trends about the linking atoms are similar. The two elongated bonds in each structure, C(1′)—C(7′) = 1·569 (3) and 1·559 (2) Å and C(9′)—C(10′) = 1·580 (3) and 1·583 (2) Å in (5*a*) and 5(*b*), respectively, are consistent with values observed for the parent cage and its derivatives (Watson, Nagl, Marchand & Reddy, 1989; Marchand, Arney, Gilardi & Flippen-Anderson, 1987; Marchand, Annapurna, Reddy, Watson & Nagl, 1988). In the other half of the molecule the ketone moiety imposes some geometric constraints, and internal strain is relieved partially by the elongation of four bonds to distances longer than 1·565 Å.

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References

ALLINGER, N. L. & YUH, Y. H. (1980). *MM*2 and *MMPI* subroutines for IBM PC. Updated by L. C. ROHREN (1984); adapted by J. J. GAJEWSKI & K. E. GILBERT from copy supplied by O. NELSON & C. SCOTT OWEN. Obtained from Serena Software, 489 Serena Lane, Bloomington, IN 47401, USA.

FLIPPEN-ANDERSON, J. L., GILARDI, R., GEORGE, C., MARCHAND, A. P., JIN, P.-W. & DESHPANDE, M. N. (1988). *Acta Cryst.* C**44**, 1617–1619.

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

LENOIR, D. (1977). *Synthesis*, pp. 553–554.

LERMAN, B. M., GALIN, F. Z., UMANSKAYA, L. A. & TOLSTIKOV, G. A. (1978). *J. Org. Chem. USSR*, **14**, 2536–2541.

MARCHAND, A. P. (1989). *Advances in Theoretically Interesting Molecules;* Vol. 1, edited by R. P. THUMMEL, pp. 357–397. Greenwich, CT: JAI Press.

MARCHAND, A. P., ANNAPURNA, P., REDDY, S. P., WATSON, W. H. & NAGL, A. (1988). *J. Org. Chem.* **54**, 187–193.

MARCHAND, A. P., ARNEY, B. E. JR, GILARDI, R. & FLIPPEN-ANDERSON, J. L. (1987). *J. Org. Chem.* **52**, 3455–3457.

MCMURRY, J. E. (1983). *Acc. Chem. Res.* **16**, 405–411; and references cited therein.

Nicolet Instrument Corporation (1986). *SHELXTL* for Desktop 30 (Microeclipse). PN-269-1040340. Siemens Analytical X-ray Instruments, Inc., Madison, Wisconsin, USA.

PONS, J.-M. & SANTELLI, M. (1988). *Tetrahedron*, **44**, 4295–4312; and references cited therein.

WATSON, W. H., NAGL, A., MARCHAND, A. P. & REDDY, G. M. (1989). *Acta Cryst.* C. Accepted.

Acta Cryst. (1989). C**45**, 1342–1346

A Diels–Alder Cycloadduct of Methylcyclopentadiene with 2,6-Dimethyl-*p*-benzoquinone and the Intramolecular Photocyclization Product of this Cycloadduct

BY WILLIAM H. WATSON,* ANTE NAGL† AND RAM P. KASHYAP

Department of Chemistry, Texas Christian University, Fort Worth, Texas 76129, *USA*

AND ALAN P. MARCHAND* AND DALIAN ZHAO

Department of Chemistry, University of North Texas, PO Box 5068, *Denton, Texas* 76203, *USA*

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Abstract. 3,4a,6-Trimethyl-1,4,4a,8a-tetrahydro-1,4-methanonaphthalene-5,8-dione, (1*b*), $C_{14}H_{16}O_2$, $M_r = 216{\cdot}28$, monoclinic, $P2_1/a$, $a = 11{\cdot}291$ (1), $b = 8{\cdot}267$ (1), $c = 12{\cdot}860$ (1) Å, $\beta = 107{\cdot}20$ (1)°, $V = 1146{\cdot}6$ (2) Å^3, $Z = 4$, $D_x = 1{\cdot}253$ g cm^{-3}, λ(Mo $K\alpha$) = 0·71073 Å, $\mu = 0{\cdot}77$ cm^{-1}, $F(000) = 464$, $T = 295$ K, $R = 0{\cdot}0445$ for 1279 reflections. 1,2,10-Trimethylpentacyclo[5.4.0.0^{2,6}.0^{3,10}.0^{5,9}]undecane-8,11-dione, (2), $C_{14}H_{16}O_2$, $M_r = 216{\cdot}28$, orthorhombic, *Pcab*, $a = 10{\cdot}998$ (2), $b = 11{\cdot}828$ (2), $c = 17{\cdot}179$ (2) Å, $V = 2234{\cdot}5$ Å^3, $Z = 8$, $D_x = 1{\cdot}286$ g cm^{-3}, λ(Mo $K\alpha$) = 0·71073 Å, $\mu = 0{\cdot}79$ cm^{-1}, $F(000) = 928$, $T = 295$ K, $R = 0{\cdot}0378$ for 1186 observed reflections. Structure (1*b*) contains a norbornene moiety fused to a cyclohexenedione ring. The two sets of C atoms which are joined in the photochemical reaction to form the four-membered ring in (2) are separated by

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* Authors to whom correspondence should be addressed.

† On leave of absence from the Faculty of Science, University of Zagreb, Yugoslavia.

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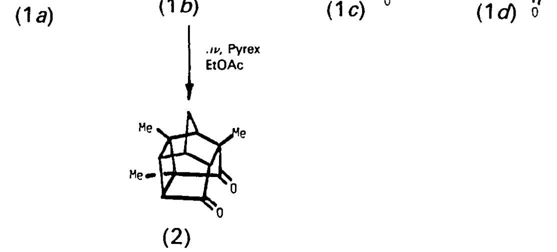
3·671 (3) and 3·685 (5) Å. Compound (2) contains a norbornane system fused to two five-membered rings and a four-membered ring. Two bonds in the structure are elongated to 1·585 (3) and 1·591 (3) Å. The two keto C atoms are separated by 2·542 (3) Å with the O atoms pointing away from each other. The heats of formation of (1*b*) and (2) are estimated by molecular mechanics calculations to be $-163·5$ and $-250·7$ kJ mol^{-1} with strain energies of 85·1 and 138·5 kJ mol^{-1}, respectively.

Introduction. As part of a continuing study of Diels–Alder reactions of substituted cyclopentadienes with substituted *p*-benzoquinones (Marchand, Suri, Earlywine, Powell & van der Helm, 1984; Flippen-Anderson, Gilardi, George, Marchand & Jin, 1987; Marchand, 1989; Watson & Nagl, 1988), the reaction of a mixture of 1- and 2-methylcyclopentadienes (Csicsery, 1960) with 2,6-dimethyl-*p*-benzoquinone has been studied. When freshly cracked methylcyclopentadiene dimer (20% molar excess) was heated with a benzene solution of the dienophile in a sealed tube at 363 K for 5 h, a mixture of four products {NMR consistent with all four possible *endo* [4 + 2] cycloadducts, (1*a*)–(1*d*)} was obtained in 77% yield. Partial separation of this mixture of isomeric products was performed by using flash column chromatography on silica gel. A gradient elution scheme that employed 1–2% ethyl acetate–hexane as eluent led to the isolation of a pure isomer from the mixture, m.p. 376–377 K. The fact that this cycloadduct isomer possesses the *endo* configuration was shown by its facile intramolecular photocyclization to afford the corresponding trimethylpentacyclo[5.4.0.0^{2,6}.0^{3,10}.0^{5,9}]undecane-8,11-dione, (2), m.p. 360–361 K. The proton NMR spectrum of the Diels–Alder cycloadduct indicated the presence of only two vinyl protons (Marchand & Zhao, 1989), and the compound may possess either structure (1*b*) or (1*c*) but not (1*a*) or (1*d*). In order to resolve ambiguities regarding (i) the structure of the Diels–Alder cycloadduct [*i.e.* (1*b*) or (1*c*)] and (ii) the structure of the cage diketone, (2), the structures of both compounds were elucidated by X-ray diffraction.



Scheme 1

Experimental. All data were collected on a Nicolet *R*3*m*/*μ* update of a $P2_1$ diffractometer using the *ω*-scan mode with a variable scan rate of 4 to 29·3° min^{-1} and graphite-monochromated Mo *Kα* radiation. All computer programs supplied by Nicolet for Desktop 30 Microeclipse and Nova 4/C configuration (Nicolet Instrument Corporation, 1986); atomic scattering factors and anomalous-dispersion corrections from *International Tables for X-ray Crystallography* (1974).

A colorless single crystal of (1*b*) with dimensions 0·50 × 0·50 × 0·50 mm was used to collect data in the range $3 \le 2\theta \le 45°$; lattice parameters from a least-squares refinement of 25 reflections ($25·08 \le 2\theta \le 33·62°$); systematic absences ($h0l$, $h = 2n + 1$; $0k0$, $k = 2n + 1$) and Laue symmetry $2/m$ consistent with space group $P2_1/a$; intensity data were corrected for a 5% decrease in the intensities of check reflections 608 and 434; 1496 unique reflections collected ($-11 \le h \le 11$, $0 \le k \le 8$, $0 \le l \le 13$), $1279 \ge 3\sigma(I)$, equivalent reflections averaged; Lorentz–polarization corrections applied, a *ψ*-scan-based empirical absorption correction applied (transmission factors 0·880 to 0·924); structure solved by direct methods, block-cascade least-squares refinement, H atoms found in difference map and refined with isotropic thermal parameters; final $R = 0·0445$, $wR = 0·0621$ for 209 parameters and 1279 reflections ($R = 0·0521$ and $wR = 0·0639$ for all 1496 reflections), $S = 1·465$, $(\Delta/\sigma)_{\max} = 0·013$, largest peaks in the final difference map of 0·13 and $-0·22$ e Å^{-3}; $\sum w(|F_o| - |F_c|)^2$ refined with $w = [\sigma^2(F_o) + 0·00121F_o^2]^{-1}$. Atomic positional parameters are listed in Table 1 while bond distances and valence angles are given in Table 2.

A colorless single crystal of (2) with dimensions 0·33 × 0·33 × 0·38 mm was used to collect all data, $0 \le 2\theta \le 45°$; lattice parameters from a least-squares refinement of 25 reflections ($25·03 \le 2\theta \le 29·73°$); systematic absences ($0kl$, $l = 2n + 1$; $h0l$, $h = 2n + 1$; $hk0$, $k = 2n + 1$) and Laue symmetry consistent with space group *Pcab*; monitored reflections 119 and 361 showed deviations of less than ±2%; 1464 independent reflections ($0 \le h \le 11$, $0 \le k \le 12$, $0 \le l \le 18$), $1186 \ge 3\sigma(I)$; Lorentz–polarization corrections applied, a *ψ*-scan-based empirical absorption correction applied (transmission factors 0·739 to 0·750); structure solved by direct methods, block-cascade least-squares refinement, H atoms found in a difference map and refined with isotropic thermal

Table 1. *Atomic coordinates* (×10⁴) *and isotropic thermal parameters* (Å² × 10³) *for compound* (1*b*)

Equivalent isotropic *U* defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x	*y*	*z*	*U*
C(1)	2746 (2)	9652 (3)	9189 (2)	53 (1)
C(2)	3295 (2)	8023 (3)	9071 (2)	52 (1)
C(3)	2427 (2)	7128 (3)	8423 (2)	47 (1)
C(4)	1263 (2)	8168 (3)	8070 (2)	46 (1)
C(4a)	1507 (2)	9486 (3)	7284 (2)	45 (1)
C(5)	1906 (2)	8701 (3)	6383 (2)	45 (1)
O(5)	1277 (2)	7627 (2)	5839 (1)	66 (1)
C(6)	3027 (2)	9259 (3)	6113 (2)	48 (1)
C(7)	3821 (2)	10253 (3)	6778 (2)	58 (1)
C(8)	3689 (2)	10865 (3)	7803 (2)	58 (1)
O(8)	4516 (2)	11658 (2)	8411 (2)	87 (1)
C(8a)	2518 (2)	10540 (3)	8088 (2)	52 (1)
C(9)	1427 (2)	9150 (3)	9112 (2)	57 (1)
C(10)	2507 (3)	5441 (3)	8031 (2)	67 (1)
C(11)	330 (2)	10447 (3)	6717 (2)	69 (1)
C(12)	3213 (2)	8615 (3)	5084 (2)	68 (1)

Table 2. *Bond distances* (Å) *and valence angles* (°) *for compound* (1*b*)

C(1)—C(2)	1·509 (3)	C(1)—C(8a)	1·547 (3)
C(1)—C(9)	1·521 (4)	C(2)—C(3)	1·311 (3)
C(3)—C(4)	1·523 (3)	C(3)—C(10)	1·495 (3)
C(4)—C(4a)	1·564 (3)	C(4)—C(9)	1·531 (3)
C(4a)—C(5)	1·508 (3)	C(4a)—C(8a)	1·560 (3)
C(4a)—C(11)	1·534 (3)	C(5)—O(5)	1·220 (3)
C(5)—C(6)	1·482 (3)	C(6)—C(7)	1·325 (3)
C(6)—C(12)	1·498 (4)	C(7)—C(8)	1·460 (4)
C(8)—O(8)	1·216 (3)	C(8)—C(8a)	1·496 (4)

C(2)—C(1)—C(8a)	107·0 (2)	C(2)—C(1)—C(9)	100·0 (2)
C(8a)—C(1)—C(9)	100·1 (2)	C(1)—C(2)—C(3)	108·4 (2)
C(2)—C(3)—C(4)	106·8 (2)	C(2)—C(3)—C(10)	129·2 (2)
C(4)—C(3)—C(10)	124·0 (2)	C(3)—C(4)—C(4a)	106·6 (2)
C(3)—C(4)—C(9)	99·6 (2)	C(4a)—C(4)—C(9)	101·4 (2)
C(4)—C(4a)—C(5)	110·2 (2)	C(4)—C(4a)—C(8a)	101·3 (2)
C(5)—C(4a)—C(8a)	115·0 (2)	C(4)—C(4a)—C(11)	112·0 (2)
C(5)—C(4a)—C(11)	105·8 (2)	C(8a)—C(4a)—C(11)	112·8 (2)
C(4a)—C(5)—O(5)	119·9 (2)	C(4a)—C(5)—C(6)	121·1 (2)
O(5)—C(5)—C(6)	118·9 (2)	C(5)—C(6)—C(7)	120·2 (2)
C(5)—C(6)—C(12)	116·7 (2)	C(7)—C(6)—C(12)	123·1 (2)
C(6)—C(7)—C(8)	124·6 (2)	C(7)—C(8)—O(8)	120·2 (2)
C(7)—C(8)—C(8a)	119·4 (2)	O(8)—C(8)—C(8a)	120·3 (2)
C(1)—C(8a)—C(4a)	103·4 (2)	C(1)—C(8a)—C(8)	112·9 (2)
C(4a)—C(8a)—C(8)	117·7 (2)	C(1)—C(9)—C(4)	93·7 (2)

Table 3. *Atomic coordinates* (×10⁴) *and isotropic thermal parameters* (Å² × 10³) *for compound* (2)

Equivalent isotropic *U* defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x	*y*	*z*	*U*
C(1)	−1677 (2)	8455 (2)	6189 (1)	38 (1)
C(2)	−2618 (2)	9317 (2)	5848 (1)	39 (1)
C(3)	−1966 (2)	10477 (2)	5790 (1)	43 (1)
C(4)	−2836 (2)	11280 (2)	6197 (1)	54 (1)
C(5)	−2893 (2)	10601 (2)	6944 (1)	47 (1)
C(6)	−3241 (2)	9408 (2)	6656 (1)	44 (1)
C(7)	−2331 (2)	8558 (2)	7009 (1)	45 (1)
C(8)	−1571 (2)	9261 (2)	7545 (1)	47 (1)
O(8)	−1082 (2)	8977 (1)	8143 (1)	72 (1)
C(9)	−1557 (2)	10414 (2)	7176 (1)	44 (1)
C(10)	−890 (2)	10353 (2)	6353 (1)	41 (1)
C(11)	−525 (2)	9134 (2)	6225 (1)	41 (1)
O(11)	498 (1)	8781 (1)	6143 (1)	65 (1)
C(12)	−1555 (3)	7275 (2)	5865 (2)	55 (1)
C(13)	−3357 (2)	8978 (2)	5152 (1)	61 (1)
C(14)	135 (2)	11197 (2)	6269 (1)	63 (1)

Table 4. *Bond distances* (Å) *and valence angles* (°) *for compound* (2)

C(1)—C(2)	1·564 (3)	C(1)—C(7)	1·585 (3)
C(1)—C(11)	1·499 (3)	C(1)—C(12)	1·506 (3)
C(2)—C(3)	1·549 (3)	C(2)—C(6)	1·548 (3)
C(2)—C(13)	1·498 (3)	C(3)—C(4)	1·516 (3)
C(3)—C(10)	1·533 (3)	C(4)—C(5)	1·513 (3)
C(5)—C(6)	1·540 (3)	C(5)—C(9)	1·536 (3)
C(6)—C(7)	1·541 (3)	C(7)—C(8)	1·492 (3)
C(8)—O(8)	1·205 (3)	C(8)—C(9)	1·502 (3)
C(9)—C(10)	1·591 (3)	C(10)—C(11)	1·510 (3)
C(10)—C(14)	1·511 (3)	C(11)—O(11)	1·207 (2)

C(2)—C(1)—C(7)	88·9 (1)	C(2)—C(1)—C(11)	103·0 (1)
C(7)—C(1)—C(11)	107·7 (1)	C(2)—C(1)—C(12)	121·6 (2)
C(7)—C(1)—C(12)	116·1 (2)	C(11)—C(1)—C(12)	115·8 (2)
C(1)—C(2)—C(3)	107·1 (1)	C(1)—C(2)—C(6)	90·2 (1)
C(3)—C(2)—C(6)	101·5 (1)	C(1)—C(2)—C(13)	118·8 (2)
C(3)—C(2)—C(13)	115·8 (2)	C(6)—C(2)—C(13)	119·5 (2)
C(2)—C(3)—C(4)	103·4 (1)	C(2)—C(3)—C(10)	103·4 (1)
C(4)—C(3)—C(10)	104·9 (2)	C(3)—C(4)—C(5)	94·8 (2)
C(4)—C(5)—C(6)	102·9 (2)	C(4)—C(5)—C(9)	104·8 (2)
C(6)—C(5)—C(9)	100·9 (2)	C(2)—C(6)—C(5)	104·0 (2)
C(2)—C(6)—C(7)	91·1 (1)	C(5)—C(6)—C(7)	107·9 (2)
C(1)—C(7)—C(6)	89·7 (1)	C(1)—C(7)—C(8)	109·6 (2)
C(6)—C(7)—C(8)	104·1 (2)	C(7)—C(8)—O(8)	128·2 (2)
C(7)—C(8)—C(9)	104·5 (2)	O(8)—C(8)—C(9)	127·3 (2)
C(5)—C(9)—C(8)	103·2 (2)	C(5)—C(9)—C(10)	102·6 (2)
C(8)—C(9)—C(10)	109·7 (2)	C(3)—C(10)—C(9)	101·5 (1)
C(3)—C(10)—C(11)	101·7 (2)	C(9)—C(10)—C(11)	107·1 (2)
C(3)—C(10)—C(14)	116·9 (2)	C(9)—C(10)—C(14)	113·4 (2)
C(11)—C(10)—C(14)	114·7 (2)	C(1)—C(11)—C(10)	107·0 (2)
C(1)—C(11)—O(11)	126·6 (2)	C(10)—C(11)—O(11)	126·4 (2)

parameters; final $R = 0{\cdot}0378$, $wR = 0{\cdot}0389$ for 210 parameters and 1186 reflections ($R = 0{\cdot}0505$ and $wR = 0{\cdot}0401$ for all 1464 reflections), $S = 1{\cdot}484$, $(\Delta/\sigma)_{\max} = 0{\cdot}014$, largest peaks in the final difference map of 0·13 and −0·13 e Å^{-3}; $w = [\sigma^2(F_o) + 0{\cdot}00020F_o^2]^{-1}$; isotropic extinction correction, $F = F_c/[1{\cdot}0 + 0{\cdot}002gF_c^2/\sin(2\theta)]^{0{\cdot}25}$ (Nicolet Instrument Corporation, 1986) with g refined to 0·00195 (5). Atomic positional parameters are listed in Table 3 while bond distances and valence angles are listed in Table 4.*

Discussion. Figs. 1 and 2 present drawings of compounds (1*b*) and (2). In compound (1*b*) a norbornene moiety is fused to a cyclohexenedione ring. The cyclohexenedione is approximately planar (0·06 Å r.m.s.d.) and makes an angle of 126·4 (6)° with C(1)—C(8a)—C(4a)—C(4) (0·012 Å r.m.s.d.) of the norbornene group. The two keto O atoms are out of the plane by 0·287 (5) and 0·145 (5) Å. The C(1)—C(8a)—C(4a)—C(4) and C(1)—C(2)—C(3)—C(4) (0·005 Å r.m.s.d.) planes make angles of 120·9 (5) and 127·6 (3)° with C(1)—C(9)—C(4). The distances between atoms C(2) to C(7) and C(3) to C(6) which are joined during the photochemical

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* Lists of H-atom coordinates, anisotropic thermal parameters and structure factors have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 51906 (23 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

reaction to produce (2) are 3·671 (3) and 3·685 (3) Å, respectively. The three methyl groups on one side of the molecule lead to an elongation of bonds relative to those of the unsubstituted side. As with most norbornane and norbornene structures, the C(4a)—C(8a) ethane-type bond [1·560 (3) Å] is elongated (Dallinga & Toneman, 1968; Marchand, Chou, Ekstrand & van der Helm, 1976; Watson, Nagl, Marchand & Deshpande, 1989); however, in (*1b*) the C(4)—C(4a) distance of 1·564 (3) Å is statistically equivalent. This lengthening may be attributed to the methyl substituents at C(4a). Molecular mechanics calculations are consistent with this observation.

In compound (2) the norbornane moiety is fused to two five-membered rings and a four-membered ring. The C(1)—C(7) and C(9)—C(10) bonds of 1·585 (3) and 1·591 (3) Å are significantly longer than the others in the structure. This lengthening has been attributed to a through bond coupling involving the keto groups (Ōsawa & Kanematsu, 1986; Marchand, Huang, Kaya, Baker, Jemmis & Dixon, 1987; Watson, Nagl, Marchand, Reddy & Reddy, 1989). The normal molecular mechanics parameters (Allinger & Yu, 1980) cannot reproduce these distances. The two keto C atoms C(9) and C(11) are separated by 2·542 (3) Å while the two keto O atoms are separated by 3·849 (2) Å. The C(7)—C(8)—C(9)—O(8) and C(1)—C(11)—C(10)—O(11) groups are approximately planar with the central C atoms, C(8) and C(11), lying out of the planes of the other three atoms by only 0·003 (3) and 0·015 (3) Å, respectively. The two groups are not coplanar but make an angle of 64·8 (4)°. The two five-membered rings exhibit slightly distorted envelope conformation with C(9) and C(10) being the flaps.

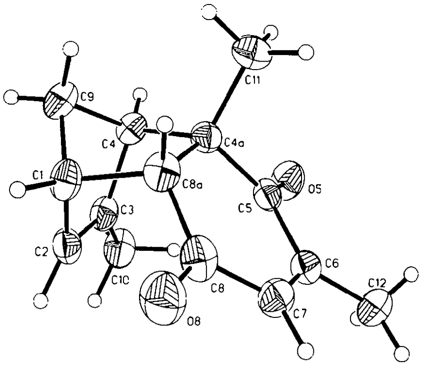

Fig. 1. Drawing of compound (*1b*) with thermal ellipsoids drawn at the 30% probability level. H atoms represented by spheres of arbitrary size.

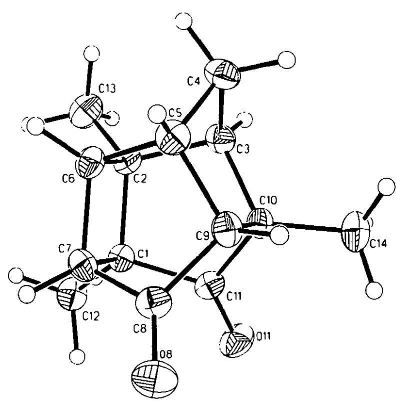

Fig. 2. Drawing of compound (2) with thermal ellipsoids drawn at the 30% probability level. H atoms represented by spheres of arbitrary size.

Molecular mechanics calculations using the program parameters (Allinger & Yu, 1980) estimate the heats of formation for (*1b*) and (2) to be − 163·5 and − 250·7 kJ mol^{-1}, respectively. The strain energies in the two molecules are 141·7 and 253·1 kJ mol^{-1} with 85·1 and 138·9 kJ attributed to angle strain and 42·4 and 128·2 kJ to torsional strain for (*1b*) and (2). The formation of the additional four-membered ring and two five-membered rings in (2) accounts for the angle-strain differences and the increased number of eclipsing interactions adds to the torsional strain. The cage structure (2) has a calculated crystal density of 1·286 g cm^{-3} which may be compared with the 1·253 g cm^{-3} value for (*1b*). The cage structure is a spacially more compact way of arranging the atoms which is of significance for the potential applications of high energy organic molecules.

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References

Allinger, N. L. & Yu, Y. H. (1980). *MM2* with *MMPI* subroutines for IBM PC. Updated by L. C. Rohren (1984). Adapted by J. J. Gagewski and K. E. Gilbert from copy supplied by O. Nelson and G. Scott Owen. Obtained from Serena Software, 489 Serena Lane, Bloomington, IN 47401, USA.

Csicsery, S. M. (1960). *J. Org. Chem.* **25**, 518–521.

Dallinga, G. & Toneman, L. H. (1968). *Recl Trav. Chim. Pays-Bas*, **87**, 795–804.

Flippen-Anderson, J. L., Gilardi, R. D., George, C. F., Marchand, A. P. & Jin, P. W. (1987). *Acta Cryst.* **C43**, 2151–2153.

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

Marchand, A. P. (1989). *Advances in Theoretically Interesting Molecules*, Vol. 1, edited by R. P. Thummel, pp. 357–397. Greenwich, CT: JAI Press.

Marchand, A. P., Chou, T.-C., Ekstrand, J. D. & van der Helm, D. (1976). *J. Org. Chem.* **41**, 1438–1444.

Marchand, A. P., Huang, C., Kaya, R., Baker, A. D., Jemmis, E. D. & Dixon, D. A. (1987). *J. Am. Chem. Soc.* **109**, 7095–7101.

Marchand, A. P., Suri, S. C., Earlywine, A. D., Powell, D. R. & van der Helm, D. (1984). *J. Org. Chem.* **49**, 670–675.

Marchand, A. P. & Zhao, D. (1989). In preparation.

Nicolet Instrument Corporation (1986). *SHELXTL* for Desktop 30 (Microeclipse). PN-269–1040340, April.

Ōsawa, E. & Kanematsu (1986). *Molecular Structure and Energetics*, Vol. 3, edited by J. F. Liebman & A. Greenberg, pp. 329–369. New York: VCH.

Watson, W. H. & Nagl, A. (1988). *Acta Cryst.* C**44**, 1852–1853.

Watson, W. H., Nagl, A., Marchand, A. P. & Deshpande, M. N. (1989). *Acta Cryst.* C**45**, 1339–1342.

Watson, W. H., Nagl, A., Marchand, A. P., Reddy, G. M. & Reddy, S. P. (1989). *Acta Cryst.* C**45**, 659–661.

Acta Cryst. (1989). C**45**, 1346–1348

Structure of 1-Amino-2-(9-methyl-1,8-dioxo-1,2,3,4,5,6,7,8,9,10-decahydro-10-acridinyl)propenedinitrile

By S. Selladurai, K. Subramanian* and S. Natarajan

Department of Physics, College of Engineering, Anna University, Madras-25, India

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Abstract. $C_{18}H_{18}N_4O_2$, $M_r = 322{\cdot}4$, triclinic, $P\bar{1}$, $a = 8{\cdot}5138\,(6)$, $b = 9{\cdot}407\,(3)$, $c = 11{\cdot}838\,(1)$ Å, $\alpha = 70{\cdot}76\,(2)$, $\beta = 79{\cdot}485\,(8)$, $\gamma = 79{\cdot}51\,(2)°$, $V = 872{\cdot}5$ Å^3, $Z = 2$, $D_x = 1{\cdot}23$ g cm^{-3}, $\lambda(\mathrm{Cu}\ K\alpha) = 1{\cdot}5418$ Å, $\mu = 6{\cdot}36$ cm^{-1}, $F(000) = 340$, $T = 296$ K, $R = 0{\cdot}049$ for 3302 observed reflections with $I > 3\sigma(I)$. The central acridinyl ring and one of the side rings adopt sofa conformations, the other side ring is midway between a half-chair and a sofa conformation. The 9-methyl group adopts an axial position on the acridinyl ring, with the dicyano group equatorial. The structure is stabilized by two N—H···O intermolecular hydrogen bonds N(18)···O(11) and N(18)···O(13) with N···O distances and N—H···O angles 2·84 (3), 2·81 (3) Å and 142 (2), 130 (2)° respectively.

Introduction. The title compound was supplied by Professor V. T. Ramakrishnan, Department of Chemistry, University of Madras, India. Aminoacridine derivatives have general uses in medicine especially as antibacterials for wound therapy (Acheson, 1956). For this reason the crystal structure of the title compound is of considerable interest to our continuing investigations.

Experimental. The crystals were obtained from acetone/ethanol as very small transparent yellow-coloured, rectangular blocks. A specimen with dimensions 0·25 × 0·35 × 0·65 mm was selected. The unit-cell parameters were obtained by measuring the 2θ values of 25 medium angle ($35 < 2\theta < 45°$) reflections (least-squares treatment). Intensity data were collected on an Enraf–Nonius CAD-4 diffractometer. Graphite-crystal-monochromated Cu $K\alpha$ radiation was used to measure 3594 independent reflections with $4 \leq 2\theta \leq 150°$ ($h\ 0 \rightarrow 10$, $k\ -11 \rightarrow 11$, $l\ -13 \rightarrow 14$), of which 3302 were considered observed [$I > 3\sigma(I)$, where $\sigma(I)$ was determined from counting statistics]. Lorentz, polarization and absorption (ω scans, $T_{\min} = 0{\cdot}903$, $T_{\max} = 0{\cdot}989$) corrections were applied.

The structure was determined by direct methods using *SHELX*86 (Sheldrick, 1986) and refinement was carried out by full-matrix least-squares method using *SHELX*76 (Sheldrick, 1976), with anisotropic temperature factors for non-H atoms. All 18 H atoms in the structure were located in a difference Fourier map and not refined. Subsequent refinement converged to $R = 0{\cdot}049$ and $wR = 0{\cdot}059$. Final difference density within $\pm 0{\cdot}17$ e Å^{-3}. $w = 8{\cdot}002/[\sigma^2 F_o + 0{\cdot}00008|F_o|^2]$. Ratio of final shift to e.s.d. $< 0{\cdot}1$. The atomic scattering factors used for all the atoms were as provided in the *SHELX*76 program. Computer programs: molecular illustrations drawn using *MOLDRAW* (Ugliengo, Borzani & Viterbo, 1988); *PARST* (Nardelli, 1983) for geometrical calculations.

Discussion. Table 1 contains the positional and equivalent isotropic thermal parameters of the non-H atoms.† Table 2 contains torsion angles and

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* To whom correspondence should be addressed.

† Lists of structure factors, anisotropic thermal parameters, H-atom coordinates, bond lengths and angles involving non-H atoms, bond lengths and angles involving H atoms, and least-squares-planes calculations have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 51884 (21 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

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