

with the assumed triple bond within the C₂ group, there is no reason to assume the presence of true thorium–thorium bonds.

The four N–Th bonds are directed toward the corners of a slightly distorted tetrahedron, as illustrated in Fig. 1. Within experimental error, the N–Th bonds are of equal length, 2.36 Å. A great many compounds have structures with a nitrogen atom tetrahedrally bonded to four thorium atoms, and averages of the reported values for the N–Th bond lengths in such compounds range from 2.34 to 2.41 Å (Benz & Zachariasen, 1966, 1969, 1970; Bowman & Arnold, 1971).

When a thorium atom is bonded to both carbon atoms of a C₂ pair, the bond length is considerably longer (3.06 Å) than the value of 2.82 Å, corresponding to binding to only one carbon of a pair. The mean Th–C bond length of 2.98 Å is large compared with the Th–N bond length of 2.36 Å. However, it should be remembered that the carbon atom is effectively univalent with respect to thorium in ThCN and that, hence, the strength of a Th–C bond is only 0.25. In the ThC structure, the bond strength is 0.67 and the bond length is 2.67 Å. The ThC₂ structure (Bowman, Krikorian, Arnold, Wallace & Nereson, 1968) also contains C₂ groups, but the carbon–carbon distance of 1.32 ± 0.04 Å corresponds to the formula ThC=C and, thus, the carbon is effectively divalent with respect to thorium. In ThC₂, the mean strength of a Th–C bond is 0.40 and the mean bond length is 2.74 Å.

The weakness of the Th–C bonds in the ThCN structure must cause excellent cleavage parallel to

(001), and must also be responsible for the low density of the compound. The volume per thorium atom in ThCN is 50.4 Å³, compared to the values of 46.4 Å³ for ThC₂ and 43.8 Å³ for ThO₂.

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The Crystal and Molecular Structure of 12-Methyl-11,13-dioxo-12-aza-pentacyclo[4.4.3.0.^{1,6}0^{2,10}.0^{5,7}]trideca-3,8-diene

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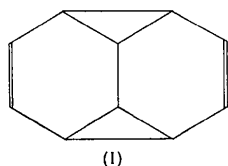
(Received 17 September 1971)

Crystals of the title compound, C₁₃H₁₁O₂N, are orthorhombic, space group *Fdd2*, *Z* = 8, *a* = 27.289 ± 0.004, *b* = 13.108 ± 0.005, *c* = 9.165 ± 0.002 Å, *D_x* = 1.362 g.cm⁻³. Data for 454 reflections were collected at room temperature on a Picker automated diffractometer. The structure was solved by direct methods, using a computerized multiple-solution procedure, and it was refined by full-matrix least-squares methods to a final *R* on *F* of 0.046, with 377 observed reflections. The average e.s.d.'s of the bond lengths and angles not involving hydrogen are about 0.008 Å and 0.5°, respectively. Bond lengths and angles were corrected for thermal motion. Some C–C single bonds are found to be significantly shorter than the normal C–C single bond length of 1.54 Å. The shortening of the C–C bonds is explained by the conjugation of the C=O bond and C=C bond with the three-membered ring.

Introduction

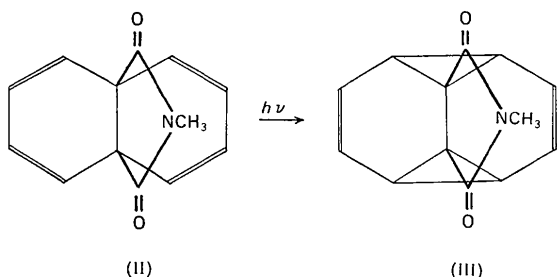
Crystals of what was presumed to be the title compound, C₁₃H₁₁O₂N (III), were obtained from Professor

David Ginsburg of the Technion, Haifa, Israel. This compound is the first stable derivative of the long sought, but never isolated, tetracyclo[4.4.0.0^{2,10}.0^{5,7}]deca-3,8-diene (I).



Compounds in the $(CH)_{10}$ series have been studied extensively because of the possibilities they offer for fluxional isomerism (see, e.g., Altman, Babad, Rubin & Ginsburg, 1969).

The compound we studied is the product of the photochemical rearrangement of 12-methyl-11,13-dioxo-12-aza[4.4.3]propella-2,4,7,9-tetraene (II).



Experimental

The crystals of the substance are white and tend to grow as parallelepipeds with an octagonal face per-

pendicular to an elongated prism axis. Weissenberg photographs showed them to be orthorhombic, with systematic extinctions of hkl for $h+k \neq 2n$, $k+l \neq 2n$; $0kl$ for $k+l \neq 4n$; $h0l$ for $h+l \neq 4n$, and $hk0$ for $h,k \neq 2n$, establishing the space group as $Fdd2$. The lattice constants, as determined from diffractometer data, are $a=27.289 \pm 0.004$, $b=13.108 \pm 0.005$, and $c=9.165 \pm 0.002$ Å. The only reasonable calculated density is for $Z=8$, viz., 1.362 g.cm $^{-3}$; the molecules thus lie on twofold axes.

Intensity data were collected on a Picker automatic four-circle diffractometer, using $Cu K\alpha$ radiation, with a scintillation counter and pulse-height analyzer. The moving-crystal moving-counter technique ($\theta-2\theta$ scan) was used with a 2θ scan speed of 1.0° min $^{-1}$. The scan range included both $K\alpha_1$ and $K\alpha_2$ peaks. For each reflection, the base width of the scan was 1.5° . Two sets of intensity equivalent data (hkl and $hk\bar{l}$) with $2\theta < 126^\circ$ were collected and their intensities averaged. Two reflections (0,12,0 and 551) were monitored during the two days required to collect the data, and they showed variations not greater than those expected from counting statistics. Corrections for absorption, negligible for a crystal of the size used, were not made. Of the 454 independent reflections collected, 377 were assigned as observed with intensity higher than $2.33\sigma_I$, where σ_I is defined by $\sigma_I = Lp(C+k^2B+0.0025C^2)^{1/2}$ in which C is the total count from the scan, k is the ratio of scanning time to the total background count B , and Lp is the Lorentz-polarization correction.

Structure determination and refinement

Preliminary scale and temperature factors were derived from Wilson's (1942) method. Initial attempts were made to solve the structure by rotating an idealized model of the molecule along the twofold axis to estimate the orientation of the molecule in the unit cell. This attempt was unsuccessful. Accordingly, statistical methods were employed to solve the crystal structure.

A three-dimensional E map, computed with the 84 phases obtained from a computerized multiple-solution procedure (Tsai, Donohue & Streib, 1969), revealed all nonhydrogen atoms in the asymmetric unit (Fig. 1).

The approximate coordinates of the nine nonhydrogen atoms obtained from the E map were refined by full-matrix least-squares methods, using the program of Sparks, Long & Trueblood (unpublished) and all 377 data. After five cycles with isotropic temperature factors, the value of R dropped to 11.8%, and the residual, $\sum w(|F_{obs}| - K|F_{calc}|)^2$, was reduced to 26.2% of its initial value. The refinement was carried further by refining anisotropic thermal parameters. After three cycles, R dropped to 6.8%, and the residual was 7.3% of its initial value. A Hughes (1941) weighting scheme, with $(w)^{1/2} = 1$ for reflections with $F_{obs} \leq 4F_{min}$ and $(w)^{1/2} = 4F_{min}/F_{obs}$ for $F_{obs} > 4F_{min}$ ($F_{min} = 18.2$), was used.

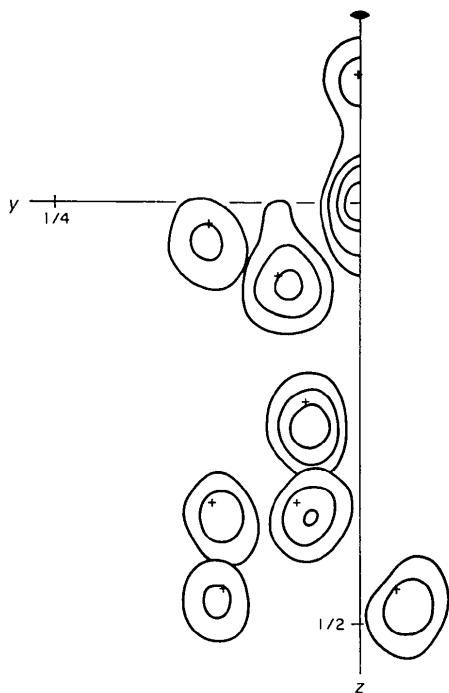


Fig. 1. A composite E map viewed along an axis perpendicular to the plane of the five-membered ring. 84 terms with $|E| > 1.2$ and the phases determined from a computerized multiple-solution procedure were used. Crosses mark the final atomic positions.

At this point, a difference Fourier synthesis was computed in order to find the hydrogen atoms. All the

Table 1. Observed and calculated structure factors

The asterisks indicate reflections designated as unobserved.

h	k	l	F _o	F _c	h	k	l	F _o	F _c	h	k	l	F _o	F _c	h	k	l	F _o	F _c
0	0	0	32.9	32.9	1	1	1	25.5	26.0	7	7	1	3.6	3.9	11	11	3	3.5	2.9
0	0	2	17.0	16.7	1	1	3	10.4	10.9	7	7	3	15.9	15.3	11	11	5	10.2	10.4
0	0	4	12.4	12.1	1	1	5	6.7	7.1	7	7	5	12.6	12.0	11	11	7	7.4	6.8
0	0	6	9.5	9.1	1	1	7	4.9	5.1	7	7	7	12.2	11.5	11	11	9	7.5	7.2
0	0	8	7.2	6.8	1	1	9	3.7	3.9	7	7	9	12.2	11.5	11	11	11	6.7	6.3
0	0	10	5.7	5.6	1	1	11	3.0	3.0	7	7	11	13.0	12.7	11	11	13	6.0	5.7
0	0	12	4.7	4.7	1	1	13	2.5	2.5	7	7	13	13.0	12.7	11	11	15	5.5	5.6
0	0	14	3.9	3.9	1	1	15	2.2	2.2	7	7	15	13.0	12.7	11	11	17	5.2	5.2
0	0	16	3.1	3.1	1	1	17	1.9	1.9	7	7	17	13.0	12.7	11	11	19	5.0	5.0
0	0	18	2.7	2.7	1	1	19	1.7	1.7	7	7	19	13.0	12.7	11	11	21	4.8	4.8
0	0	20	2.4	2.4	1	1	21	1.6	1.6	7	7	21	13.0	12.7	11	11	23	4.6	4.6
0	0	22	2.2	2.2	1	1	23	1.5	1.5	7	7	23	13.0	12.7	11	11	25	4.5	4.5
0	0	24	2.1	2.1	1	1	25	1.4	1.4	7	7	25	13.0	12.7	11	11	27	4.4	4.4
0	0	26	2.0	2.0	1	1	27	1.3	1.3	7	7	27	13.0	12.7	11	11	29	4.3	4.3
0	0	28	1.9	1.9	1	1	29	1.2	1.2	7	7	29	13.0	12.7	11	11	31	4.2	4.2
0	0	30	1.8	1.8	1	1	31	1.1	1.1	7	7	31	13.0	12.7	11	11	33	4.1	4.1
0	0	32	1.7	1.7	1	1	33	1.0	1.0	7	7	33	13.0	12.7	11	11	35	4.0	4.0
0	0	34	1.6	1.6	1	1	35	0.9	0.9	7	7	35	13.0	12.7	11	11	37	3.9	3.9
0	0	36	1.5	1.5	1	1	37	0.8	0.8	7	7	37	13.0	12.7	11	11	39	3.8	3.8
0	0	38	1.4	1.4	1	1	39	0.7	0.7	7	7	39	13.0	12.7	11	11	41	3.7	3.7
0	0	40	1.3	1.3	1	1	41	0.6	0.6	7	7	41	13.0	12.7	11	11	43	3.6	3.6
0	0	42	1.2	1.2	1	1	43	0.5	0.5	7	7	43	13.0	12.7	11	11	45	3.5	3.5
0	0	44	1.1	1.1	1	1	45	0.4	0.4	7	7	45	13.0	12.7	11	11	47	3.4	3.4
0	0	46	1.0	1.0	1	1	47	0.3	0.3	7	7	47	13.0	12.7	11	11	49	3.3	3.3
0	0	48	0.9	0.9	1	1	49	0.2	0.2	7	7	49	13.0	12.7	11	11	51	3.2	3.2
0	0	50	0.8	0.8	1	1	51	0.1	0.1	7	7	51	13.0	12.7	11	11	53	3.1	3.1
0	0	52	0.7	0.7	1	1	53	0.0	0.0	7	7	53	13.0	12.7	11	11	55	3.0	3.0
0	0	54	0.6	0.6	1	1	55	0.0	0.0	7	7	55	13.0	12.7	11	11	57	2.9	2.9
0	0	56	0.5	0.5	1	1	57	0.0	0.0	7	7	57	13.0	12.7	11	11	59	2.8	2.8
0	0	58	0.4	0.4	1	1	59	0.0	0.0	7	7	59	13.0	12.7	11	11	61	2.7	2.7
0	0	60	0.3	0.3	1	1	61	0.0	0.0	7	7	61	13.0	12.7	11	11	63	2.6	2.6
0	0	62	0.2	0.2	1	1	63	0.0	0.0	7	7	63	13.0	12.7	11	11	65	2.5	2.5
0	0	64	0.1	0.1	1	1	65	0.0	0.0	7	7	65	13.0	12.7	11	11	67	2.4	2.4
0	0	66	0.0	0.0	1	1	67	0.0	0.0	7	7	67	13.0	12.7	11	11	69	2.3	2.3
0	0	68	0.0	0.0	1	1	69	0.0	0.0	7	7	69	13.0	12.7	11	11	71	2.2	2.2
0	0	70	0.0	0.0	1	1	71	0.0	0.0	7	7	71	13.0	12.7	11	11	73	2.1	2.1
0	0	72	0.0	0.0	1	1	73	0.0	0.0	7	7	73	13.0	12.7	11	11	75	2.0	2.0
0	0	74	0.0	0.0	1	1	75	0.0	0.0	7	7	75	13.0	12.7	11	11	77	1.9	1.9
0	0	76	0.0	0.0	1	1	77	0.0	0.0	7	7	77	13.0	12.7	11	11	79	1.8	1.8
0	0	78	0.0	0.0	1	1	79	0.0	0.0	7	7	79	13.0	12.7	11	11	81	1.7	1.7
0	0	80	0.0	0.0	1	1	81	0.0	0.0	7	7	81	13.0	12.7	11	11	83	1.6	1.6
0	0	82	0.0	0.0	1	1	83	0.0	0.0	7	7	83	13.0	12.7	11	11	85	1.5	1.5
0	0	84	0.0	0.0	1	1	85	0.0	0.0	7	7	85	13.0	12.7	11	11	87	1.4	1.4
0	0	86	0.0	0.0	1	1	87	0.0	0.0	7	7	87	13.0	12.7	11	11	89	1.3	1.3
0	0	88	0.0	0.0	1	1	89	0.0	0.0	7	7	89	13.0	12.7	11	11	91	1.2	1.2
0	0	89	0.0	0.0	1	1	91	0.0	0.0	7	7	91	13.0	12.7	11	11	93	1.1	1.1
0	0	90	0.0	0.0	1	1	93	0.0	0.0	7	7	93	13.0	12.7	11	11	95	1.0	1.0
0	0	91	0.0	0.0	1	1	95	0.0	0.0	7	7	95	13.0	12.7	11	11	97	0.9	0.9
0	0	92	0.0	0.0	1	1	97	0.0	0.0	7	7	97	13.0	12.7	11	11	99	0.8	0.8
0	0	93	0.0	0.0	1	1	99	0.0	0.0	7	7	99	13.0	12.7	11	11	101	0.7	0.7
0	0	94	0.0	0.0	1	1	101	0.0	0.0	7	7	101	13.0	12.7	11	11	103	0.6	0.6
0	0	95	0.0	0.0	1	1	103	0.0	0.0	7	7	103	13.0	12.7	11	11	105	0.5	0.5
0	0	96	0.0	0.0	1	1	105	0.0	0.0	7	7	105	13.0	12.7	11	11	107	0.4	0.4
0	0	97	0.0	0.0	1	1	107	0.0	0.0	7	7	107	13.0	12.7	11	11	109	0.3	0.3
0	0	98	0.0	0.0	1	1	109	0.0	0.0	7	7	109	13.0	12.7	11	11	111	0.2	0.2
0	0	99	0.0	0.0	1	1	111	0.0	0.0	7	7	111	13.0	12.7	11	11	113	0.1	0.1
0	0	100	0.0	0.0	1	1	113	0.0	0.0	7	7	113	13.0	12.7	11	11	115	0.0	0.0

hydrogen atoms, except the three of the methyl group, were well resolved. By considering the known geometry of the methyl group, the most likely positions for these hydrogen atoms were obtained. In the least-squares refinement, the methyl hydrogen atoms were considered to be disordered. A model of six methyl hydrogen atoms with half weight was used. Several cycles of anisotropic least-squares refinement on the carbon, nitrogen and oxygen parameters, and simultaneous isotropic least-squares refinement on hydrogen parameters, keeping the methyl hydrogen contribution to the structure factors constant, dropped *R* to 4.6%. A weighting function based on counting statistics [$1/w=1/\sigma(F)$], was used. The results from both weighting schemes were approximately the same. The final electron density map computed with F_{obs} values is shown in Fig. 2. This composite drawing is a view along an axis perpendicular to the plane of the five-membered ring.

The final values of the observed and calculated

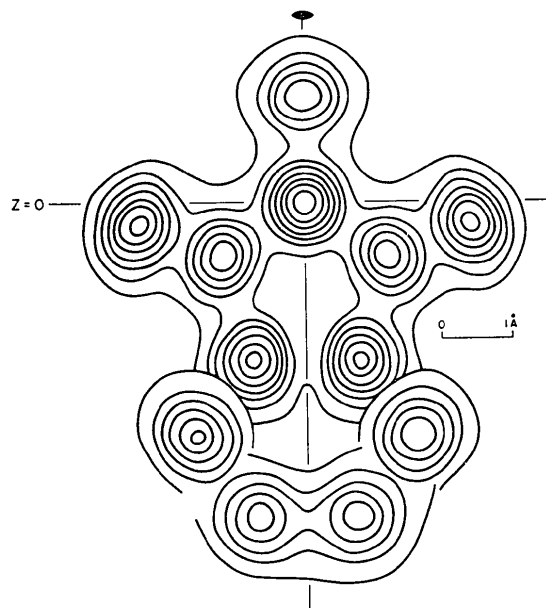


Fig. 2. Composite drawing of the final electron density map viewed along an axis perpendicular to the plane of the five-membered ring.

Table 2. Positional and thermal parameters, with their e.s.d.'s, of nonhydrogen atoms ($\times 10^4$)

The temperature factor is in the form: $\exp(-b_{11}h^2 - b_{22}k^2 - b_{33}l^2 - b_{12}hk - b_{13}hl - b_{23}kl)$

	x	y	z	b ₁₁	b ₂₂	b ₃₃	b ₁₂	b ₁₃	b ₂₃
C(1)	0	0	-1602 (10)	42 (3)	112 (6)	167 (13)	26 (6)	0	0
C(2)	412 (2)	675 (4)	842 (8)	28 (1)	67 (3)	197 (9)	-6 (3)	12 (6)	6 (8)
C(3)	267 (2)	434 (3)	2376 (8)	31 (1)	52 (2)	173 (8)	-8 (3)	0 (5)	3 (7)
C(4)	214 (3)	1233 (4)	3560 (9)	60 (2)	49 (3)	205 (8)	-5 (4)	-37 (7)	-17 (8)
C(5)	884 (3)	500 (4)	3547 (9)	44 (2)	83 (4)	208 (9)	-23 (4)	-45 (7)	-11 (11)
C(6)	-438 (3)	1103 (4)	4609 (9)	65 (2)	71 (3)	183 (9)	45 (5)	15 (8)	-22 (9)
C(7)	921 (3)	-345 (4)	4597 (8)	49 (2)	91 (4)	187 (9)	22 (5)	-47 (7)	-5 (9)
N	0	0	0	27 (2)	68 (3)	169 (10)	-6 (4)	0	0
O	824 (2)	1348 (3)	342 (7)	62 (1)	101 (3)	241 (7)	-73 (4)	35 (6)	58 (7)

structure factors are compared in Table 1. The final positional and thermal parameters and their standard deviations are given for nonhydrogen atoms in Table 2 and for nonmethyl hydrogen atoms in Table 3.

Table 3. *Positional ($\times 10^4$) and thermal parameters of hydrogen atoms*

	<i>x</i>	<i>y</i>	<i>z</i>	<i>B</i> (\AA^2)
H(4)	341 (28)	1881 (37)	3369 (60)	6.52 (1.16)
H(5)	1437 (32)	832 (41)	3252 (65)	8.22 (1.31)
H(6)	-576 (29)	1655 (41)	5345 (73)	7.68 (1.35)
H(7)	1335 (37)	-243 (49)	5664 (85)	10.90 (1.77)

The bond lengths and bond angles were corrected by fitting through the observed temperature-factor parameters to a rigid-body model (Schomaker & Trueblood, 1968). The quality of the fit of the rigid-body model is shown in Table 4, which lists the individual observed atomic mean-square displacement tensors and the cal-

culated atomic displacement tensors derived from the rigid-body parameters.

Discussion

The shape of the molecule, as it exists in the crystal, is given in Fig. 3. Bond lengths and angles corresponding to the positional parameters, both before and after librational corrections, are listed in Table 5.

Table 5. *Bond distances, angles, and their e.s.d.'s*

	Uncorrected	E.s.d.	Corrected
C(1)-N	1.468 \AA	0.009 \AA	1.469 \AA
C(2)-N	1.373	0.005	1.381
C(2)-C(3)	1.462	0.010	1.467
C(2)-O	1.223	0.006	1.224
C(3)-C(3')	1.465	0.005	1.478
C(3)-C(4)	1.511	0.009	1.516
C(3)-C(5)	1.515	0.009	1.521
C(4)-C(5)	1.505	0.007	1.512
C(4)-C(6)	1.491	0.009	1.496
C(5)-C(7)	1.469	0.009	1.474
C(6)-C(7')	1.298	0.007	1.304
C(1)-N-C(2)	124.2°	0.3°	124.0°
C(2)-N-C(2')	111.6	0.4	111.9
N-C(2)-O	123.8	0.6	124.0
N-C(2)-C(3)	108.2	0.3	108.0
O-C(2)-C(3)	128.0	0.6	128.0
C(2)-C(3)-C(3')	106.0	0.5	106.0
C(3)-C(4)-C(5)	60.3	0.4	60.3
C(3)-C(5)-C(4)	60.0	0.4	60.0
C(4)-C(3)-C(5)	59.7	0.4	59.7
C(4)-C(3)-C(3')	120.0	0.5	120.0
C(5)-C(3)-C(3')	119.2	0.5	119.2
C(3)-C(5)-C(7)	116.8	0.4	116.7
C(4)-C(5)-C(7)	120.6	0.6	120.6
C(5)-C(4)-C(6)	121.0	0.5	121.0
C(4)-C(6)-C(7')	124.7	0.6	124.8
C(5)-C(7)-C(6')	123.7	0.6	123.8
C(4)-H(4)	0.895 \AA		C(6)-C(4)-H(4) 114.8°
C(5)-H(5)	1.084		C(3)-C(5)-H(5) 117.8
C(6)-H(6)	1.018		C(4)-C(5)-H(5) 115.1
C(7)-H(7)	1.220		C(7)-C(5)-H(5) 115.3
			C(4)-C(6)-H(6) 121.6
			C(7)-C(6)-H(6) 113.5
C(3)-C(4)-H(4)	120.2°		C(5)-C(7)-H(7) 117.9
C(5)-C(4)-H(4)	114.6		C(6)-C(7)-H(7) 117.1

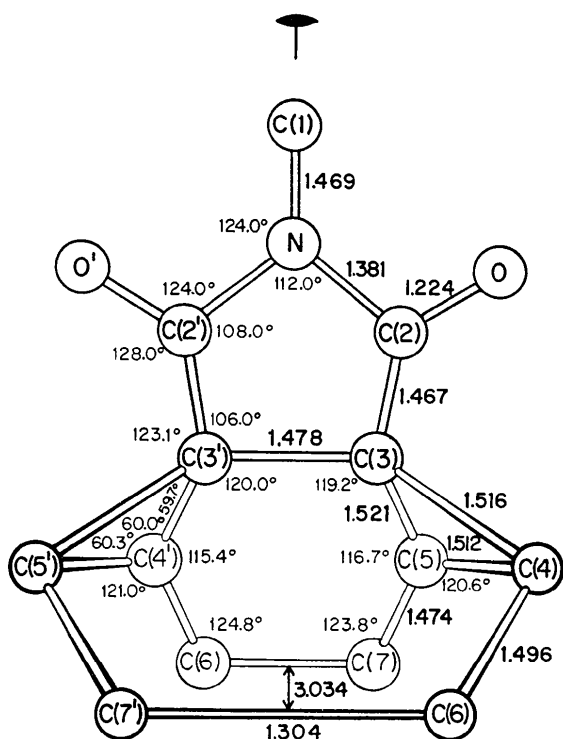


Fig. 3. Bond lengths (\AA) and bond angles ($^\circ$) in the molecule.

Table 4. *Mean-square atomic displacement tensors ($\text{\AA}^2 \times 10^4$)*

	U_{11}^{obs}	U_{11}^{cal}	U_{22}^{obs}	U_{22}^{cal}	U_{33}^{obs}	U_{33}^{cal}	U_{12}^{obs}	U_{12}^{cal}	U_{13}^{obs}	U_{13}^{cal}	U_{23}^{obs}	U_{23}^{cal}
C(1)	636	669	975	968	710	732	147	156	0	0	0	0
C(2)	425	492	587	578	839	801	-35	-95	46	26	18	70
C(3)	464	650	455	423	735	761	-46	-56	-1	-22	8	6
C(4)	913	919	429	422	871	849	-31	-71	-148	-73	-53	-47
C(5)	666	593	719	748	884	875	-131	-133	-179	-143	-33	-49
C(6)	981	1038	616	554	777	793	256	241	62	31	-66	-55
C(7)	749	756	792	854	794	819	128	184	-190	-174	-15	-34
N	403	430	587	623	717	731	33	34	0	0	0	0
O	944	865	875	859	1024	1008	-421	-355	140	94	176	180

The average estimated standard deviations in distances and angles involving heavy atoms are about 0.0076 \AA and 0.46° respectively. The C(1)-N distance is 1.469 (9) \AA , which is about the same as the corre-

sponding average value of 1.472 (5) Å for a trivalent nitrogen atom given by Sutton (1965). The C(2)–N distance of 1.381 (5) Å compares well with the average values (1.385 Å) for such bonds (uncorrected) in succinimide (Mason, 1961), and it indicates the combination of unsaturated effects on the five-membered ring and the resonance effect due to the presence of a carbon–oxygen double bond, shortening the bond length. The C(2)–N bond length of 1.381 (5) Å also agrees well with the two average C–N single-bond lengths of 1.385 and 1.374 Å in thymine–uracil (Donohue, 1968).

The C(2)–O double-bond distance is 1.224 (6) Å, comparable to those in ketones, in which the average distance is 1.215 (5) Å (Sutton, 1965). It also agrees well with the two average C=O double-bond distances of 1.224 and 1.233 Å in thymine–uracil.

The bond length of 1.467 (10) Å for C(2)–C(3) appears shorter than the average C–C single bond distance of 1.506 Å in succinimide (Mason, 1961) and the average C–C single bond distance of 1.49 Å in succinic anhydride (Ehrenberg, 1965); also it is even shorter than the average C–C bond lengths (1.515 Å) of the five-membered ring in deoxyadenosine monohydrate (Watson, Sutor & Tollin, 1965) and of 1.524 Å in adenosine 3'-phosphate dihydrate (Sundaralingam, 1966). The shortening of this bond length indicates that in addition to the conjugative effect in the five-membered ring and the resonance effect in the C=O double bond, the unsaturated character of the cyclopropane ring contributes an unusual effect in bond shortening. The shortening of the C–C bond between the cyclopropyl ring and the C–O double bond appeared also in cyclopropane-carbohydrazide (un-

corrected) as 1.478 Å (Chesnut & Marsh, 1958), and in cyclopropanecarboxamide (corrected) as 1.487 Å (Long, Maddox & Trueblood, 1969).

The central bond C(3)–C(3') has the bond length of 1.478 (5) Å. Statistically, it appears significantly different from the average C–C bond lengths of the five-membered ring mentioned above. This result also implies the conjugative effect of the two cyclopropyl rings.

The average C–C distance in the cyclopropyl ring in this structure is 1.516 Å, a value close to that in cyclopropyl cyanide (1.513 Å, Friend & Dailey, 1958) and in *sym*-tricyanocyclopropane (1.518 Å, Hartman & Hirshfeld, 1966), but somewhat greater than those in cyclopropanecarbohydrazide (1.497 Å, Chesnut & Marsh, 1958), in cyclopropanecarboxamide (1.498 Å, Long *et al.*, 1969), and in bicyclopropyl (1.507 Å, Eraker & Rømming, 1967).

The bond distances of C(5)–C(7), 1.474 (9) Å, and of C(4)–C(6), 1.496 (9) Å, appear shorter than the average C–C single bond lengths of 1.54 Å (Sutton, 1965). This indicates the resonance effect between the cyclopropyl ring and the C–C double bond. The similar effect of the shortening of the C–C single bond length between cyclopropyl ring and the C–C double bond can be seen in 2,5-dimethyl-7,7-dicyanonorcaradiene (Fritchie, 1966), which has the average C–C single bond length 1.480 Å.

The C(6)–C(7') double-bond length of 1.304 (7) Å is significantly shorter than the C–C double bond length of 1.332 Å in ethylene (Bartell & Bonham, 1959). However, the short C–C double-bond length of 1.300 Å was observed in the case like cyclopropene (Kasai, Meyers, Eggers & Wiberg, 1959). The intramolecular

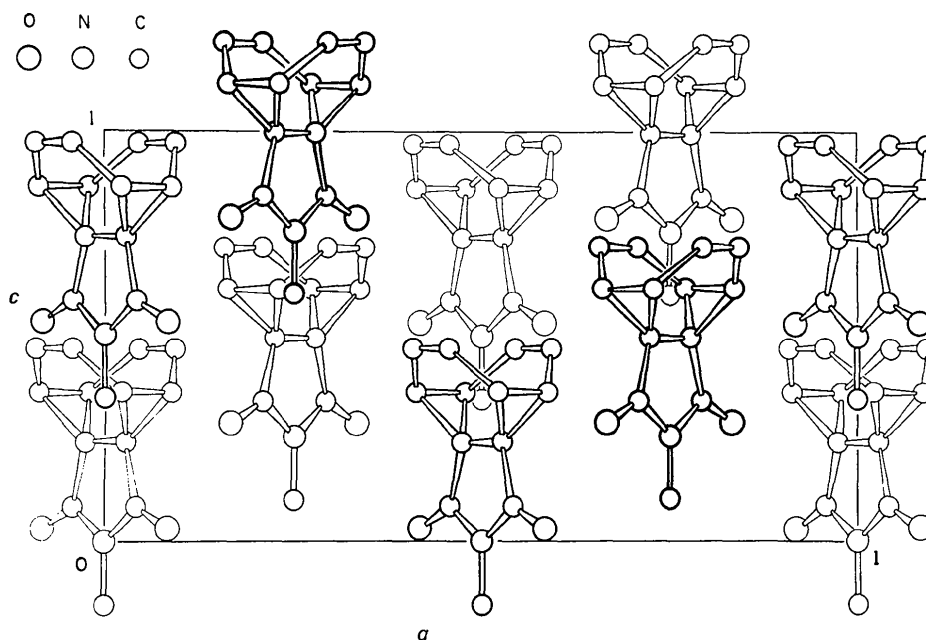


Fig. 4. Projection of the structure along the *b* axis. Hydrogen atoms are deleted for clarity.

nonbonded distance between the two double bonds C(6)–C(7') and C(6')–C(7) is 3.034 Å.

The inner angles of the five-membered ring agree well with those found in succinimide (Mason, 1961) and succinic anhydride (Ehrenberg, 1965). The inner angles of the cyclopropyl ring and other angles are normal in view of the known molecular geometry of other organic compounds.

The equation of the least-squares plane fitted through the five-membered ring is $0.77871x - 0.62738y - 0.00080z = 0.00062$. The atomic deviations from this plane and the weight of each atom are given in Table 6.

Table 6. Deviations from least-squares plane

Atoms with weight equal to one are included in the mean-plane calculations.

	w	Deviation		w	Deviation
N	1	-0.00062 Å	C(1)	1	0.00054 Å
C(2)	1	-0.00060	C(2')	1	0.00060
O	1	0.00012	O'	1	-0.00012
C(3)	1	0.00056	C(3')	1	-0.00056
C(4)	0	-0.73214	C(4')	0	0.73214
C(5)	0	0.78008	C(5')	0	-0.78008

Views of the structure along the *b* and *c* axes are shown in Figs. 4 and 5. As can be seen in Fig. 3, the

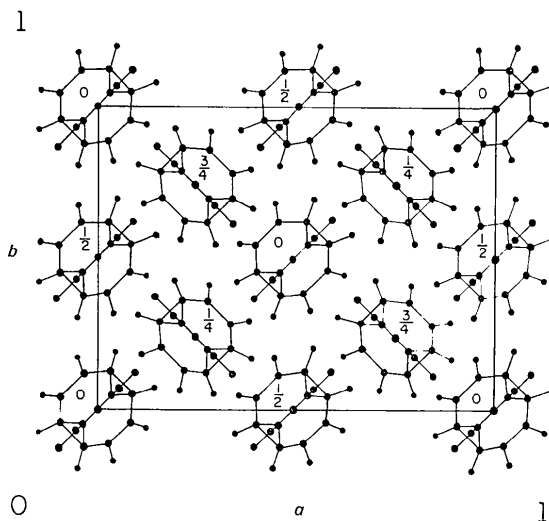


Fig. 5. Projection of the structure along the *c* axis.

molecule contains a twofold symmetry along the long axis of the molecule, the long axis being coincident with the twofold axis of the unit cell. None of the intermolecular distances is shorter than the sum of the van der Waals radii for the atoms involved. Since no close contacts are involved in the molecular packing, there might be some freedom of molecular motion in the crystal. That this is indeed true is shown by the large amplitudes (7, 4, 3°) of libration along the three axes of libration.

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