

measured with graphite-monochromated Mo K α radiation ($\lambda = 0.7017 \text{ \AA}$), and the ω - 2θ scan mode [scan width 1.2° (θ), scan speed 0.04° (θ) s^{-1}]. 2162 reflections were collected in the 2θ range 6° to 44° and, using the criterion $I_{rel} > 2\sigma(I_{rel})$, 1724 reflections were considered 'observed'. A Lorentz-polarization correction was applied; no correction was made for absorption.

The structure was solved by the automatic centrosymmetric routine of the *SHELX* program system (Sheldrick, 1976), in which an *E* map yielded 22 of the 25 heavy atoms, and subsequent least-squares refinement, followed by a difference map, showed the remaining heavy atoms. The final refinement was carried out with the heavy atoms treated anisotropically, the methyl H atoms refined as rigid groups and the remaining H atoms constrained at 1.08 \AA from their respective C atoms, their positions being dictated by the geometry of the molecule. The isotropic temperature factors of the H atoms, treated as three single parameters, refined to $U = 0.072 \text{ \AA}^2$ (aromatic H), $U = 0.053 \text{ \AA}^2$ (methine, methylene H) and $U = 0.137 \text{ \AA}^2$ (methyl H). The refinement converged to $R = 0.073$ and $R_w = \sum w^{1/2}|F_o - F_c| / \sum w^{1/2}|F_o| = 0.062$ with $w = 1/\sigma^2$.*

* Lists of structure factors and anisotropic thermal parameters have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 32929 (13 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 13 White Friars, Chester CH1 1NZ, England.

Discussion. The analysis shows that the major isomer, m.p. 183°C , formed in the Diels-Alder reaction is the title compound. Tables 1 and 2 show the final coordinates. Fig. 1 shows a perspective view of the molecule with the atomic nomenclature. Principal bond lengths and angles are listed in Tables 3 and 4. All bond lengths are in good agreement with accepted values (Kennard, 1962) and there are no intermolecular close contacts less than 3.5 \AA .

All calculations were carried out on a Univac 1106 computer at the University of Cape Town.

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References

- GILES, R. G. F. & ROOS, G. H. P. (1976). *J. Chem. Soc. Perkin Trans. 1*, pp. 2057-2060.
 KENNARD, O. (1962). *International Tables for X-ray Crystallography*, Vol. III, pp. 275-276. Birmingham: Kynoch Press.
 SHELDRICK, G. M. (1976). Private communication.
 WOLINSKY, J. & LOGIN, R. B. (1970). *J. Org. Chem.* **35**, 1987-1989.

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The Monoadduct between [4.4.3]Propella-2,4-diene and 4-Phenyl-1,2,4-triazoline-3,5-dione*

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Abstract. C₂₁H₂₃N₃O₂, monoclinic, $P2_1/c$, $a = 14.142$ (2), $b = 7.228$ (1), $c = 18.014$ (3) \AA , $\beta = 108.27$ (1) $^\circ$, $M_r = 349.43$, $Z = 4$, $D_x = 1.328 \text{ g cm}^{-3}$. The two six-membered rings are in the boat form, both folded towards the central five-membered ring. The 1,2,4-triazoline-3,5-dione ring is *anti* with respect to the latter. The bonding around the two vicinal atoms N(1) and N(3) is pyramidal, while around the third, N(2), it is planar. The cyclopentane ring has an envelope shape

with C(12) flapping towards the unsubstituted six-membered ring.

Introduction. The crystal structure of the title compound is of interest in connexion with stereochemical studies of Diels-Alder adducts of [4.4 X]propella-2,4-dienes ($X \equiv$ substituted rings) (Korat, Tatarsky & Ginsburg, 1972; Kalo, Vogel & Ginsburg, 1977).

Intensities from a colourless crystal, $0.4 \times 0.2 \times 0.2 \text{ mm}$, were collected on a semi-automatic Stoe-Weissenberg diffractometer with graphite-mono-

* Configuration of Diels-Alder Adducts. I.

Table 1. *Final fractional coordinates* ($\times 10^4$; for H $\times 10^3$)

E.s.d.'s are given in parentheses.

	x	y	z		x	y	z
O(1)	-538 (4)	8048 (6)	7130 (3)	H(2)	110 (7)	993 (10)	820 (6)
O(2)	104 (3)	2839 (6)	8663 (3)	H(3)	42 (7)	964 (11)	942 (6)
N(1)	852 (4)	7033 (7)	8116 (3)	H(4)	90 (8)	687 (11)	1012 (6)
N(2)	-445 (3)	5138 (6)	7705 (3)	H(5)	183 (7)	475 (11)	973 (6)
N(3)	1055 (4)	5374 (6)	8570 (3)	H(71)	272 (7)	481 (12)	852 (6)
C(1)	2390 (4)	8495 (8)	8881 (3)	H(72)	374 (8)	409 (12)	938 (6)
C(2)	1225 (4)	8727 (9)	8582 (4)	H(81)	439 (7)	512 (11)	857 (6)
C(3)	823 (5)	8629 (10)	9281 (4)	H(82)	460 (7)	643 (11)	934 (6)
C(4)	1053 (4)	7131 (9)	9698 (4)	H(91)	394 (8)	752 (11)	785 (6)
C(5)	1662 (4)	5795 (7)	9399 (3)	H(92)	426 (7)	907 (11)	864 (6)
C(6)	2648 (4)	6681 (8)	9372 (3)	H(101)	238 (8)	740 (11)	779 (6)
C(7)	3232 (5)	5300 (9)	9031 (4)	H(102)	262 (7)	952 (11)	783 (6)
C(8)	4129 (6)	6155 (11)	8861 (5)	H(111)	253 (8)	1115 (12)	955 (6)
C(9)	3849 (6)	7814 (11)	8363 (5)	H(112)	314 (8)	1139 (11)	910 (6)
C(10)	2771 (5)	8405 (10)	8164 (4)	H(121)	394 (8)	1002 (11)	1041 (6)
C(11)	2864 (5)	10104 (8)	9424 (4)	H(122)	437 (7)	878 (12)	999 (6)
C(12)	3669 (6)	9234 (12)	10125 (4)	H(131)	276 (7)	755 (11)	1056 (6)
C(13)	3276 (5)	7347 (8)	10219 (3)	H(132)	373 (7)	615 (11)	1049 (6)
C(14)	-116 (5)	6860 (8)	7596 (4)	H(17)	-61 (8)	163 (11)	715 (6)
C(15)	221 (5)	4281 (7)	8362 (4)	H(18)	-213 (7)	47 (11)	626 (6)
C(16)	-1341 (4)	4303 (8)	7205 (3)	H(19)	-358 (7)	229 (11)	594 (6)
C(17)	-1319 (5)	2483 (9)	6967 (4)	H(20)	-375 (7)	540 (11)	628 (6)
C(18)	-2192 (5)	1686 (8)	6462 (4)	H(21)	-216 (8)	666 (10)	712 (6)
C(19)	-3051 (5)	2788 (9)	6208 (4)				
C(20)	-3073 (5)	4539 (11)	6456 (4)				
C(21)	-2206 (5)	5360 (8)	6959 (3)				

chromatized Mo $K\alpha$ radiation ($\lambda = 0.71069 \text{ \AA}$) with the $\omega/2\theta$ scan technique; 2064 reflexions were measured, of which 1431 were significantly above background [$F_o > 3\sigma(F_o)$]. The structure was solved by direct methods with *MULTAN 74* (Main, Woolfson, Lessinger, Germain & Declercq, 1974) and refined by least-squares analysis in two blocks. H atoms were

located on a difference map. These were included in the refinement at positions calculated on the basis of the heavy-atom positions. Constant temperature factors ($U = 0.076 \text{ \AA}^2$) were assigned to the H atoms but not refined. Scattering factors for C, N, O and H were taken from Hanson, Herman, Lea & Skillman (1964). The final R for 1431 reflexions was 0.093. Final

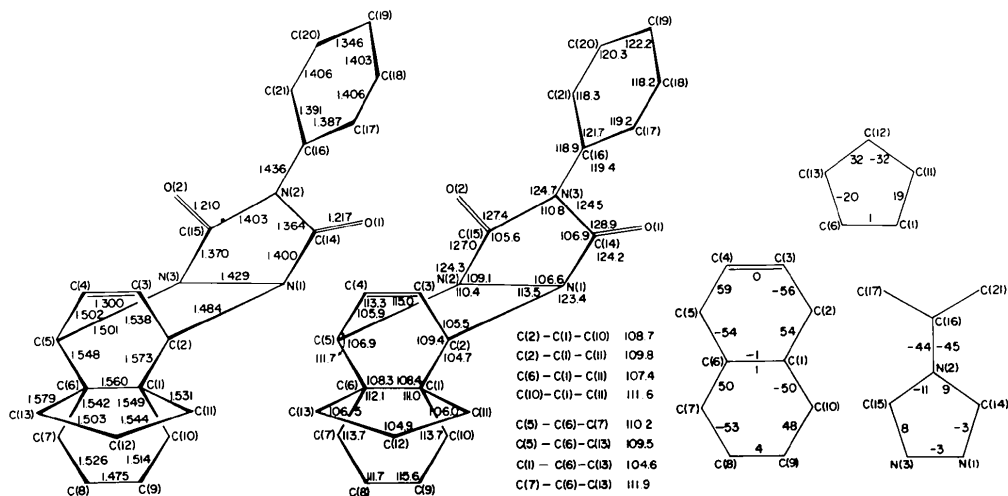


Fig. 1. Bond distances (Å), bond angles and torsion angles (°).

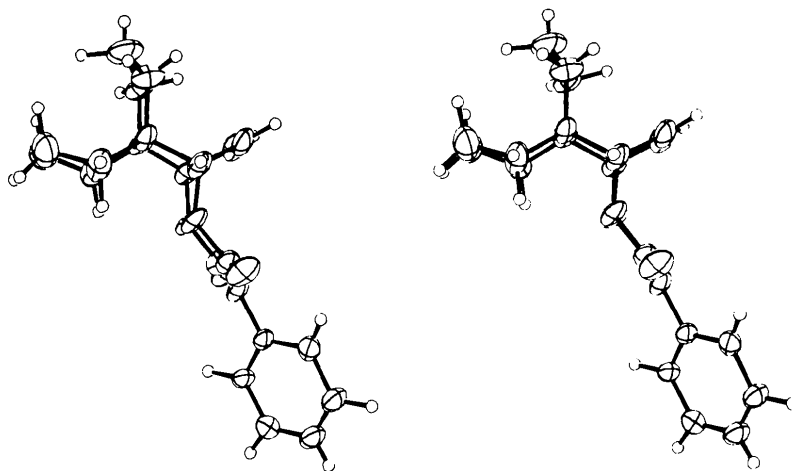


Fig. 2. Stereoscopic view of the molecule, showing vibration ellipsoids at the 50% probability level (Johnson, 1965).

positional parameters are listed in Table 1.* Corresponding interatomic distances, angles and torsion angles involving C, N and O atoms are shown in Fig. 1; the C—H distances (not shown) lie in the range 0.83–1.22 Å.

Discussion. The rather high value of R is probably due to systematic errors in the intensity measurements which may affect also the unexpected differences between chemically equivalent bond lengths and angles. The shortening of the non-bonded intramolecular C(2)···C(5) distance (2.544 Å) compared with C(7)···C(10) (2.695 Å) is due to the strain imposed by the substituent. This strain causes the closure of the bond angles at the atoms involved in the double bond,

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C(3) (115.0°) and C(4) (113.3°), and also the puckering of the substituted six-membered ring (see bond angles and torsion angles in Fig. 1).

The cyclopentane ring has an envelope shape with the central C(12) atom flapping towards the unsubstituted six-membered ring.

The 1,2,4-triazoline part has an envelope shape with N(2) flapping away from the C(3)=C(4) double bond (Fig. 2). The bonding around the N(2) atom is planar (the sum of the valency angles is 360.0°) while around the two vicinal N(1) and N(3) atoms it is pyramidal (the sums of the valency angles are 343.5 and 343.8° respectively). The values found in 4-phenyl-2,4,6-triazatricyclo[5.2.2.0^{2,6}]undecane-3,5-dione (I) are 359.9 , 350.3 , 353.7° (van der Ende, Offereins & Romers, 1974). The angle between the two mean planes described by atoms N(1), N(3), C(14), C(15) and N(1), N(3), C(2), C(5) is 139.5° in the title compound compared with 149.3° in (I).

The dihedral angle between the phenyl group and the

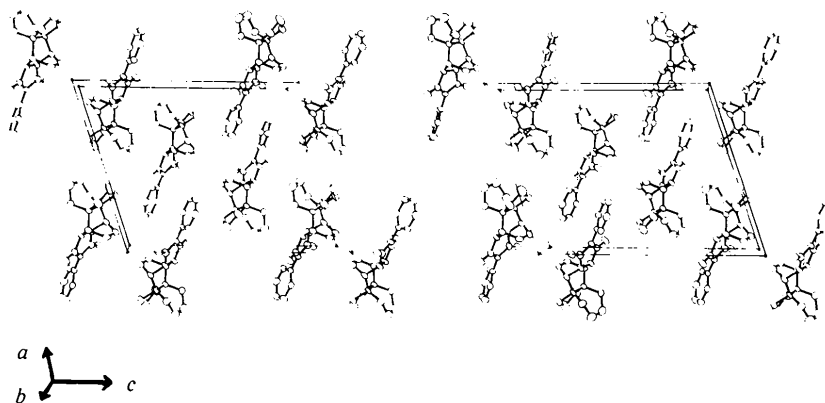


Fig. 3. Stereoscopic view of packing in the unit cell. The origin is at the lower left-hand corner of the unit cell.

mean plane of 1,2,4-triazacyclopentane is 45° compared with 75.3° in (I). The N(2)—C(16) bond length (1.436 Å) is therefore longer than the other N—C(sp^2) bonds in the molecule (1.364–1.403 Å). The differences can be ascribed to interaction of the N lone-pair electrons with the two carbonyl groups [C(14), C(15)] while there is no interaction with the phenyl group. The same difference was also observed by van der Ende, Offereins & Romers (1974); 1.431, 1.357–1.403 Å.

The packing of molecules in the unit cell is shown in Fig. 3. Intermolecular distances shorter than van der Waals contacts have not been found.

The author wishes to thank Professor D. Ginsburg and Dr J. Kalo for providing a sample of the compound.

References

- ENDE, C. VAN DER, OFFEREINS, B. & ROMERS, C. (1974). *Acta Cryst.* **B30**, 1947–1952.
 HANSON, H. P., HERMAN, F., LEA, J. D. & SKILLMAN, S. (1964). *Acta Cryst.* **17**, 1040–1044.
 JOHNSON, C. K. (1965). *ORTEP*. Report ORNL-3794. Oak Ridge National Laboratory, Tennessee.
 KALO, J., VOGEL, E. & GINSBURG, D. (1977). To be published.
 KORAT, M., TATARSKY, D. & GINSBURG, D. (1972). *Tetrahedron*, **28**, 2315–2332.
 MAIN, P., WOOLFSON, M. M., LESSINGER, L., GERMAIN, G. & DECLERCQ, J. P. (1974). *MULTAN 74. A System of Computer Programs for the Automatic Solution of Crystal Structures from X-ray Diffraction Data*. Univs. York, England, and Louvain, Belgium.

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The Monoadduct between 12-Thia[4.4.3]propella-2,4-diene *anti*-12-Oxide and 4-Phenyl-1,2,4-triazoline-3,5-dione*

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Abstract. $C_{20}H_{21}N_3O_3S$, monoclinic, $P2_1/n$, $a = 7.344$ (2), $b = 27.029$ (6), $c = 9.165$ (2) Å, $\beta = 100.75$ (3)°, $M_r = 383.48$, $Z = 4$, $D_x = 1.306$ g cm $^{-3}$. The two six-membered rings are in the boat form; the substituted ring is folded away from, the other ring towards, the central five-membered ring. The 4-phenyl-1,2,4-triazoline-3,5-dione ring is *syn* with respect to the latter. The central five-membered ring has an envelope shape with the S—O(1) bond flapping towards the unsubstituted six-membered ring.

Introduction. The configuration of the title compound is of interest in connexion with stereochemical studies of the Diels–Alder adducts of [4.4. X]propella-2,4-dienes ($X \equiv$ substituted rings) (Korat, Tatarsky & Ginsburg, 1972; Kalo, Vogel & Ginsburg, 1977). Intensities from a colourless crystal, $0.4 \times 0.3 \times 0.3$ mm, were collected on a semi-automatic Stoe–Weissenberg diffractometer with graphite-monochromatized Mo $K\alpha$ radiation ($\lambda = 0.71069$ Å) with the $\omega/2\theta$ scanning technique; 2295 reflexions were measured, of which 1562 were significantly above background [$F_o > 3\sigma(F_o)$]. The structure was solved by direct methods

with *MULTAN 74* (Main, Woolfson, Lessinger, Germain & Declercq, 1974) and refined by least-squares analysis in two blocks. H atoms were located on a difference map. These were included in the refinement at positions calculated on the basis of the heavy-atom positions. A constant temperature factor ($U = 0.076$ Å 2) was assigned to the H atoms; their positional and vibrational parameters were not refined. Scattering factors for S, C, N, O and H were taken from Hanson, Herman, Lea & Skillman (1964). The final R for 1562 reflexions was 0.104. The final positional parameters are listed in Table 1.† Corresponding interatomic distances, angles and torsion angles involving S, C, N and O atoms are shown in Fig. 1.

Discussion. The rather high value of R is probably due to systematic errors in the intensity measurements which may also affect the differences between chemically equivalent bond lengths and angles.

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* Configuration of Diels–Alder Adducts. II.