

N—N single bond of hydrazine (Morino, Iijima & Murata, 1960). It is also in agreement with the N—N bond length of 1.443 (3) Å present in the 2-phenylpyrazolidin-3-one studied by Ege *et al.* (1983). In contrast, the N1—N2 bond length in (4) is slightly longer than the 1.396 (9) Å N—N bond distance in 1,2-diacetylhydrazine (Shintani, 1960) and longer still when compared with the N—N bond length of 1.346 (6) Å in tetraformyl hydrazine (Hinderer & Hess, 1974).

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## Structures of Diels-Alder Reaction Adducts of Certain Propellanes with 4-Phenyl-1,2,4-triazoline-3,5-dione

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**Abstract.** 1a,8b-Dihydro-5-phenyl-2*H*,8*H*-2,8-etheno-1b,8a-(methanoxymethano)-4*H*-oxireno[3,4]cyclobuta[1,2-*d*][1,2,4]triazolo[1,2-*A*]pyridazine-4,6(5*H*)-dione: isomer (1), C<sub>18</sub>H<sub>15</sub>N<sub>3</sub>O<sub>4</sub>, *M<sub>r</sub>* = 337.33, orthorhombic, *P*2<sub>1</sub>2<sub>1</sub>2<sub>1</sub>, *a* = 23.866 (11), *b* = 9.571 (5), *c* = 6.686 (3) Å, *V* = 1527.22 Å<sup>3</sup>, *Z* = 4, *D<sub>x</sub>* = 1.467 g cm<sup>-3</sup>, Mo *K*α, λ = 0.71069 Å, μ = 0.991 cm<sup>-1</sup>, *F*(000) = 704, *R* = 0.059 for 1560 reflections; isomer (2), C<sub>18</sub>H<sub>15</sub>N<sub>3</sub>O<sub>4</sub>, *M<sub>r</sub>* = 337.33, orthorhombic, *P*2<sub>1</sub>2<sub>1</sub>2<sub>1</sub>, *a* = 24.104 (12), *b* = 9.774 (5), *c* = 6.463 (3) Å, *V* = 1522.63 Å<sup>3</sup>, *Z* = 4, *D<sub>x</sub>* = 1.461 g cm<sup>-3</sup>, Mo *K*α, λ = 0.71069 Å, μ = 0.994 cm<sup>-1</sup>, *F*(000) = 704, *R* = 0.095 for 866 reflections. 1a,8b-Dihydro-1a,8b-dimethyl-5-phenyl-2*H*,8*H*-2,8-etheno-1b,8a-(methanoxymethano)-4*H*-oxireno[3,4]cyclobuta[1,2-*d*][1,2,4]triazolo[1,2-*a*]pyridazine-4,6(5*H*)-dione: isomer (3), C<sub>20</sub>H<sub>19</sub>N<sub>3</sub>O<sub>4</sub>, *M<sub>r</sub>* = 365.39, orthorhombic, *P*2<sub>1</sub>2<sub>1</sub>2<sub>1</sub>, *a* = 13.096 (6), *b* =

12.653 (6), *c* = 10.696 (5) Å, *V* = 1772.36 Å<sup>3</sup>, *Z* = 4, *D<sub>x</sub>* = 1.369 g cm<sup>-3</sup>, Mo *K*α, λ = 0.71069 Å, μ = 0.908 cm<sup>-1</sup>, *F*(000) = 768, *R* = 0.066 for 1758 reflections; isomer (4), C<sub>20</sub>H<sub>19</sub>N<sub>3</sub>O<sub>4</sub>, *M<sub>r</sub>* = 365.39, monoclinic, *P*2<sub>1</sub>/*c*, *a* = 26.724 (13), *b* = 11.985 (6), *c* = 11.453 (6) Å, β = 99.96 (2)°, *V* = 3612.96 Å<sup>3</sup>, *Z* = 8, *D<sub>x</sub>* = 1.343 g cm<sup>-3</sup>, Mo *K*α, λ = 0.71069 Å, μ = 0.891 cm<sup>-1</sup>, *F*(000) = 1536, *R* = 0.036 for 2649 reflections. 13-Hydroxy-13-methyl-14-methylene-2-phenyl-6*H*,9*H*-5a,8a-ethano-5,9-etheno-1*H*,5*H*,8*H*-furo[3,4-*d*][1,2,4]triazolo[1,2-*a*]pyridazine-1,3(2*H*)-dione (5), C<sub>20</sub>H<sub>19</sub>N<sub>3</sub>O<sub>4</sub>, *M<sub>r</sub>* = 365.39, monoclinic, *P*2<sub>1</sub>/*c*, *a* = 13.474 (7), *b* = 11.422 (6), *c* = 11.534 (6) Å, β = 97.73 (2)°, *V* = 1758.95 Å<sup>3</sup>, *Z* = 4, *D<sub>x</sub>* = 1.379 g cm<sup>-3</sup>, Mo *K*α, λ = 0.71069 Å, μ = 0.915 cm<sup>-1</sup>, *F*(000) = 768, *R* = 0.070 for 2095 reflections. Crystal structures of the Diels-Alder adducts of 4-phenyl-1,2,4-triazoline-3,5-dione with various

Table 1. *Crystallographic and experiential details*

	(1)	(2)	(3)	(4)	(5)
Crystal shape	Prism	Needle	Needle	Needle	Needle
Crystal size (mm)	0.5 × 0.3 × 0.2	0.3 × 0.1 × 0.1	0.3 × 0.2 × 0.2	0.4 × 0.3 × 0.2	0.5 × 0.4 × 0.2
$\Delta\omega$ (°)	1.2	1.2	1.15	1.2	1.0
Scan time (s)	24	48	23	*	20
Background time (s)	20	20	20	*	20
$\theta_{\max}$ (°)	25	21	25	22	24
<i>hkl</i> range					
<i>h</i>	0→8	0→25	0→15	-28→27	-15→15
<i>k</i>	0→11	0→10	0→15	0→12	0→13
<i>l</i>	0→7	0→6	0→12	0→12	0→13
Reflections measured	1581	1159	1809	4412	3070
Unique reflections	1560	1076	1758	4410	2680
Significant†	1560	866	1758	2649	2095
Weighting coefficients‡					
<i>k</i>	0.4701	0.9536	0.6398	1.0000	1.3815
<i>g</i>	0.0042	0.0186	0.0040	0.0436	0.0008
$(\Delta/\sigma)_{\max}$	0.7	0.1	0.3	0.9	1.0
$(\Delta\rho)_{\max}$ (e Å <sup>-3</sup> )	0.3	0.4	0.3	0.2	0.2
$(\Delta\rho)_{\min}$ (e Å <sup>-3</sup> )	-0.4	-0.4	-0.2	-0.4	-0.3
<i>wR</i>	0.063	0.097	0.070	0.036	0.067
<i>R</i>	0.059	0.095§	0.066	0.036	0.070

\* The data were measured with a CAD-4 diffractometer with variable scan time and background.

† (1)  $F_o > 0.0$ ; (2)  $F_o > 2\sigma(F_o)$ ; (3)  $F_o > 0.0$ ; (4)  $F_o > 6\sigma(F_o)$ ; (5)  $F_o > 1.5\sigma(F_o)$ .

‡  $w = k/[\sigma^2(F_o) + gF_o^2]$ .

§ The amount of available material was small and the crystals obtained were of poor quality (thin-needle, poorly diffracted).

related propellanes consisting of a cyclohexadiene ring and four- and five-membered rings show that the title dienophile attack is, as predicted, *syn* to the five-membered ether ring whose  $\alpha$ -H atoms exert less repulsion than the H or the O atoms of the  $\alpha$ -epoxy of the cyclobutane ring.

**Introduction.** Several propellane substrates have been used in the study of the steric effects exerted by different substituents on the direction of attack by a dienophile in a Diels-Alder reaction. The propellanes prepared by Maier, Sayrac & Reisenauer (1982) have been used to modulate the competition between various rings in exerting larger (or smaller) steric repulsion for the approach of 4-substituted 1,2,4-triazoline-3,5-diones which undergo a Diels-Alder reaction with a cyclohexadiene ring (Ashkenazi, Kafatory, Sayrac, Maier & Ginsburg, 1984, 1986). It has been shown by Gleiter & Ginsburg (1979) that a larger repulsion is exerted, for example, by a cyclopropane ring than by a cyclobutane, a cyclopentane, or a cyclohexane ring in the same propelladiene molecule (Ashkenazi, Kafatory, Grimme, Heger, Vogel & Ginsburg, 1979), or by a tetrahydrofuran ring than by a tetrahydrothiophene ring (Ashkenazi, Olikier & Ginsburg, 1978), or by a cyclobutane ring rather than a six-membered ring in various oxidation states in the same propelladiene (Kalo, Photis, Paquette, Vogel & Ginsburg, 1976). We compare here the molecular structures of five adducts all having in common a propellane skeleton possessing four-, five- and six-membered rings.

**Experimental.** The various compounds were obtained by a Diels-Alder reaction of 4-substituted 1,2,4-triazoline-3,4-diones with the cyclohexadiene deriva-

Table 2. *Positional parameters ( $\times 10^4$ ) and equivalent isotropic displacement parameters ( $\text{\AA}^2 \times 10^3$ ) for (1) with e.s.d.'s in parentheses*

	$U_{\text{eq}} = \frac{1}{3} \text{trace } U.$			
	<i>x</i>	<i>y</i>	<i>z</i>	$U_{\text{eq}}$
O(1)	6356 (1)	3401 (4)	8082 (6)	55 (1)
O(2)	7539 (1)	47 (4)	5595 (6)	44 (1)
O(3)	5082 (2)	-648 (5)	2857 (5)	60 (1)
O(4)	5443 (2)	-2159 (5)	8380 (7)	67 (1)
N(1)	6219 (1)	1117 (4)	7115 (6)	36 (1)
N(2)	7085 (1)	1936 (3)	7097 (6)	33 (1)
N(3)	6587 (1)	59 (4)	6293 (5)	32 (1)
C(1)	5678 (2)	1229 (5)	5978 (8)	40 (1)
C(2)	5844 (2)	1483 (6)	3830 (9)	49 (1)
C(3)	6194 (2)	447 (7)	3089 (8)	50 (1)
C(4)	6334 (2)	-622 (5)	4497 (7)	36 (1)
C(5)	5802 (2)	-1323 (5)	5298 (8)	39 (1)
C(6)	5409 (2)	-197 (5)	6237 (7)	37 (1)
C(7)	6532 (2)	2292 (4)	7487 (7)	35 (1)
C(8)	7121 (2)	630 (4)	6227 (7)	47 (1)
C(9)	7555 (2)	2834 (4)	7484 (7)	35 (1)
C(10)	7600 (2)	3458 (5)	9341 (9)	46 (1)
C(11)	8053 (2)	4319 (6)	9698 (12)	62 (1)
C(12)	8455 (2)	4532 (6)	8245 (13)	66 (2)
C(13)	8405 (2)	3917 (5)	6430 (12)	66 (2)
C(14)	7948 (2)	3044 (5)	6001 (9)	47 (1)
C(15)	5304 (2)	-1776 (7)	3956 (8)	53 (1)
C(16)	4934 (2)	-748 (6)	4901 (7)	44 (1)
C(17)	5888 (2)	-2376 (6)	6962 (10)	54 (1)
C(18)	5321 (2)	-674 (7)	8373 (8)	54 (1)

tives. Crystallographic data, details of intensity measurements and structure refinement are given in Table 1. Densities were not measured. A Philips PW 1100 four-circle computer-controlled diffractometer with graphite-monochromated Mo  $K\alpha$  radiation was used for data collection; measurements by  $\theta/2\theta$ -scan mode [for compound (4), a CAD-4 diffractometer was used]. Lattice parameters were calculated using 25 reflections for each compound [ $\theta$  range: (1) 4→13; (2) 3→12; (3) 5→13; (4) 6→15; (5) 3→10]. Three reflections (for each compound) were monitored and showed variation in intensities of *ca* 5%. Absorption and extinction corrections

Table 3. Positional parameters ( $\times 10^4$ ) and equivalent isotropic displacement parameters ( $\text{\AA}^2 \times 10^3$ ) for (2) with e.s.d.'s in parentheses

	$U_{eq} = \frac{1}{3} \text{trace } U.$			
	x	y	z	$U_{eq}$
O(1)	6340 (3)	3439 (9)	8254 (15)	51 (2)
O(2)	7525 (4)	49 (11)	6123 (13)	50 (2)
O(3)	4892 (5)	-1952 (11)	4830 (17)	78 (3)
O(4)	5448 (5)	-1868 (11)	8843 (16)	80 (4)
N(1)	6206 (4)	1182 (9)	7381 (15)	38 (2)
N(2)	7071 (4)	1955 (8)	7509 (13)	30 (2)
N(3)	6579 (4)	133 (10)	6688 (14)	37 (3)
C(1)	5698 (5)	1293 (12)	6042 (19)	41 (3)
C(2)	5900 (5)	1410 (14)	3816 (20)	47 (3)
C(3)	6235 (5)	434 (14)	3199 (19)	48 (4)
C(4)	6361 (5)	-655 (12)	4882 (18)	39 (3)
C(5)	5826 (5)	-1273 (11)	5612 (18)	38 (3)
C(6)	5408 (5)	-84 (11)	6332 (18)	35 (3)
C(7)	6528 (5)	2350 (11)	7708 (18)	32 (3)
C(8)	7114 (5)	656 (11)	6637 (16)	28 (3)
C(9)	7532 (5)	2821 (21)	7917 (20)	47 (4)
C(10)	7572 (5)	3407 (12)	9876 (21)	44 (3)
C(11)	8029 (6)	4232 (14)	10274 (23)	55 (4)
C(12)	8416 (6)	4527 (12)	8788 (29)	63 (5)
C(13)	8358 (7)	3936 (16)	6789 (25)	67 (4)
C(14)	7923 (5)	3073 (14)	6376 (22)	55 (3)
C(15)	5397 (6)	1657 (17)	3911 (21)	62 (5)
C(16)	5009 (6)	-547 (17)	4557 (27)	61 (4)
C(17)	5860 (6)	-2230 (16)	7444 (21)	62 (4)
C(18)	5276 (7)	-514 (16)	8499 (22)	68 (5)

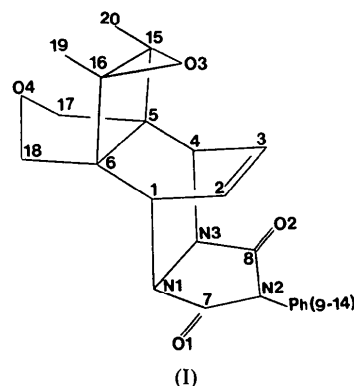
Table 4. Positional parameters ( $\times 10^4$ ) and equivalent isotropic displacement parameters ( $\text{\AA}^2 \times 10^3$ ) for (3) with e.s.d.'s in parentheses

	$U_{eq} = \frac{1}{3} \text{trace } U.$			
	x	y	z	$U_{eq}$
O(1)	512 (3)	2102 (3)	10067 (3)	61 (1)
O(2)	-219 (3)	120 (4)	6629 (4)	80 (1)
O(3)	4698 (2)	1505 (3)	6467 (3)	54 (1)
O(4)	3415 (3)	-134 (3)	8350 (3)	54 (1)
N(1)	1532 (3)	1367 (3)	8516 (3)	41 (1)
N(2)	-111 (3)	980 (3)	8548 (4)	45 (1)
N(3)	1296 (3)	740 (3)	7441 (3)	44 (1)
C(1)	2264 (3)	2230 (3)	8210 (4)	38 (1)
C(2)	1817 (3)	2841 (4)	7142 (5)	47 (1)
C(3)	1599 (4)	2270 (4)	6149 (4)	47 (1)
C(4)	1840 (3)	1098 (4)	6301 (4)	43 (1)
C(5)	2971 (3)	945 (4)	6633 (4)	39 (1)
C(6)	3241 (3)	1650 (3)	7778 (4)	36 (1)
C(7)	623 (3)	1562 (4)	9152 (2)	44 (1)
C(8)	258 (4)	573 (4)	7420 (5)	55 (1)
C(9)	-1151 (3)	874 (4)	8922 (4)	44 (1)
C(10)	-1894 (4)	218 (4)	9907 (5)	50 (1)
C(11)	-2426 (4)	124 (5)	10236 (6)	62 (1)
C(12)	-3168 (4)	653 (5)	9585 (5)	61 (2)
C(13)	-2905 (4)	1277 (5)	8629 (5)	67 (2)
C(14)	-1894 (4)	1417 (5)	8284 (5)	63 (1)
C(15)	3727 (3)	1610 (4)	5849 (4)	47 (1)
C(16)	3971 (3)	2287 (4)	6923 (4)	42 (1)
C(17)	3266 (4)	-169 (4)	7016 (5)	54 (1)
C(18)	3697 (4)	900 (4)	8733 (5)	35 (1)
C(19)	4333 (4)	3401 (4)	7054 (5)	61 (1)
C(20)	3722 (4)	1789 (5)	4468 (4)	65 (1)

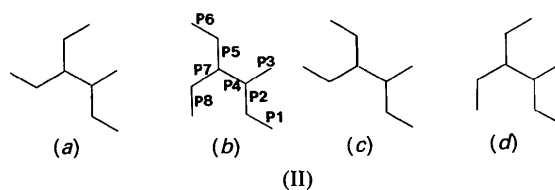
were not applied. Structures were solved either by *MULTAN80* (Main *et al.*, 1980) or by *SHELX76* (Sheldrick, 1976), and refined by full-matrix least squares (based on *F*) with anisotropic atomic displacement parameters for O, N and C atoms, isotropic for H. All H-atom positions (see structural commentary) were located either from difference Fourier maps or calculated (Sheldrick, 1976). Atomic scattering factors and  $f'$ ,  $f''$  values were obtained

from *International Tables for X-ray Crystallography* (1974, Vol. IV).

**Discussion.** Final positional parameters for (1)–(5) are given in Tables 2–6 respectively [atomic notation in (1)].\* Molecular stereoviews for the molecules (1)–(5) are shown in Figs. 1–5, respectively. A comparison of bond lengths and angles is given in Tables 7 and 8.



**Molecular conformation.** A schematic drawing of the molecule projected along the propellane bridging atoms is shown below (II) with the notation of various planes and the four possible conformations with respect to the bonding of the epoxide and ether O atoms. Comparison of the angles between the planes is given in Table 9. Three compounds, (1) (4) and (5) (in the latter we consider the direction of the C—OH bond), adopt the (*a*)-type conformation while (2) adopts conformation (*c*) and (3) adopts a (*d*)-type conformation. The most significant differences in the angles between the various planes are in *P4—P5* and *P5—P7*. While the *P4—P7* angle is similar in all, *P4—P5* opens up, *P5—P7* closes for compounds (1) and (4) (*A* and *B*) compared with (2) and (3). It seems that the epoxide O atom (and its lone-pair electron) is exerting steric repulsion on the C(2)=C(3) double bond. Also, as expected, the angle between *P7* and *P8* in (2) opens up owing to the steric repulsion in the (*c*)-type conformation.



\* Lists of structure factors, atomic displacement parameters and H-atom parameters have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 55077 (59 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England. [CIF reference: GE0276]

Table 5. Positional parameters ( $\times 10^4$ ) and equivalent isotropic displacement parameters ( $\text{\AA}^2 \times 10^4$ ) for (4) with *e.s.d.*'s in parentheses

Molecule A	$U_{\text{eq}} = \frac{1}{3} \text{ trace } U$			$U_{\text{eq}}$
	x	y	z	
O(1)	-599 (1)	-475 (2)	1028 (2)	541 (5)
O(2)	-780 (1)	2425 (2)	3451 (2)	528 (5)
O(3)	1317 (1)	2015 (2)	2517 (2)	552 (5)
O(4)	1087 (1)	-59 (2)	5158 (2)	529 (5)
N(1)	-60 (1)	270 (2)	2650 (2)	354 (5)
N(2)	853 (1)	924 (2)	2192 (2)	380 (5)
N(3)	-121 (1)	1212 (2)	3416 (2)	344 (5)
C(1)	415 (1)	390 (2)	2104 (2)	372 (6)
C(2)	347 (1)	1482 (2)	1464 (2)	435 (7)
C(3)	280 (1)	2338 (3)	2136 (2)	405 (7)
C(4)	297 (1)	2063 (2)	3413 (2)	344 (6)
C(5)	782 (1)	1465 (2)	3940 (2)	348 (6)
C(6)	857 (1)	440 (2)	3158 (2)	386 (6)
C(7)	-510 (1)	179 (2)	1851 (2)	376 (7)
C(8)	-606 (1)	1626 (2)	3068 (2)	370 (6)
C(9)	-1356 (1)	1112 (2)	1580 (2)	390 (7)
C(10)	1431 (1)	1545 (3)	449 (3)	566 (8)
C(11)	-1913 (1)	1765 (3)	-119 (3)	722 (11)
C(12)	-2325 (1)	1548 (3)	428 (4)	753 (11)
C(13)	-2243 (1)	1116 (3)	1543 (4)	739 (11)
C(14)	-1762 (1)	885 (3)	2133 (3)	583 (3)
C(15)	1308 (1)	1906 (2)	3768 (2)	448 (7)
C(16)	1383 (1)	923 (2)	3035 (2)	486 (7)
C(17)	810 (1)	958 (3)	5169 (2)	430 (7)
C(18)	907 (1)	-542 (2)	4011 (3)	493 (8)
C(19)	1851 (2)	292 (4)	2879 (5)	851 (14)
C(20)	1647 (1)	2661 (4)	4567 (3)	593 (9)

Molecule B	$U_{\text{eq}} = \frac{1}{3} \text{ trace } U$			$U_{\text{eq}}$
	x	y	z	
O(1)	4273 (1)	2570 (2)	5974 (2)	497 (5)
O(2)	4294 (1)	-536 (2)	8284 (2)	559 (5)
O(3)	6357 (1)	1480 (2)	8657 (2)	642 (5)
O(4)	5997 (1)	-310 (2)	5658 (2)	519 (5)
N(1)	4882 (1)	1213 (1)	6545 (2)	334 (5)
N(2)	4125 (1)	1003 (2)	7061 (2)	378 (5)
N(3)	4887 (1)	227 (2)	7273 (2)	347 (5)
C(1)	5338 (1)	1942 (2)	6939 (2)	398 (7)
C(2)	5342 (1)	2158 (3)	8235 (3)	496 (8)
C(3)	5359 (1)	1254 (3)	8883 (3)	512 (8)
C(4)	5357 (1)	177 (2)	8211 (2)	389 (6)
C(5)	5798 (1)	135 (2)	7518 (2)	361 (6)
C(6)	5792 (1)	1224 (2)	6763 (2)	394 (6)
C(7)	4410 (1)	1706 (2)	6470 (2)	360 (6)
C(8)	4422 (1)	162 (2)	7634 (2)	380 (6)
C(9)	3601 (1)	1179 (2)	7146 (2)	418 (7)
C(10)	3479 (1)	1530 (2)	8200 (3)	496 (8)
C(11)	2975 (1)	1722 (3)	8272 (3)	619 (9)
C(12)	2604 (1)	1545 (3)	7303 (3)	679 (10)
C(13)	2733 (1)	1189 (3)	6265 (3)	763 (11)
C(14)	3234 (1)	1006 (3)	6171 (3)	639 (9)
C(15)	6352 (1)	414 (2)	8084 (2)	481 (7)
C(16)	6338 (1)	1454 (2)	7396 (3)	520 (7)
C(17)	5781 (1)	-784 (2)	6604 (2)	443 (7)
C(18)	5785 (1)	787 (3)	5519 (2)	481 (8)
C(19)	6740 (5)	2099 (5)	6914 (6)	899 (15)
C(20)	6760 (1)	-401 (4)	8534 (4)	716 (11)

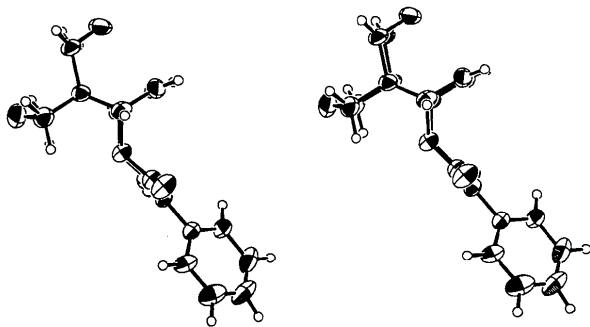


Fig. 1. Stereoscopic view of (1), down C(1)—C(4).

Table 6. Positional parameters ( $\times 10^4$ ) and equivalent isotropic displacement parameters ( $\text{\AA}^2 \times 10^4$ ) for (5) with *e.s.d.*'s in parentheses

	$U_{\text{eq}} = \frac{1}{3} \text{ trace } U$			$U_{\text{eq}}$
	x	y	z	
C(1)	4145 (2)	-270 (3)	-4148 (3)	575 (8)
O(2)	4001 (2)	2332 (2)	-1170 (2)	474 (7)
O(3)	8037 (2)	754 (2)	141 (2)	502 (8)
O(4)	7683 (2)	2484 (3)	-3340 (2)	570 (8)
N(1)	5358 (2)	836 (3)	-3037 (3)	399 (8)
N(2)	3772 (2)	1177 (3)	-2858 (3)	405 (8)
N(3)	5307 (2)	1665 (3)	-2107 (3)	375 (8)
C(1)	6184 (3)	-36 (3)	-2732 (3)	368 (9)
C(2)	6062 (3)	-530 (3)	-1557 (4)	417 (10)
C(3)	6054 (3)	262 (4)	-720 (3)	411 (10)
C(4)	6144 (3)	1505 (3)	-1134 (3)	348 (9)
C(5)	7105 (3)	1671 (3)	-1693 (3)	339 (9)
C(6)	7150 (3)	686 (3)	-2626 (3)	348 (9)
C(7)	4402 (3)	484 (4)	-3430 (3)	412 (10)
C(8)	4322 (3)	1774 (3)	-1935 (3)	373 (10)
C(9)	2713 (3)	1162 (3)	-3127 (4)	422 (10)
C(10)	2131 (3)	884 (4)	-2276 (4)	502 (11)
C(11)	1105 (4)	823 (4)	-2573 (5)	671 (14)
C(12)	664 (4)	1024 (5)	-3697 (5)	719 (15)
C(13)	1260 (4)	1313 (5)	-4536 (4)	737 (15)
C(14)	2285 (3)	1394 (4)	-4257 (4)	541 (12)
C(15)	8104 (3)	1191 (3)	-997 (3)	386 (9)
C(16)	8063 (3)	196 (3)	-1878 (3)	367 (9)
C(17)	7177 (3)	2806 (3)	-2369 (3)	457 (10)
C(18)	7325 (3)	1347 (4)	-3734 (3)	454 (11)
C(19)	8595 (3)	-756 (4)	-1934 (4)	575 (12)
C(20)	9014 (3)	1983 (4)	-951 (4)	556 (11)

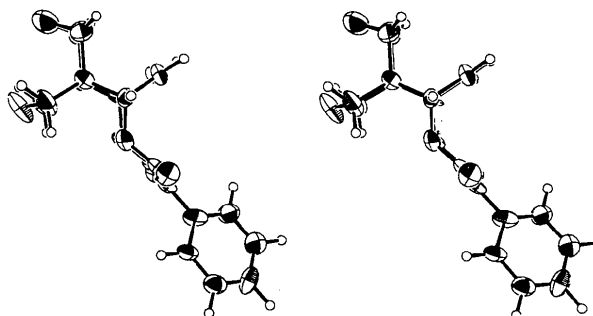


Fig. 2. Stereoscopic view of (2), down C(1)—C(4).

The degree of planarity of the triazolidione-bonded N atoms [N(1) and N(3)] is measured by  $\alpha_{\text{av}}$  [the average of the bond angle at N(1) and N(3)] (Kaftory & Agmon, 1984).  $\alpha_{\text{av}}$  values range from 111.1 to 114.0° and are somewhat larger (as expected) than those observed in compounds where the triazolidione is bonded to a cyclopentene [see (I) and (III) in Agmon, Kaftory, Nelson & Blackstock (1986) where  $\alpha_{\text{av}}$  is 110.5 and 111.2°].

**Packing.** The packing of molecules in (5) is interesting (see Fig. 6). The molecules are arranged in a zigzag manner along the *c* axis, interacting by a hydrogen bond between the hydroxyl group and the ether O atom. The O(3)⋯O(4) distance is 2.752 (5) Å and the O(3)⋯H⋯O(4) angle is 169.9 (3)°.

**Bond lengths and angles.** Most chemically equivalent bond lengths in the five compounds are similar

[except for those in (5) which are affected by the absence of the three-membered epoxy ring], but one significant difference should be noted. The O(3)—C(15) [or C(16) in the three-membered epoxy ring is longer in (3) and (4) (1.435 – 1.457 Å) than in (1) and (2) (1.39 – 1.415 Å). The major chemical difference between these two groups of systems is the replacement of the two H atoms in the three-membered epoxy ring [in (1) and (2)] by methyl groups [in (3) and (4)]. The effects of substitutions in small rings are known and have been carefully studied by Allen (1982).

Some significant differences between bond angles are observed, especially those between bonds within different planes. These differences are connected to

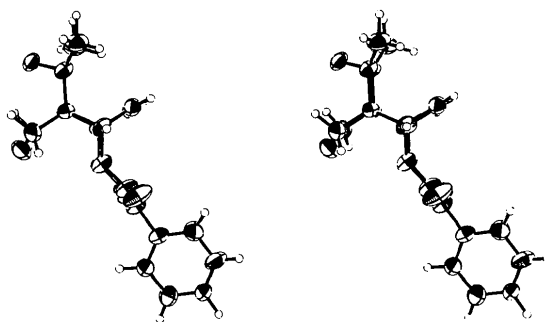


Fig. 3. Stereoscopic view of (3), down C(1)—C(4).

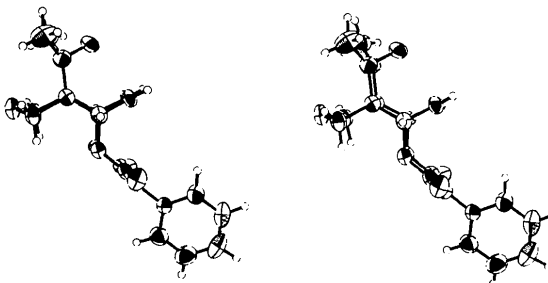


Fig. 4. Stereoscopic view of (4), down C(1)—C(4).

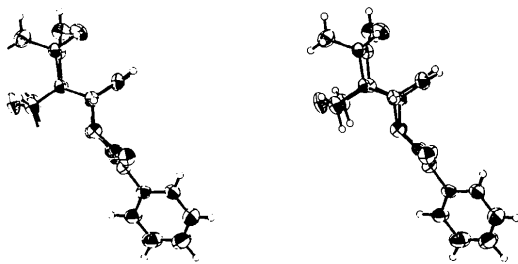


Fig. 5. Stereoscopic view of (5), down C(1)—C(4).

Table 7. Comparison of bond lengths (Å)

	(1)	(2)	(3)	(4A)	(4B)	(5)
O(1)—C(7)	1.209 (5)	1.21 (1)	1.202 (5)	1.218 (3)	1.207 (2)	1.212 (4)
O(2)—C(8)	1.219 (5)	1.20 (1)	1.198 (6)	1.206 (3)	1.209 (3)	1.215 (4)
O(3)—C(15)	1.410 (7)	1.39 (1)	1.440 (5)	1.444 (3)	1.435 (3)	1.419 (4)
O(3)—C(16)	1.415 (6)	1.41 (1)	1.457 (5)	1.435 (3)	1.436 (3)	
O(4)—C(17)	1.438 (7)	1.39 (1)	1.441 (6)	1.429 (3)	1.429 (3)	1.435 (5)
O(4)—C(18)	1.450 (6)	1.40 (1)	1.419 (6)	1.440 (3)	1.430 (3)	1.437 (4)
N(1)—N(3)	1.449 (4)	1.43 (1)	1.431 (5)	1.456 (2)	1.446 (2)	1.440 (4)
N(1)—C(1)	1.503 (5)	1.50 (1)	1.498 (5)	1.516 (3)	1.504 (3)	1.500 (4)
N(1)—C(7)	1.371 (5)	1.40 (1)	1.393 (5)	1.385 (3)	1.383 (3)	1.368 (4)
N(2)—C(7)	1.388 (5)	1.37 (1)	1.372 (5)	1.383 (3)	1.387 (3)	1.390 (5)
N(2)—C(8)	1.381 (5)	1.39 (1)	1.398 (6)	1.386 (3)	1.377 (2)	1.391 (4)
N(2)—C(9)	1.437 (5)	1.42 (1)	1.426 (5)	1.422 (2)	1.439 (3)	1.420 (4)
N(3)—C(4)	1.493 (5)	1.49 (1)	1.483 (5)	1.512 (3)	1.505 (2)	1.491 (4)
N(3)—C(8)	1.386 (5)	1.39 (1)	1.377 (5)	1.381 (2)	1.379 (3)	1.375 (5)
C(1)—C(2)	1.510 (8)	1.52 (1)	1.504 (6)	1.496 (3)	1.504 (4)	1.497 (5)
C(1)—C(6)	1.518 (6)	1.53 (1)	1.535 (5)	1.537 (3)	1.530 (3)	1.533 (5)
C(2)—C(3)	1.514 (7)	1.51 (1)	1.515 (6)	1.513 (4)	1.510 (4)	1.523 (5)
C(3)—C(4)	1.508 (7)	1.55 (2)	1.526 (6)	1.492 (3)	1.502 (3)	1.509 (5)
C(4)—C(5)	1.533 (6)	1.50 (1)	1.535 (5)	1.513 (3)	1.532 (3)	1.534 (5)
C(5)—C(6)	1.561 (6)	1.61 (1)	1.557 (6)	1.554 (3)	1.565 (3)	1.564 (3)
C(5)—C(15)	1.550 (6)	1.55 (1)	1.546 (6)	1.544 (3)	1.545 (3)	1.570 (4)
C(5)—C(17)	1.516 (7)	1.51 (1)	1.518 (6)	1.524 (3)	1.515 (3)	1.522 (5)
C(6)—C(16)	1.537 (6)	1.56 (1)	1.550 (5)	1.547 (3)	1.538 (3)	1.511 (4)
C(6)—C(18)	1.514 (7)	1.50 (1)	1.516 (6)	1.521 (3)	1.515 (3)	1.530 (5)
C(9)—C(10)	1.381 (7)	1.39 (1)	1.379 (6)	1.378 (3)	1.370 (4)	1.374 (6)
C(9)—C(14)	1.380 (6)	1.39 (1)	1.373 (7)	1.376 (4)	1.368 (3)	1.377 (5)
C(10)—C(11)	1.380 (7)	1.39 (1)	1.403 (7)	1.365 (4)	1.382 (4)	1.381 (6)
C(11)—C(12)	1.381 (9)	1.37 (2)	1.369 (7)	1.382 (5)	1.371 (4)	1.370 (7)
C(12)—C(13)	1.35 (1)	1.42 (2)	1.337 (8)	1.361 (5)	1.363 (5)	1.378 (7)
C(13)—C(14)	1.403 (7)	1.37 (2)	1.386 (7)	1.373 (4)	1.378 (4)	1.379 (6)
C(15)—C(16)	1.466 (7)	1.49 (2)	1.468 (6)	1.481 (3)	1.471 (3)	1.521 (5)
C(15)—C(20)			1.494 (6)	1.480 (4)	1.489 (4)	1.519 (5)
C(16)—C(19)			1.494 (7)	1.499 (5)	1.502 (6)	1.309 (5)

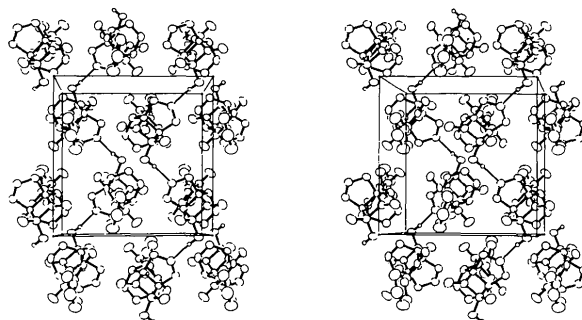


Fig. 6. Stereoscopic view of the packing of (5).

the various conformations. For example, the bond angle C(1)—C(6)—C(16) in (2) and (3) is smaller by *ca* 6° than the same angle in (1) and (4); consistent with the small angle between *P*<sub>4</sub> and *P*<sub>5</sub> in (2) and (3) compared with (1) and (4). Another example is the bond angles C(15)—C(5)—C(17) and C(16)—C(6)—C(18) which are larger by up to 8° in (2) and (3) compared with (1) and (4); these are fully consistent with the differences in angles between planes *P*<sub>5</sub> and *P*<sub>7</sub>.

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Table 8. Comparison of bond angles (°)

	(1)	(2)	(3)	(4A)	(4B)	(5)
C(15)—O(3)—C(16)	62.5 (3)	64 (1)	60.9 (2)	61.9 (1)	61.7 (1)	
C(17)—O(4)—C(18)	106.7 (4)	111.0 (1)	110.5 (3)	105.0 (2)	104.2 (1)	107.7 (2)
N(3)—N(1)—C(1)	112.3 (3)	112.5 (8)	111.7 (3)	111.4 (1)	112.1 (1)	111.7 (2)
N(3)—N(1)—C(7)	108.2 (3)	106.5 (8)	107.9 (3)	106.4 (1)	107.6 (1)	107.6 (2)
C(1)—N(1)—C(7)	120.0 (3)	118.7 (9)	122.2 (3)	115.4 (1)	117.0 (1)	121.2 (3)
C(7)—N(2)—C(8)	111.1 (3)	111.6 (9)	111.3 (3)	109.9 (2)	111.0 (1)	110.3 (3)
C(7)—N(2)—C(9)	124.2 (3)	124.2 (9)	126.0 (3)	125.4 (2)	124.5 (2)	123.3 (3)
C(8)—N(2)—C(9)	124.7 (3)	123.9 (9)	122.5 (3)	123.5 (1)	124.3 (1)	126.1 (3)
N(1)—N(3)—C(4)	111.4 (3)	113.1 (9)	112.8 (3)	111.2 (1)	111.6 (1)	111.9 (2)
N(1)—N(3)—C(8)	107.0 (3)	109.1 (8)	108.1 (3)	107.7 (1)	107.5 (1)	108.1 (2)
C(4)—N(3)—C(8)	121.2 (3)	119.9 (9)	120.5 (3)	114.5 (1)	117.9 (1)	123.3 (2)
N(1)—C(1)—C(2)	105.5 (3)	106.7 (9)	106.3 (3)	104.2 (1)	105.2 (2)	107.1 (2)
N(1)—C(1)—C(6)	104.0 (3)	103.8 (9)	104.6 (3)	105.3 (1)	104.7 (1)	104.9 (2)
C(2)—C(1)—C(6)	111.4 (4)	109 (1)	110.2 (3)	111.3 (2)	111.0 (2)	109.0 (3)
C(1)—C(2)—C(3)	114.6 (4)	115 (1)	114.7 (4)	114.4 (2)	114.2 (2)	114.6 (3)
C(2)—C(3)—C(4)	114.2 (4)	114 (1)	113.8 (4)	114.7 (2)	115.1 (2)	113.8 (3)
N(3)—C(4)—C(3)	105.4 (3)	105.3 (9)	106.6 (3)	104.7 (2)	104.1 (2)	105.6 (2)
N(3)—C(4)—C(5)	104.2 (3)	105.3 (9)	103.6 (3)	105.2 (1)	105.7 (1)	105.3 (2)
C(3)—C(4)—C(5)	111.2 (3)	109 (1)	110.3 (3)	112.0 (1)	111.1 (2)	111.0 (2)
C(4)—C(5)—C(6)	109.7 (3)	109.8 (9)	109.2 (3)	109.4 (2)	108.9 (1)	108.2 (2)
C(4)—C(5)—C(15)	123.7 (4)	117 (1)	115.1 (3)	121.5 (2)	122.9 (2)	117.2 (2)
C(4)—C(5)—C(17)	115.8 (3)	117 (1)	115.1 (3)	117.3 (2)	116.7 (2)	115.8 (3)
C(6)—C(5)—C(15)	88.0 (3)	87.9 (9)	88.2 (3)	89.2 (1)	88.2 (1)	89.4 (2)
C(6)—C(5)—C(17)	104.2 (4)	104.8 (9)	105.2 (3)	103.1 (1)	103.2 (1)	104.4 (2)
C(15)—C(5)—C(17)	110.0 (4)	116 (1)	119.3 (3)	110.8 (2)	111.0 (2)	116.7 (3)
C(1)—C(6)—C(5)	108.7 (3)	108.3 (9)	108.9 (3)	109.0 (1)	109.3 (1)	109.3 (2)
C(1)—C(6)—C(16)	123.7 (4)	116 (1)	116.0 (3)	121.9 (1)	123.1 (2)	117.6 (3)
C(1)—C(6)—C(18)	115.9 (4)	117 (1)	114.9 (3)	116.3 (2)	115.9 (2)	115.1 (3)
C(5)—C(6)—C(16)	88.5 (3)	87.9 (9)	88.5 (3)	88.1 (1)	88.3 (1)	88.7 (2)
C(5)—C(6)—C(18)	104.7 (4)	101.6 (9)	105.1 (3)	104.3 (1)	103.2 (2)	104.2 (2)
C(16)—C(6)—C(18)	110.1 (3)	118 (1)	118.7 (3)	111.6 (2)	111.3 (2)	117.0 (3)
O(1)—C(7)—N(1)	126.3 (3)	124 (1)	127.0 (4)	126.3 (2)	126.5 (2)	127.2 (3)
O(1)—C(7)—N(2)	127.4 (3)	129 (1)	127.1 (4)	125.8 (2)	127.2 (2)	126.3 (3)
N(1)—C(7)—N(2)	106.3 (3)	106.6 (9)	105.9 (3)	107.8 (2)	106.4 (1)	106.5 (3)
O(2)—C(8)—N(2)	127.7 (3)	129 (1)	127.3 (4)	126.6 (2)	126.8 (2)	127.5 (3)
O(2)—C(8)—N(3)	125.6 (3)	126 (1)	126.8 (4)	126.1 (2)	126.2 (2)	126.7 (3)
N(2)—C(8)—N(3)	106.6 (3)	104.8 (9)	105.7 (4)	107.2 (2)	106.9 (1)	105.7 (3)
N(2)—C(9)—C(10)	118.7 (3)	118 (1)	119.4 (4)	119.6 (2)	119.2 (2)	120.0 (3)
N(2)—C(9)—C(14)	119.2 (4)	120 (1)	119.4 (4)	119.7 (2)	119.5 (2)	119.1 (3)
C(10)—C(9)—C(14)	122.0 (4)	122 (1)	121.2 (4)	120.6 (2)	121.3 (2)	120.9 (3)
C(9)—C(10)—C(11)	118.3 (5)	118 (1)	117.7 (4)	119.5 (2)	119.1 (2)	118.9 (4)
C(10)—C(11)—C(12)	120.8 (6)	122 (1)	121.0 (5)	120.6 (3)	120.1 (2)	121.2 (4)
C(11)—C(12)—C(13)	120.3 (5)	119 (1)	119.7 (4)	119.0 (3)	119.9 (3)	119.0 (4)
C(12)—C(13)—C(14)	120.7 (6)	120 (1)	121.7 (5)	121.6 (3)	120.9 (3)	120.8 (4)
C(9)—C(14)—C(13)	117.9 (5)	119 (1)	118.7 (5)	118.7 (3)	118.7 (2)	119.1 (3)
O(3)—C(15)—C(5)	112.1 (4)	109 (1)	105.5 (3)	109.2 (2)	108.5 (2)	116.0 (2)
O(3)—C(15)—C(16)	58.9 (3)	58.7 (9)	60.1 (2)	58.7 (1)	59.2 (1)	110.8 (3)
C(5)—C(15)—C(16)	91.5 (4)	92 (1)	91.9 (3)	90.9 (2)	91.5 (2)	88.2 (2)
O(3)—C(16)—C(6)	110.7 (13)	108 (1)	104.3 (3)	109.3 (2)	110.1 (1)	
O(3)—C(16)—C(15)	58.5 (3)	56.9 (9)	59.0 (2)	59.3 (1)	59.1 (1)	
C(6)—C(16)—C(15)	92.0 (3)	92 (1)	91.4 (3)	91.8 (1)	92.0 (2)	93.3 (2)
O(4)—C(17)—C(5)	106.7 (3)	108 (1)	105.9 (3)	105.9 (2)	105.5 (2)	104.5 (2)
O(4)—C(18)—C(6)	105.7 (4)	111 (1)	106.3 (3)	104.6 (2)	105.8 (2)	105.3 (2)
O(3)—C(15)—C(20)			118.1 (3)	116.4 (2)	118.5 (2)	109.1 (3)
C(5)—C(15)—C(20)			128.0 (4)	127.2 (2)	126.4 (2)	116.3 (3)
C(16)—C(15)—C(20)			133.3 (4)	135.1 (2)	133.6 (2)	115.2 (3)
O(3)—C(16)—C(19)			117.7 (3)	117.1 (2)	117.1 (3)	
C(6)—C(16)—C(19)			129.1 (4)	127.7 (2)	126.7 (3)	133.6 (3)
C(15)—C(16)—C(19)			133.9 (4)	131.8 (3)	132.8 (3)	133.0 (3)

Table 9. Comparison of angles (°) between planes in (1)–(5)

The notation of the plane is shown in the scheme (II).

	P1—P2	P2—P3	P2—P4	P3—P4	P4—P5	P4—P7	P5—P6	P5—P7
(1)	136.8	119.8	114.7	125.5	125.2	124.5	114.1	110.2
(2)	137.1	121.5	115.7	122.8	117.4	125.1	111.0	117.6
(3)	139.3	121.3	114.8	123.8	116.8	123.8	106.3	119.6
(4A)	128.9	118.2	115.8	126.0	123.3	125.2	111.8	111.5
(4B)	132.4	119.6	115.3	125.2	124.4	124.2	111.5	111.3
(5)	140.8	120.9	115.7	123.3	118.6	123.6	—	117.4

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