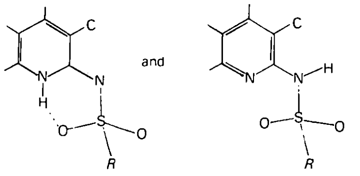
the methoxy groups to be coplanar with the phenyl ring has resulted in some close contacts between C(17) and C(3), and C(18) and C(6) and the H atoms attached to them, which have been relieved by increase of the angles C(3)—C(4)—O(5) and C(6)—C(5)—O(6) from 120 to 124·2 (5) and 123·8 (5)° respectively.

The tetrahedral coordination around the S atom is distorted, with the O(1)—S(1)—O(2) angle being 116·6 (2)°, larger than the normal tetrahedral value, possibly due to repulsions between the sulphonamide O atoms (Kálmán, Duffin & Kucsman, 1971). The S(1)—O(1) and S(1)—O(2) bond lengths of 1·430 (3) and 1·456 (4) Å indicate π-bond character of over 60% (Cruickshank, 1961), while the S(1)—C(1) and S(1)—N(1) bond lengths of 1·777 (5) and 1·589 (4) Å are very close to values obtained for similar structures (Hamodrakas *et al.*, 1978).

Some structural differences between this structure and others of the same family (Hamodrakas *et al.*, 1978; Eliopoulos *et al.*, 1983) occur, in addition to the position of the methyl group attached to the phenyl ring. The length of the bond N(1)—C(7) [1·334 (6) Å] suggests a possible partial double bond. The final electron density map shows no peaks close to N(1) while a peak with height one third of an electron appears 1·07 (3) Å from the pyridine N(2). This peak, 2·06 (3) Å from O(2), suggests a hydrogen bond [N(2)—H···O(2)]. This unusual situation could be explained by tautomerism of the two forms with the balance shifted in favour of the first form.



In the 4-methyl member of this series of compounds (Eliopoulos *et al.*, 1983), where an intermolecular hydrogen-bond scheme N(1)···H—N(2) was proposed, the hydrogen peak was observed 1·08 Å from N(2).

We wish to thank Dr Ch. Camoutsis for providing the material, Mr D. Akrigg for assisting us with the calculations and diagrams and the Leeds University Computing Service for the provision of computing facilities.

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Structure of 11,12-Dichloro-3,8-bis(2,4,6-trimethylphenyl)-1,4,7,10-tetraoxa-2,9-diazadispiro[4.0.4.2]dodeca-2,8,11-triene (DTDD), $C_{24}H_{22}Cl_2N_2O_4$

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Abstract. $M_r = 473{\cdot}36$, monoclinic $P2_1/c$, $a = 16{\cdot}661\ (3)$, $b = 9{\cdot}011\ (1)$, $c = 24{\cdot}231\ (7)$ Å, $\beta = 139{\cdot}01\ (1)°$, $Z = 4$, $V = 2386{\cdot}2$ Å^3, $D_x = 1{\cdot}318$, $D_m = 1{\cdot}307$ Mg m^{-3}, m.p. = 448–449 K, λ(Mo $K\alpha$) = 0·71069 Å, $\mu = 0{\cdot}305$ mm^{-1}, $F(000) = 984$, $T = 293$ K. Final $R = 0{\cdot}056$ for 815 independent non-zero

reflections. The molecule is established to be a cyclobutenobis(spirodioxazole). Its characteristic feature is the system formed by the nearly tetragonal cyclobutene ring and the two dioxazole rings perpendicular to it. The molecule appears to have a mirror plane almost perpendicular to a pair of opposite edges of the tetragonal ring.

Introduction. In the course of study of the 1,3-dipolar cycloaddition reaction of several substituted benzonitrile oxides with substituted cyclobutenediones a series of new compounds was prepared in the Laboratory of Organic Chemistry of this University (Argyropoulos, 1978). As chemical and spectroscopic data regarding the selectivity of the cycloaddition of mesitonitrile oxide to the cyclobutenedione ring were in some cases inconclusive, an X-ray structure analysis was considered necessary and to this end the crystal structure of the title compound (DTDD hereafter) was carried out.

Experimental. Pure colourless crystals, 0·34 × 0·22 × 0·12 mm. D_m measured by flotation in KBr solution. Computer-controlled Philips PW1100 four-circle single-crystal diffractometer. Cell constants by least-squares analysis of θ angles of 66 strong reflections. Graphite-monochromated Mo $K\alpha$, ω-scan mode. Three standard reflections, no intensity variation. 7629 measured reflections, $\theta = 3{\cdot}5$–$36°$, 4569 unique reflections, $R_{\rm int} = 0{\cdot}119$, 815 with $I > 2\sigma(I)$. Index range h 0→17, k 0→10, l −14→14. No absorption correction. Direct methods with *MULTAN*77 (Main, Lessinger, Woolfson, Germain & Declercq, 1977), all non-H atoms located on E map. Full-matrix least squares (F magnitudes) with XRAY76 (Stewart, Machin, Dickinson, Ammon, Heck & Flack, 1976); positional and anisotropic thermal parameters refined. Atomic scattering factors and anomalous-dispersion correction for Cl from *International Tables for X-ray Crystallography* (1974). H atoms of phenyl rings placed at calculated positions and those of methyl groups located on a difference Fourier map; positional parameters refined [except for H1(C12), H2(C23), H3(C23)], isotropic temperature factors as those of bonded C atoms. $w = 1/[A + B\sigma^2(F)]$ with $A = 2{\cdot}0$, $B = 0{\cdot}8$; $R = 0{\cdot}056$, $R_w = 0{\cdot}050$, $S = 1{\cdot}55$. $(\Delta/\sigma)_{\max} = 1{\cdot}26$, $(\Delta/\sigma)_{\rm mean} = 0{\cdot}26$. Final $\Delta\rho$ excursions $-0{\cdot}28$–$0{\cdot}26$ e Å^{-3}.

Discussion. The final positional parameters and equivalent isotropic temperature coefficients are given in Table 1.[*] Interatomic distances and angles are given in Table 2. The structural and conformational details of the molecule are shown in Fig. 1. The compound is clearly a cyclobutenobis(spirodioxazole). The characteristic feature of the molecule is the system formed by the nearly tetragonal cyclobutene ring B and the two dioxazole rings $D(1)$ and $D(2)$ perpendicular to it [$\angle BD(1) = 88{\cdot}2(0{\cdot}2)$, $\angle BD(2) = 88{\cdot}8(1{\cdot}3)°$]. The plane of the tetragonal ring almost bisects the two five-membered dioxazole rings. The two benzene rings are oriented in such a way that they form angles of 71·3(0·1) and 66·9(0·2)° with the corresponding dioxazole rings to which they are linked. The molecule appears to have a mirror plane almost perpendicular to the edges C(1)—C(4) and C(2)—C(3) of the tetragonal ring. All rings are planar to a good approximation (see deposition footnote).

The geometries of the cyclobutene ring and the dioxazole rings are normal, except for the C(1)—C(4) bond distance, 1·62 (4) Å, which is considerably larger than a normal C—C bond (~1·54 Å). This may be attributed to stereochemical repulsion between the two dioxazole rings, which is caused by the fact that the intramolecular O—O distances tend to their normally accepted value, 2·84 Å (Bondi, 1964) [O(1)···O(4) = 2·80 (2), O(2)···O(3) = 2·83 (2) Å].

A slightly larger C—C bond distance, 1·643 Å, has recently been reported in tetramethyl 3,7-diacetoxy-1,5-dimethyltricyclo[3.3.0.0^{2,8}]octa-3,6-diene-2,4,6,8-tetracarboxylate (Gompper & Schwarzensteiner, 1982).

Table 1. *Atomic coordinates and equivalent isotropic temperature factors* (Å^2) *for the non*-H *atoms*

Here and throughout this paper the e.s.d.'s are given in parentheses and refer to the last digit.

	x	y	z	$B_{\rm eq}$*
Cl(1)	−0·2308 (4)	0·2444 (5)	−0·1299 (3)	5·8
Cl(2)	0·0610 (4)	0·3513 (5)	−0·0442 (3)	7·9
O(1)	−0·0582 (8)	0·2865 (10)	0·0801 (6)	5·3
O(2)	−0·0330 (9)	0·0490 (11)	0·0649 (7)	6·0
O(3)	0·1888 (10)	0·1305 (11)	0·1276 (7)	4·4
O(4)	0·1628 (8)	0·3686 (11)	0·1450 (6)	4·9
N(1)	−0·0923 (10)	0·0473 (12)	0·0888 (8)	5·3
N(2)	0·3029 (11)	0·1902 (11)	0·2046 (8)	4·3
C(1)	−0·0270 (14)	0·1963 (19)	0·0498 (14)	4·7
C(2)	−0·0826 (15)	0·2453 (19)	−0·0332 (11)	3·9
C(3)	0·0233 (19)	0·2808 (18)	−0·0011 (15)	5·6
C(4)	0·1012 (15)	0·2434 (21)	0·0882 (12)	4·3
C(5)	−0·1006 (13)	0·1859 (15)	0·0963 (10)	4·7
C(6)	−0·1574 (19)	0·2405 (18)	0·1172 (15)	4·0
C(7)	−0·0948 (17)	0·2368 (19)	0·2013 (17)	5·8
C(8)	−0·1456 (18)	0·2982 (17)	0·2200 (10)	4·2
C(9)	−0·2618 (24)	0·3663 (21)	0·1598 (21)	7·6
C(10)	−0·3200 (23)	0·3721 (24)	0·0802 (21)	7·3
C(11)	−0·2749 (21)	0·3160 (22)	0·0547 (16)	6·0
C(12)	0·0307 (15)	0·1659 (20)	0·2720 (10)	6·5
C(13)	−0·3153 (15)	0·4394 (19)	0·1830 (11)	7·3
C(14)	−0·3431 (16)	0·3176 (17)	−0·0338 (13)	6·5
C(15)	0·2834 (13)	0·3217 (16)	0·2109 (10)	5·3
C(16)	0·3776 (14)	0·4308 (16)	0·2811 (10)	4·4
C(17)	0·4668 (13)	0·4780 (16)	0·2906 (10)	2·8
C(18)	0·5571 (13)	0·5698 (16)	0·3593 (9)	3·8
C(19)	0·5600 (16)	0·6188 (17)	0·4140 (10)	3·9
C(20)	0·4666 (21)	0·5650 (21)	0·4007 (14)	6·2
C(21)	0·3725 (16)	0·4733 (19)	0·3336 (12)	6·8
C(22)	0·4708 (19)	0·4305 (19)	0·2332 (14)	7·3
C(23)	0·6566 (18)	0·7232 (26)	0·4862 (13)	8·1
C(24)	0·2781 (19)	0·4193 (24)	0·3240 (13)	8·0

* $B_{\rm eq} = \frac{8}{3}\pi^2$ trace $\tilde{\mathbf{U}}$.

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[*] Lists of structure factors, anisotropic thermal parameters of the non-H atoms, H-atom parameters, C—H bond distances, intermolecular distances and least-squares-planes' calculations have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 38787 (14 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

The two Cl atoms lie on the plane of the cyclobutene ring at distances C(2)–Cl(1) = 1·69 (1) and C(3)–Cl(2) = 1·70 (4) Å. The Cl–Cl distance is normal [3·69 (1) Å].

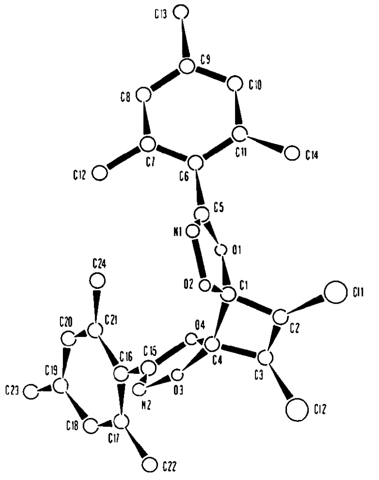

Fig. 1. Clinographic projection of the DTDD molecule.

Table 2. *Interatomic distances* (Å) *and angles* (°)

Cl(1)–C(2)	1·69 (1)	C(5)–C(6)	1·46 (5)
Cl(2)–C(3)	1·70 (4)	C(6)–C(7)	1·43 (5)
C(1)–C(2)	1·51 (4)	C(7)–C(8)	1·34 (5)
C(2)–C(3)	1·32 (4)	C(8)–C(9)	1·41 (3)
C(3)–C(4)	1·50 (4)	C(9)–C(10)	1·36 (7)
C(4)–C(1)	1·62 (4)	C(10)–C(11)	1·38 (7)
O(1)–C(1)	1·43 (4)	C(11)–C(6)	1·45 (3)
O(1)–C(5)	1·38 (3)	C(7)–C(12)	1·52 (2)
O(2)–C(1)	1·40 (2)	C(9)–C(13)	1·52 (6)
O(2)–N(1)	1·47 (3)	C(11)–C(14)	1·49 (5)
N(1)–C(5)	1·28 (2)	C(15)–C(16)	1·51 (2)
O(3)–C(4)	1·40 (2)	C(16)–C(17)	1·39 (4)
O(3)–N(2)	1·42 (1)	C(17)–C(18)	1·39 (2)
O(4)–C(4)	1·45 (2)	C(18)–C(19)	1·36 (4)
O(4)–C(15)	1·39 (2)	C(19)–C(20)	1·42 (5)
N(2)–C(15)	1·27 (2)	C(20)–C(21)	1·39 (2)
		C(21)–C(16)	1·39 (5)
		C(17)–C(22)	1·51 (5)
		C(19)–C(23)	1·51 (2)
		C(21)–C(24)	1·49 (5)

C(2)–C(1)–C(4)	84·5 (2·2)	C(5)–C(6)–C(7)	121·5 (1·9)
C(2)–C(1)–O(2)	122·1 (1·8)	C(5)–C(6)–C(11)	120·8 (3·0)
C(4)–C(1)–O(1)	112·9 (1·5)	C(7)–C(6)–C(11)	117·3 (3·6)
C(1)–C(2)–C(3)	94·7 (1·8)	C(6)–C(7)–C(8)	120·3 (1·9)
C(1)–C(2)–Cl(1)	129·0 (2·1)	C(7)–C(8)–C(9)	123·5 (3·0)
C(3)–C(2)–Cl(1)	136·2 (2·5)	C(8)–C(9)–C(10)	115·9 (4·5)
C(2)–C(3)–C(4)	96·5 (3·0)	C(9)–C(10)–C(11)	125·2 (2·6)
C(2)–C(3)–Cl(2)	132·7 (1·9)	C(10)–C(11)–C(6)	117·7 (3·1)
C(4)–C(3)–Cl(2)	130·8 (2·0)	C(6)–C(7)–C(12)	121·4 (3·2)
C(3)–C(4)–C(1)	84·2 (1·8)	C(8)–C(7)–C(12)	118·3 (2·8)
C(3)–C(4)–O(3)	118·0 (2·7)	C(8)–C(9)–C(13)	122·6 (3·4)
C(1)–C(4)–O(4)	115·7 (2·4)	C(10)–C(9)–C(13)	121·4 (2·4)
O(1)–C(1)–O(2)	106·3 (2·6)	C(6)–C(11)–C(14)	118·1 (3·6)
C(1)–O(1)–C(5)	103·6 (1·5)	C(10)–C(11)–C(14)	124·1 (2·1)
O(1)–C(5)–N(1)	117·8 (2·4)	C(15)–C(16)–C(17)	117·2 (2·4)
C(5)–N(1)–O(2)	102·8 (1·8)	C(15)–C(16)–C(21)	118·6 (2·2)
N(1)–O(2)–C(1)	108·2 (1·8)	C(17)–C(16)–C(21)	124·1 (1·5)
O(1)–C(5)–C(6)	119·0 (1·4)	C(16)–C(17)–C(18)	115·9 (2·4)
N(1)–C(5)–C(6)	123·1 (2·2)	C(17)–C(18)–C(19)	124·6 (2·4)
O(3)–C(4)–O(4)	106·9 (1·0)	C(18)–C(19)–C(20)	116·2 (1·7)
C(4)–O(3)–N(2)	107·0 (1·1)	C(19)–C(20)–C(21)	123·1 (3·4)
O(3)–N(2)–C(15)	107·5 (1·1)	C(20)–C(21)–C(16)	116·0 (3·0)
N(2)–C(15)–O(4)	115·3 (1·1)	C(16)–C(17)–C(22)	123·3 (1·5)
C(15)–O(4)–C(4)	103·2 (1·2)	C(18)–C(17)–C(22)	120·7 (2·3)
O(4)–C(15)–C(16)	117·9 (1·4)	C(18)–C(19)–C(23)	123·6 (2·7)
N(2)–C(15)–C(16)	126·8 (1·3)	C(20)–C(19)–C(23)	120·2 (2·8)
		C(20)–C(21)–C(24)	120·3 (3·2)
		C(16)–C(21)–C(24)	123·6 (1·7)

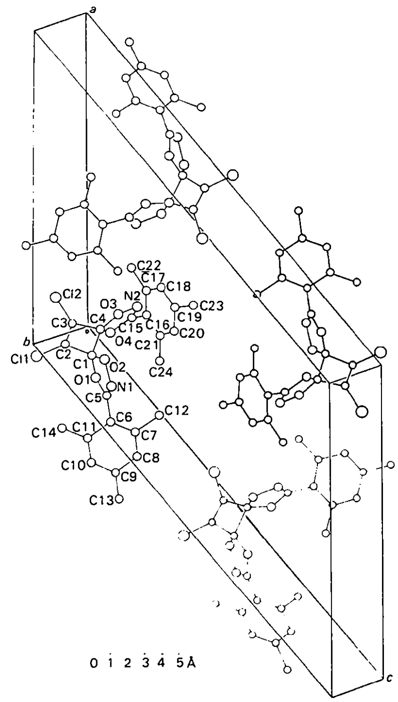

Fig. 2. Clinographic projection of DTDD showing the molecular packing in the unit cell.

The geometrical features of the benzene rings are in good agreement with the usually accepted values. Mean bond lengths are 1·39 Å in both rings.

A clinographic projection of the structure is shown in Fig. 2. A list of selected intermolecular distances has been deposited.*

We thank Professor N. E. Alexandrou and Dr N. Argyropoulos, Laboratory of Organic Chemistry, Aristotle University of Thessaloniki, for providing the DTDD crystals and the University of Thessaloniki for the use of its computer facilities.

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* See deposition footnote.

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