The Crystal and Molecular Structure of 4'-Methylene-1,2-di-*m*-bromophenyl-1',2',6',7'-tetra-phenylspiro{pyrazolidine-4,8'-[8'H,4'H]-benzo[1,2-c: 4,5-c']dipyrazoline}-3,5,3',5'-tetraone, $C_{47}H_{30}N_6O_4Br_2$

BY L. FANFANI, A. NUNZI, P. F. ZANAZZI AND A. R. ZANZARI Istituto di Mineralogia, Università di Perugia, 06100 Perugia, Italy

(Received 16 May 1973; accepted 19 September 1973)

The crystal and molecular structure of the title compound has been determined by single-crystal X-ray diffraction analysis. Crystals are triclinic, space group $P\overline{1}$, with a=12.775, b=12.702, c=13.158 Å, $\alpha=111.33^{\circ}$, $\beta=97.42^{\circ}$, $\gamma=88.71^{\circ}$. The structure was solved by Patterson and Fourier methods, and refined by least-squares methods to a final R value of 0.067 for 2230 observed reflexions.

Introduction

Pyrazolidine and its derivatives are interesting, from a pharmaceutical point of view, as antipyretic agents. In an attempt to obtain new active compounds, recent studies on the chemical behaviour of 1,2-diphenylpyrazolidine-3,5-dione showed that the action of acetic anhydride on this compound (reaction A) gives a new complex substance which crystallizes in yellowish tiny irregular tablets with high melting point $(323-325^{\circ}C)$ (Mangiavacchi, Lazzi & Ridi, 1966). The same compound was also obtained by reaction of 1,2-diphenylpyrazolidine-3,5-dione with 1-(1,2-diphenyl-3,5-diketo-4-pyrazolidine)-1-(1,2-diphenyl-3,5-diketo-4-pyrazolidiniliden)ethane (reaction B). Further chemical studies to characterize the reaction product (Mangiavacchi, 1968) have assigned it the elementary formula $C_{47}H_{32}N_6O_4$; the molecular structure proposed on the basis of infrared and n.m.r. spectra as well as chemical evidence, was:



Since some doubts about the molecular structure still remained, the X-ray analysis of a brominated derivative of the compound was undertaken. The structural study showed that the proposed formula is not correct, the right one being:



Experimental

Crystals of the title compound, $C_{47}H_{30}N_6O_4Br_2$, were prepared and kindly supplied by Dr Mangiavacchi of the Institute of Pharmaceutical Chemistry of Siena University. They are very small pale yellowish needles with rhombic section, melting point 302–304 °C.

Preliminary rotation, Weissenberg and precession photographs showed that they are triclinic and gave rough unit-cell constants which were subsequently refined by the least-squares method from fourteen accurate 2θ values measured on a diffractometer. The refined values are: $a=12.775\pm0.003$, $b=12.702\pm$ 0.001, $c=13.158\pm0.002$ Å, $\alpha=111.33\pm0.01^{\circ}$, $\beta=$ $97.42\pm0.02^{\circ}$, $\gamma=88.71\pm0.01^{\circ}$.* Cell volume is 1971.7 A³.

The space group $P\overline{1}$ was assumed at the beginning of the present investigation on the basis of morphological evidence. The number of molecules in the unit cell was also assumed to be 2, according to the empirical criterion proposed by Kempster & Lipson (1972). Both assumptions were later confirmed by structural analysis. The calculated density is 1.500 g cm⁻³.

The needle crystals were elongated along $[\overline{1}01]$. The reflexions were collected and indexed according to a

^{*} This cell is not a Delaunay reduced cell. The Delaunay reduction gives a cell with parameters: a=12.775, b=12.702, c=14.591 Å; $\alpha=122.86^\circ$, $\beta=95.86^\circ$, $\gamma=91.29^\circ$. The transformation matrix from our cell to Delaunay cell is $[100/0\overline{10}/011]$.

B-centered cell having a' = [101]; b' = b; $c' = [\overline{1}01]$. All subsequent calculations were performed with reference to the above primitive cell.

The crystal used for data collection, with dimensions $0.36 \times 0.08 \times 0.08$ mm, was mounted with its longest dimension coincident with the axis of a Hilger and Watts four-circle automatic diffractometer. Intensity data were collected by the ω -2 θ scan technique with zirconium-filtered Mo radiation. Scans of 60 s with steps of 0.01° and a count of 1 s for each step were taken across the peaks; background was counted for 20 s on each side of the peak. As a check on electronic and crystal stability during data collection three standard reflexions were monitored every 100 measurements. A total of 4750 intensities were measured within the sphere $2\theta \le 44^{\circ}$. Beyond this limit only a few re-

Table 1. Fractional atomic coordinates and thermal parameters, with standard deviations

$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		x/a	y/b	z/c	B (Å ²)
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	Br (1)	0.3951(1)	0.5953(1)	1.1159 (1)	6.86*
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	Br(2)	-0·0108 (1)	0.1800(1)	1·1069 (1)	7.01*
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	O(Ì)	0.3643(5)	0.0854 (6)	0.2887(6)	3.78(17)
$ \begin{array}{lllllllllllllllllllllllll$	O(2)	0.4492 (6)	-0.2523(6)	0.5160 (6)	3.82 (17)
	O(3)	0.0873 (6)	0.0127 (6)	0.5915(6)	3.96 (18)
	O(4)	0.3796 (6)	0.2560 (6)	0.7658 (5)	3.42(17)
	N(1)	0.2768 (7)	0.2152(7)	0.4197 (7)	3.07 (20)
	N(2)	0.2507 (6)	0.2285(7)	0.5258 (6)	2.77(20)
	N(3)	0.3103 (6)	-0.0544 (7)	0.7077 (7)	2.98 (20)
N(5) 0 - 1195 (6) 0 - 1737 (7) 0 - 7425 (7) 2 - 69 (19) N(6) 0 - 2121 (6) 0 - 2404 (7) 0 - 8025 (6) 2 - 65 (19) C(1) 0 - 2597 (8) 0 - 1042 (8) 0 - 6311 (8) 2 - 48 (23) C(2) 0 - 3162 (8) - 0 - 00697 (8) 0 - 5466 (8) 1 - 96 (22) C(4) 0 - 3730 (8) - 0 - 0 - 0478 (8) 0 - 4475 (8) 2 - 38 (23) C(5) 0 - 3257 (8) 0 - 0586 (8) 0 - 4533 (8) 2 - 33 (23) C(6) 0 - 2783 (8) 0 - 1126 (8) 0 - 5387 (8) 2 - 43 (23) C(7) 0 - 3269 (8) 0 - 1141 (9) 0 - 3774 (9) 3 - 46 (26) C(8) 0 - 0478 (8) 0 - 04718 (9) 3 - 32 (26) C(9) 0 - 1453 (9) 0 - 02072 (9) 0 - 7421 (9) 2 - 96 (25) C(10) 0 - 2962 (9) 0 - 02072 (9) 0 - 7421 (9) 2 - 96 (25) C(11) 0 - 4178 (8) - 0 - 1190 (9) 0 - 3633 (8) 3 - 25 (25) C(12) 0 - 2844 (8) 0 - 3116 (9) 0 - 3917 (9) 2 - 96 (25) C(13) 0 - 2362 (9) 0 - 04017 (12) 0 - 2619 (11) 6 - 14 (25) C(14) 0 - 2435 (9) 0 - 04017 (12) 0 - 2619 (11) 6 - 14 (25) C(14) 0 - 2435 (9) 0 - 04017 (12) 0 - 2619 (11) 6 - 14 (25) C(14) 0 - 2435 (9) 0 - 04017 (12) 0 - 2619 (11) 6 - 14 (25) C(15) 0 - 2988 (11) 0 - 9305 (11) 0 - 3359 (11) 5 - 52 (33) C(16) 0 - 3444 (10) 0 - 5005 (10) 0 - 44354 (9) - 3-88 (28) C(18) 0 - 0419 (11) 0 - 5293 (12) 0 - 4974 (11) 6 - 51 (32) C(17) 0 - 3394 (9) 0 - 04063 (10) 0 - 4656 (9) 4 - 24 (29) C(21) 0 - 04174 (12) 0 - 3709 (13) 0 - 5590 (12) 7 - 02 (38) C(22) 0 - 10409 (9) 0 - 3378 (10) 0 - 6061 (9) 4 - 44 (29) C(23) 0 - 1460 (8) 0 - 2760 (9) 0 - 8143 (8) 3 - 01 (25) C(24) 0 - 2009 (8) 0 - 3574 (9) 0 - 8762 (8) 2 - 64 (24) C(25) 0 - 1059 (9) 0 - 04105 (10) 0 - 8647 (9) 3 - 542 (26) C(23) 0 - 0463 (10) 0 - 5723 (10) 1 - 0118 (10) 4 - 73 (30) C(23) 0 - 0257 (9) 0 - 1598 (9) 0 - 9573 (9) 3 - 92 (28) C(33) - 0 -1295 (9) 0 - 1598 (9) 0 - 9573 (9) 3 - 92 (28) C(33) - 0 -1295 (9) 0 - 1598 (9) 0 - 9573 (9) 3 - 592 (23) C(34) 0 - 01393 (8) 0 - 0101 (8) 0 - 8637 (9) 3 - 556 (26) C(34) 0 - 02720 (9) - 0	N(4)	0.3709 (6)	-0·1557 (7)	0.6706 (7)	3.00 (20)
$ N(6) \qquad 0 - 2121 \ (6) \qquad 0 - 2404 \ (7) \qquad 0 - 8026 \ (6) \qquad 2 - 65 \ (19) \\ C(1) \qquad 0 - 2597 \ (8) \qquad 0 - 1042 \ (8) \qquad 0 - 6381 \ (8) \qquad 2 - 16 \ (23) \\ C(2) \qquad 0 - 3162 \ (8) \qquad - 0 - 00497 \ (8) \qquad 0 - 6381 \ (8) \qquad 2 - 48 \ (23) \\ C(3) \qquad 0 - 3665 \ (7) \qquad - 0 - 0697 \ (8) \qquad 0 - 5466 \ (8) \qquad 1 - 96 \ (22) \\ C(4) \qquad 0 - 3730 \ (8) \qquad - 0 - 0478 \ (8) \qquad 0 - 4475 \ (8) \qquad 2 - 38 \ (23) \\ C(5) \qquad 0 - 3257 \ (8) \qquad 0 - 0478 \ (8) \qquad 0 - 4475 \ (8) \qquad 2 - 38 \ (23) \\ C(5) \qquad 0 - 2783 \ (8) \qquad 0 - 1262 \ (8) \qquad 0 - 5387 \ (8) \qquad 2 - 45 \ (23) \\ C(7) \qquad 0 - 3269 \ (8) \qquad 0 - 1141 \ (9) \qquad 0 - 3774 \ (9) \qquad 3 - 346 \ (26) \\ C(8) \qquad 0 - 4011 \ (9) \qquad - 0 - 1685 \ (9) \qquad 0 - 5718 \ (9) \qquad 3 - 32 \ (26) \\ C(8) \qquad 0 - 4011 \ (9) \qquad - 0 - 1685 \ (9) \qquad 0 - 5718 \ (9) \qquad 3 - 32 \ (26) \\ C(10) \qquad 0 - 2962 \ (9) \qquad 0 - 2072 \ (9) \qquad 0 - 7421 \ (9) \qquad 2 - 95 \ (25) \\ C(11) \qquad 0 - 4178 \ (8) \qquad - 0 - 1190 \ (9) \qquad 0 - 3633 \ (8) \qquad 3 - 25 \ (25) \\ C(13) \qquad 0 - 2362 \ (9) \qquad 0 - 3070 \ (10) \qquad 0 - 2917 \ (10) \qquad 4 - 51 \ (29) \\ C(14) \qquad 0 - 2435 \ (9) \qquad 0 - 4017 \ (12) \qquad 0 - 2619 \ (11) \qquad 6 - 14 \ (35) \\ C(17) \qquad 0 - 3344 \ (10) \qquad 0 -4005 \ (10) \qquad 0 -4338 \ (10) \qquad 5 - 52 \ (33) \\ C(16) \qquad 0 - 3444 \ (10) \qquad 0 -5005 \ (10) \qquad 0 -4338 \ (10) \qquad 5 - 11 \ (32) \ C(13) \qquad 0 - 2057 \ (10) \qquad 0 - 2990 \ (10) \qquad 0 -4388 \ (9) \qquad 4 - 24 \ (29) \ C(19) \qquad - 0 -0419 \ (11) \qquad 0 -2593 \ (12) \qquad 0 -4974 \ (11) \ 6 - 31 \ (35) \ C(22) \qquad 0 - 0 4974 \ (12) \qquad 0 -3790 \ (13) \qquad 0 -5590 \ (12) \ - 7.02 \ (38) \ C(21) \qquad 0 -0 386 \ (12) \qquad 0 -378 \ (10) \qquad 0 -6647 \ (9) \qquad 3 - 38 \ (28) \ C(21) \qquad 0 -0 386 \ (12) \qquad 0 -378 \ (10) \qquad 0 -6647 \ (9) \qquad 3 - 36 \ (24) \ C(23) \ 0 -1002 \ (10) \qquad 0 -5220 \ (11) \qquad 0 -9386 \ (10) \ 5 -02 \ (31) \ C(23) \ 0 -1002 \ (10) \qquad 0 -5220 \ (11) \qquad 0 -9386 \ (10) \ 5 -02 \ (31) \ C(23) \ 0 -1038 \ (10) \ 0 -1018 \ (10) \ 4 -373 \ (30) \ C(23) \ 0 -1038 \ (10) \ 0 -1018 \ (10) \ 4 -373 \ (30) \ C(23) \ 0 -1038 \ (10) \ 0 -1038 \ (10) \ 5 -13 \ (23) \ - 0 -0386 \ (10) \ 5 -02 \ (31) \ C(23) \ 0 -0386 \ (10) \ 5 -12 \ (23) \ 0 -1038 \ (10) \ 0 -108 \ (10) \ 5 -12 \ (23) \ 0 -103$	N(5)	0.1195 (6)	0.1737 (7)	0.7425 (7)	2·69 (19)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	N(6)	0.2121 (6)	0.2404 (7)	0.8026 (6)	2.65 (19)
$\begin{array}{cccccc} C(2) & 0.3162 (8) & -0.0040 (8) & 0.6311 (8) & 2.48 (23) \\ C(3) & 0.3665 (7) & -0.0697 (8) & 0.5466 (8) & 1.96 (22) \\ C(4) & 0.3730 (8) & -0.0478 (8) & 0.4475 (8) & 2.38 (23) \\ C(5) & 0.3257 (8) & 0.0586 (8) & 0.4533 (8) & 2.43 (23) \\ C(6) & 0.783 (8) & 0.1141 (9) & 0.3774 (9) & 3.46 (26) \\ C(8) & 0.4011 (9) & -0.1685 (9) & 0.5718 (9) & 3.46 (26) \\ C(8) & 0.4011 (9) & -0.1685 (9) & 0.5718 (9) & 3.46 (26) \\ C(9) & 0.1453 (9) & 0.0894 (9) & 0.6503 (9) & 3.04 (25) \\ C(10) & 0.2962 (9) & 0.2072 (9) & 0.7421 (9) & 2.95 (25) \\ C(11) & 0.4178 (8) & -0.1190 (9) & 0.3333 (8) & 3.25 (25) \\ C(11) & 0.4135 (9) & 0.4017 (12) & 0.2619 (11) & 6.14 (35) \\ C(14) & 0.2362 (9) & 0.3070 (10) & 0.2917 (10) & 4.51 (29) \\ C(15) & 0.2988 (11) & 0.4935 (11) & 0.3359 (11) & 5.52 (33) \\ C(16) & 0.3444 (10) & 0.5005 (10) & 0.4338 (10) & 5.11 (32) \\ C(17) & 0.3949 (9) & 0.4063 (10) & 0.4654 (9) & 3.88 (28) \\ C(18) & 0.0575 (10) & 0.2090 (10) & 0.4654 (9) & 3.88 (28) \\ C(18) & 0.0575 (10) & 0.2090 (10) & 0.4654 (9) & 3.88 (28) \\ C(22) & -0.04174 (12) & 0.3709 (13) & 0.5590 (12) & 7.02 (38) \\ C(21) & -0.0419 (11) & 0.2523 (12) & 0.4974 (11) & 6.13 (35) \\ C(22) & -1.0409 (9) & 0.3578 (10) & 0.6061 (9) & 4.44 (29) \\ C(23) & 0.1460 (8) & 0.2760 (9) & 0.5413 (8) & 3.01 (25) \\ C(24) & 0.2009 (8) & 0.3554 (9) & 0.8667 (8) & 2.64 (24) \\ C(25) & 0.1059 (9) & 0.4105 (10) & 0.8667 (9) & 3.54 (26) \\ C(26) & 0.1002 (10) & 0.5723 (10) & 1.0118 (10) & 4.73 (30) \\ C(27) & 0.1832 (10) & 0.5723 (10) & 1.0118 (10) & 4.73 (30) \\ C(23) & 0.1460 (8) & 0.2760 (9) & 0.9541 (8) & 3.02 (23) \\ C(24) & 0.2009 (8) & 0.3554 (9) & 0.8657 (9) & 3.55 (27) \\ C(25) & 0.1032 (10) & 0.5198 (9) & 0.9573 (9) & 3.92 (28) \\ C(23) & 0.163 (9) & 0.1598 (10) & 0.0923 (8) & 2.78 (24) \\ C(33) & -0.1453 (9) & 0.114 (10) & 0.9738 (10) & 4.04 (29) \\ C(33) & -0.041 (9) & 0.1285 (10) & 1.00437 (10) & 4.48 (30) \\ C(44) & 0.2702 (10) & -0.2660 (10) & 0.717 (9) & 3.66 (25) \\ C(33) & -0.041 (9) & 0.2566 (10) & 0.717 (9) & 3.55 (27) \\ C(33) & 0.4599 (10) & 0.1285 (10) & 0.7718 (10) &$	C (1)	0.2597 (8)	0.1042 (8)	0.6381 (8)	2.16 (23)
$\begin{array}{cccccc} C(3) & 0.3665 (7) & -0.0697 (8) & 0.5466 (8) & 1.96 (22) \\ C(4) & 0.3730 (8) & -0.0478 (8) & 0.4475 (8) & 2.38 (23) \\ C(5) & 0.3257 (8) & 0.0586 (8) & 0.5387 (8) & 2.33 (23) \\ C(6) & 0.2783 (8) & 0.1262 (8) & 0.5387 (8) & 2.45 (23) \\ C(7) & 0.3269 (8) & 0.1141 (9) & 0.3774 (9) & 3.46 (26) \\ C(8) & 0.4011 (9) & -0.1685 (9) & 0.5718 (9) & 3.32 (26) \\ C(9) & 0.1453 (9) & 0.0894 (9) & 0.6503 (9) & 3.04 (25) \\ C(10) & 0.2962 (9) & 0.2072 (9) & 0.7421 (9) & 2.95 (25) \\ C(11) & 0.4178 (8) & -0.1190 (9) & 0.3633 (8) & 3.25 (25) \\ C(12) & 0.2844 (8) & 0.3116 (9) & 0.3917 (9) & 2.96 (25) \\ C(13) & 0.2362 (9) & 0.3070 (10) & 0.2917 (10) & 4.51 (29) \\ C(14) & 0.2435 (9) & 0.4017 (12) & 0.2619 (11) & 6.514 (35) \\ C(15) & 0.2988 (11) & 0.4935 (11) & 0.3359 (11) & 5.52 (33) \\ C(15) & 0.2988 (11) & 0.4935 (11) & 0.3359 (11) & 5.52 (33) \\ C(16) & 0.3444 (10) & 0.5005 (10) & 0.4868 (9) & 4.24 (29) \\ C(18) & 0.0575 (10) & 0.2090 (10) & 0.4868 (9) & 4.24 (29) \\ C(19) & -0.0419 (11) & 0.2593 (12) & 0.4974 (11) & 6.31 (35) \\ C(20) & -0.0474 (12) & 0.3709 (13) & 0.5590 (12) & 7.02 (38) \\ C(23) & 0.1460 (8) & 0.2760 (9) & 0.5413 (8) & 3.01 (25) \\ C(24) & 0.209 (8) & 0.3378 (10) & 0.68647 (9) & 3.54 (26) \\ C(25) & 0.1059 (9) & 0.4105 (10) & 0.8647 (9) & 3.54 (26) \\ C(26) & 0.1002 (10) & 0.5220 (11) & 0.9386 (10) & 4.02 (31) \\ C(23) & 0.2865 (8) & 0.4053 (9) & 0.9524 (8) & 2.06 (23) \\ C(33) & 0.0351 (9) & 0.5193 (9) & 0.9524 (8) & 3.02 (23) \\ C(34) & -0.1453 (9) & 0.1518 (9) & 0.9733 (9) & 3.80 (27) \\ C(33) & -0.0318 (9) & 0.1598 (9) & 0.9733 (9) & 3.82 (28) \\ C(33) & -0.0451 (9) & 0.1598 (9) & 0.9733 (9) & 3.82 (28) \\ C(33) & -0.0451 (9) & 0.1598 (9) & 0.9736 (9) & 3.80 (27) \\ C(34) & -0.0318 (9) & 0.1598 (9) & 0.9736 (9) & 3.80 (27) \\ C(35) & -0.0641 (9) & 0.1309 (9) & 0.7386 (10) & 4.23 (28) \\ C(33) & -0.1453 (9) & 0.1198 (9) & 0.7386 (10) & 4.23 (28) \\ C(33) & -0.0451 (9) & -0.2400 (9) & 0.7386 (9) & 3.80 (27) \\ C(34) & -0.270 (9) & -0.2566 (10) & 0.7517 (9) & 4.63 (28) \\ C(44) & 0.2702 (10) & -0.2400 (9) & 0.7162 (8) & $	C(2)	0.3162 (8)	-0·0040 (8)	0.6311 (8)	2.48 (23)
$\begin{array}{cccccc} C(4) & 0.3730 \ (8) & -0.0478 \ (8) & 0.4475 \ (8) & 2.38 \ (23) \\ C(5) & 0.3257 \ (8) & 0.0586 \ (8) & 0.4533 \ (8) & 2.45 \ (23) \\ C(7) & 0.3269 \ (8) & 0.1141 \ (9) & 0.3774 \ (9) & 3.46 \ (26) \\ C(8) & 0.0411 \ (9) & -0.1685 \ (9) & 0.5718 \ (9) & 3.32 \ (26) \\ C(9) & 0.1453 \ (9) & -0.0894 \ (9) & 0.6503 \ (9) & 3.04 \ (25) \\ C(10) & 0.2962 \ (9) & 0.2072 \ (9) & 0.7421 \ (9) & 2.95 \ (25) \\ C(11) & 0.478 \ (8) & -0.1190 \ (9) & 0.3633 \ (8) & 3.25 \ (25) \\ C(12) & 0.2844 \ (8) & -0.1190 \ (9) & 0.3633 \ (8) & 3.25 \ (25) \\ C(13) & 0.2362 \ (9) & 0.4017 \ (12) & 0.2619 \ (11) \ 6.14 \ (35) \\ C(14) & 0.2435 \ (9) & 0.4017 \ (12) & 0.2619 \ (11) \ 6.14 \ (35) \\ C(15) & 0.2988 \ (11) & 0.4935 \ (11) & 0.3359 \ (11) \ 5.52 \ (33) \\ C(16) & 0.3444 \ (10) & 0.5005 \ (10) & 0.4338 \ (10) \ 5.11 \ (32) \\ C(17) & 0.3944 \ (9) & 0.4063 \ (10) & 0.4654 \ (9) \ 3.48 \ (28) \\ C(18) & 0.0575 \ (10) & 0.2090 \ (10) & 0.4868 \ (9) \ 4.24 \ (29) \\ C(19) & -0.0419 \ (11) & 0.2593 \ (12) & 0.4974 \ (11) \ 6.13 \ (35) \\ C(22) & 0.1440 \ (9) & 0.3878 \ (10) & 0.6616 \ (9) \ 4.44 \ (29) \\ C(23) & 0.1460 \ (8) & 0.3754 \ (9) \ 0.8762 \ (8) \ 2.64 \ (24) \\ C(25) & 0.1059 \ (9) & 0.4105 \ (10) \ 0.8762 \ (8) \ 2.64 \ (24) \\ C(25) & 0.1059 \ (9) & 0.4353 \ (10) \ 0.6616 \ (9) \ 4.44 \ (29) \\ C(24) & 0.2009 \ (8) \ 0.3554 \ (9) \ 0.8762 \ (8) \ 2.64 \ (24) \\ C(25) & 0.1059 \ (9) \ 0.4105 \ (10) \ 0.8762 \ (8) \ 2.64 \ (24) \\ C(25) & 0.1059 \ (9) \ 0.4105 \ (10) \ 0.8867 \ (9) \ 3.62 \ (25) \\ C(30) & 0.0351 \ (9) \ 0.1598 \ (9) \ 0.9573 \ (9) \ 3.92 \ (25) \\ C(30) & 0.0351 \ (9) \ 0.1598 \ (9) \ 0.9573 \ (9) \ 3.92 \ (25) \\ C(30) & 0.0351 \ (9) \ 0.1598 \ (9) \ 0.9573 \ (9) \ 3.92 \ (25) \\ C(30) & 0.0351 \ (9) \ 0.1598 \ (9) \ 0.9573 \ (9) \ 3.92 \ (25) \\ C(30) & 0.0351 \ (9) \ 0.1598 \ (9) \ 0.9573 \ (9) \ 3.92 \ (25) \\ C(30) & 0.0351 \ (9) \ 0.1598 \ (9) \ 0.9573 \ (9) \ 3.92 \ (25) \\ C(30) & 0.0351 \ (9) \ 0.1598 \ (9) \ 0.9573 \ (9) \ 3.92 \ (25) \\ C(30) & 0.0351 \ (9) \ 0.1598 \ (9) \ 0.9573 \ (9) \ 3.96 \ (25) \ C(3$	C(3)	0.3665 (7)	-0.0697 (8)	0.5466 (8)	1.96 (22)
$\begin{array}{cccccc} C(5) & 0 \cdot 3257 \ (8) & 0 \cdot 0586 \ (8) & 0 \cdot 4533 \ (8) & 2 \cdot 33 \ (23) \\ C(6) & 0 \cdot 2783 \ (8) & 0 \cdot 1141 \ (9) & 0 \cdot 3774 \ (9) & 3 \cdot 46 \ (26) \\ C(8) & 0 \cdot 4011 \ (9) & -0 \cdot 1685 \ (9) & 0 \cdot 5718 \ (9) & 3 \cdot 32 \ (26) \\ C(9) & 0 \cdot 1433 \ (9) & 0 \cdot 0894 \ (9) & 0 \cdot 6503 \ (9) & 3 \cdot 04 \ (25) \\ C(10) & 0 \cdot 2962 \ (9) & 0 \cdot 2072 \ (9) & 0 \cdot 7421 \ (9) & 2 \cdot 95 \ (25) \\ C(11) & 0 \cdot 4178 \ (8) & -0 \cdot 1190 \ (9) & 0 \cdot 3633 \ (8) & 3 \cdot 25 \ (25) \\ C(13) & 0 \cdot 2362 \ (9) & 0 \cdot 3070 \ (10) & 0 \cdot 2917 \ (10) & 4 \cdot 51 \ (29) \\ C(14) & 0 \cdot 2435 \ (9) & 0 \cdot 4017 \ (12) & 0 \cdot 2619 \ (11) & 6 \cdot 14 \ (35) \\ C(16) & 0 \cdot 3444 \ (10) & 0 \cdot 5005 \ (10) & 0 \cdot 4338 \ (10) & 5 \cdot 11 \ (32) \\ C(16) & 0 \cdot 3444 \ (10) & 0 \cdot 5005 \ (10) & 0 \cdot 4338 \ (10) & 5 \cdot 11 \ (32) \\ C(17) & 0 \cdot 3394 \ (9) & 0 \cdot 4063 \ (10) & 0 \cdot 4654 \ (9) & 3 \cdot 88 \ (28) \\ C(18) & 0 \cdot 0575 \ (10) & 0 \cdot 2399 \ (12) & 0 \cdot 4974 \ (11) & 6 \cdot 31 \ (35) \\ C(20) & -0 \cdot 0474 \ (12) & 0 \cdot 3709 \ (13) & 0 \cdot 5590 \ (12) & 7 \cdot 02 \ (38) \\ C(21) & -0 \cdot 0386 \ (12) & 0 \cdot 4353 \ (10) & 0 \cdot 6061 \ (9) & 4 \cdot 44 \ (29) \\ C(22) & 0 \cdot 1409 \ (9) & 0 \cdot 3378 \ (10) & 0 \cdot 6061 \ (9) & 4 \cdot 44 \ (29) \\ C(23) & 0 \cdot 1460 \ (8) & 0 \cdot 2760 \ (9) & 0 \cdot 5413 \ (8) & 3 \cdot 01 \ (25) \\ C(24) & 0 \cdot 2009 \ (8) & 0 \cdot 3554 \ (9) & 0 \cdot 8762 \ (8) & 2 \cdot 64 \ (24) \\ C(25) & 0 \cdot 1059 \ (9) & 0 \cdot 4105 \ (10) & 0 \cdot 8647 \ (9) & 3 \cdot 54 \ (26) \\ C(26) & 0 \cdot 1002 \ (10) & 0 \cdot 5220 \ (11) & 0 \cdot 9386 \ (10) & 4 \cdot 23 \ (28) \\ C(23) & 0 \cdot 0351 \ (9) & 0 \cdot 1513 \ (9) & 0 \cdot 9378 \ (10) & 4 \cdot 33 \ (25) \\ C(33) & -0 \cdot 0475 \ (9) & 0 \cdot 1513 \ (9) & 0 \cdot 9737 \ (9) & 3 \cdot 54 \ (26) \\ C(25) & 0 \cdot 1059 \ (9) & 0 \cdot 1114 \ (10) & 0 \cdot 7920 \ (10) & 4 \cdot 48 \ (30) \\ C(23) & 0 \cdot 0351 \ (9) & 0 \cdot 1028 \ (9) & 0 \cdot 9737 \ (9) & 3 \cdot 54 \ (24) \\ C(33) & -0 \cdot 0143 \ (9) & 0 \cdot 1513 \ (9) & 0 \cdot 1018 \ (10) & 4 \cdot 44 \ (29) \\ C(33) & -0 \cdot 01450 \ (9) & 0 \cdot 1513 \ (9) & 0 \cdot 1018 \ (10) & 4 \cdot 44 \ (29) \\ C(33) & -0 \cdot 01450 \ (9) & -0 \cdot 2150 \ (9) & 0 \cdot 8763 \ (9) & 3 \cdot 82 \ (28) \\ C(33) & -0 \cdot 0159 \ (9) & 0 \cdot 1519 \ (9) & 0 \cdot 1$	C(4)	0.3730 (8)	-0.0478 (8)	0.4475 (8)	2.38 (23)
$\begin{array}{cccccc} C(5) & 0 \cdot 2/83 \ (8) & 0 \cdot 1262 \ (8) & 0 \cdot 5387 \ (8) & 2.45 \ (23) \\ C(7) & 0 \cdot 3269 \ (8) & 0 \cdot 1141 \ (9) & 0 \cdot 3774 \ (9) & 3 \cdot 46 \ (26) \\ C(8) & 0 \cdot 4011 \ (9) & -0 \cdot 1685 \ (9) & 0 \cdot 5718 \ (9) & 3 \cdot 32 \ (26) \\ C(9) & 0 \cdot 1453 \ (9) & 0 \cdot 0894 \ (9) & 0 \cdot 6503 \ (9) & 3 \cdot 04 \ (25) \\ C(10) & 0 \cdot 2962 \ (9) & 0 \cdot 2072 \ (9) & 0 \cdot 7421 \ (9) & 2.95 \ (25) \\ C(11) & 0 \cdot 4178 \ (8) & -0 \cdot 1190 \ (9) & 0 \cdot 3633 \ (8) & 3 \cdot 25 \ (25) \\ C(12) & 0 \cdot 2844 \ (8) & 0 \cdot 3116 \ (9) & 0 \cdot 3917 \ (9) & 2 \cdot 96 \ (25) \\ C(13) & 0 \cdot 2362 \ (9) & 0 \cdot 4017 \ (12) & 0 \cdot 2619 \ (11) & 6 \cdot 14 \ (35) \\ C(15) & 0 \cdot 2988 \ (11) & 0 \cdot 4935 \ (11) & 0 \cdot 3359 \ (11) & 5 \cdot 52 \ (33) \\ C(16) & 0 \cdot 3444 \ (10) & 0 \cdot 5005 \ (10) & 0 \cdot 4338 \ (10) \ 5 \cdot 11 \ (32) \\ C(16) & 0 \cdot 3444 \ (10) & 0 \cdot 5005 \ (10) & 0 \cdot 4338 \ (10) \ 5 \cdot 11 \ (32) \\ C(17) & 0 \cdot 3394 \ (9) & 0 \cdot 4063 \ (10) & 0 \cdot 4654 \ (9) & 3 \cdot 88 \ (28) \\ C(18) & 0 \cdot 0575 \ (10) & 0 \cdot 2090 \ (10) & 0 \cdot 4868 \ (9) & 4 \cdot 24 \ (29) \\ C(20) & -0 \cdot 0474 \ (12) & 0 \cdot 3709 \ (13) & 0 \cdot 5590 \ (12) & 7 \cdot 02 \ (38) \\ C(21) & -0 \cdot 0366 \ (12) & 0 \cdot 4353 \ (11) & 0 \cdot 6127 \ (11) & 6 \cdot 13 \ (35) \\ C(22) & 0 \cdot 1409 \ (9) & 0 \cdot 33574 \ (9) & 0 \cdot 8762 \ (8) & 2 \cdot 64 \ (24) \\ C(25) & 0 \cdot 1059 \ (9) & 0 \cdot 4105 \ (10) & 0 \cdot 8647 \ (9) & 3 \cdot 54 \ (26) \\ C(26) & 0 \cdot 1002 \ (10) & 0 \cdot 5220 \ (11) & 0 \cdot 9386 \ (10) & 5 \cdot 02 \ (31) \\ C(22) & 0 \cdot 2865 \ (8) & 0 \cdot 4053 \ (9) & 0 \cdot 98762 \ (8) & 2 \cdot 64 \ (24) \\ C(25) & 0 \cdot 1059 \ (9) & 0 \cdot 5193 \ (9) & 0 \cdot 1018 \ (10) & 4 \cdot 73 \ (30) \\ C(26) & 0 \cdot 1002 \ (10) & 0 \cdot 5220 \ (11) & 0 \cdot 9886 \ (10) & 5 \cdot 02 \ (31) \\ C(23) & -0 \cdot 0318 \ (9) & 0 \cdot 1193 \ (9) & 0 \cdot 9083 \ (8) & 2 \cdot 78 \ (24) \\ C(23) & -0 \cdot 0318 \ (9) & 0 \cdot 1598 \ (9) & 0 \cdot 9886 \ (10) & 4 \cdot 24 \ (29) \\ C(33) & -0 \cdot 1453 \ (9) & 0 \cdot 1141 \ (10) & 0 \cdot 7920 \ (10) & 4 \cdot 49 \ (30) \\ C(23) & -0 \cdot 0318 \ (9) & 0 \cdot 1598 \ (9) & 0 \cdot 9886 \ (10) & 4 \cdot 42 \ (29) \\ C(33) & -0 \cdot 0451 \ (9) & 0 \cdot 05181 \ (9) & 0 \cdot 9886 \ (10) & 4 \cdot 44 \ (29) \\ C(33) & -0 \cdot 125 \ (9) & 0 \cdot 1114 \ (10) & 0 \cdot $	C(5)	0.3257(8)	0.0586(8)	0.4533(8)	2.33 (23)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(6)	0.2/83(8)	0.1262(8)	0.5387(8)	2.45 (23)
$\begin{array}{cccccc} C(8) & 0.4011 (9) & -0.1685 (9) & 0.5718 (9) & 3.32 (2b) \\ C(9) & 0.1435 (9) & 0.0894 (9) & 0.6503 (9) & 3.04 (25) \\ C(11) & 0.2962 (9) & 0.2072 (9) & 0.7421 (9) & 2.95 (25) \\ C(11) & 0.4178 (8) & -0.1190 (9) & 0.3633 (8) & 3.25 (25) \\ C(13) & 0.2362 (9) & 0.3070 (10) & 0.2917 (10) & 4.51 (29) \\ C(14) & 0.2435 (9) & 0.4017 (12) & 0.2619 (11) & 6.14 (35) \\ C(15) & 0.2988 (11) & 0.4935 (11) & 0.3359 (11) & 5.52 (33) \\ C(16) & 0.3444 (10) & 0.5005 (10) & 0.4338 (10) & 5.11 (32) \\ C(17) & 0.3394 (9) & 0.4063 (10) & 0.4654 (9) & 3.88 (28) \\ C(18) & 0.0575 (10) & 0.2090 (10) & 0.4868 (9) & 4.24 (29) \\ C(19) & -0.0419 (11) & 0.2593 (12) & 0.4974 (11) & 6.31 (35) \\ C(22) & -0.0474 (12) & 0.3709 (13) & 0.5590 (12) & 7.02 (38) \\ C(21) & 0.0386 (12) & 0.4353 (11) & 0.6127 (11) & 6.13 (35) \\ C(22) & 0.1460 (8) & 0.2760 (9) & 0.5413 (8) & 3.01 (25) \\ C(24) & 0.2009 (8) & 0.3554 (9) & 0.8762 (8) & 2.64 (24) \\ C(25) & 0.1059 (9) & 0.4105 (10) & 0.8647 (9) & 3.54 (26) \\ C(26) & 0.1002 (10) & 0.5220 (11) & 0.9386 (10) & 5.02 (31) \\ C(27) & 0.1832 (10) & 0.5193 (9) & 1.0192 (9) & 3.82 (28) \\ C(23) & 0.0351 (9) & 0.5193 (9) & 1.0192 (9) & 3.82 (28) \\ C(23) & 0.0351 (9) & 0.1606 (8) & 0.8001 (9) & 3.06 (25) \\ C(30) & 0.0311 (9) & 0.1606 (8) & 0.8001 (9) & 3.06 (25) \\ C(31) & 0.0311 (9) & 0.1606 (8) & 0.8001 (9) & 3.06 (25) \\ C(33) & -0.1295 (9) & 0.1250 (9) & 0.9386 (10) & 4.23 (28) \\ C(33) & -0.1295 (9) & 0.1250 (9) & 0.9386 (10) & 4.23 (28) \\ C(33) & -0.1295 (9) & 0.1250 (9) & 0.8773 (9) & 3.92 (28) \\ C(33) & -0.1295 (9) & 0.1250 (9) & 0.8773 (9) & 3.92 (28) \\ C(33) & -0.1295 (9) & 0.1250 (9) & 0.8776 (9) & 3.80 (27) \\ C(36) & 0.3393 (8) & 0.0101 (8) & 0.8244 (8) & 2.54 (24) \\ C(37) & 0.4381 (9) & 0.1309 (9) & 0.7386 (9) & 3.80 (27) \\ C(36) & 0.3393 (8) & 0.0101 (8) & 0.8244 (8) & 2.54 (24) \\ C(37) & 0.4382 (10) & 0.1250 (9) & 0.7386 (9) & 3.80 (27) \\ C(36) & 0.3393 (8) & 0.0101 (8) & 0.8244 (8) & 2.54 (24) \\ C(37) & 0.4313 (10) & 0.7517 (9) & 3.66 (25) \\ C(44) & 0.2702 (10) & -0.2385 (10) & 1.0447 (10) & 4.84 (30) \\$	C(7)	0.3269(8)	0.1(9)	0.37/4(9)	3.46 (26)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(8)	0.4011(9)	-0.1685(9)	0.5/18(9)	3.32 (26)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(9)	0.1453(9)	0.0894(9)	0.6503(9)	3.04(25)
$\begin{array}{cccccc} C(12) & 0.741/8 (6) & -0.1190 (9) & 0.3033 (6) & 3.23 (25) \\ C(13) & 0.2362 (9) & 0.3070 (10) & 0.2917 (10) & 4.51 (29) \\ C(14) & 0.2435 (9) & 0.4017 (12) & 0.2619 (11) & 6.14 (35) \\ C(15) & 0.2988 (11) & 0.4935 (11) & 0.3359 (11) & 5.52 (33) \\ C(16) & 0.3444 (10) & 0.5005 (10) & 0.4338 (10) & 5.11 (32) \\ C(17) & 0.3394 (9) & 0.4063 (10) & 0.4654 (9) & 3.88 (28) \\ C(18) & 0.0575 (10) & 0.2993 (12) & 0.4974 (11) & 6.31 (35) \\ C(20) & -0.0474 (12) & 0.3709 (13) & 0.5590 (12) & 7.02 (38) \\ C(21) & 0.0386 (12) & 0.4353 (11) & 0.6127 (11) & 6.13 (35) \\ C(22) & 0.1409 (9) & 0.3878 (10) & 0.601 (9) & 4.44 (29) \\ C(23) & 0.1460 (8) & 0.2760 (9) & 0.5413 (8) & 3.01 (25) \\ C(24) & 0.209 (8) & 0.3554 (9) & 0.8762 (8) & 2.64 (24) \\ C(25) & 0.1059 (9) & 0.4105 (10) & 0.88647 (9) & 3.54 (26) \\ C(26) & 0.1002 (10) & 0.5520 (11) & 0.9386 (10) & 5.02 (31) \\ C(26) & 0.1002 (10) & 0.5723 (10) & 1.0118 (10) & 4.73 (30) \\ C(28) & 0.2757 (9) & 0.5193 (9) & 1.0192 (9) & 3.82 (28) \\ C(23) & 0.0351 (9) & 0.1606 (8) & 0.8001 (9) & 3.06 (25) \\ C(33) & -0.0318 (9) & 0.1598 (9) & 0.9573 (9) & 3.92 (28) \\ C(33) & -0.0318 (9) & 0.1598 (9) & 0.9573 (9) & 3.92 (28) \\ C(33) & -0.0318 (9) & 0.1598 (9) & 0.9573 (9) & 3.92 (28) \\ C(33) & -0.0318 (9) & 0.1309 (9) & 0.7386 (10) & 4.23 (28) \\ C(33) & -0.0318 (9) & 0.1309 (9) & 0.7386 (10) & 4.23 (28) \\ C(33) & -0.441 (9) & 0.1309 (9) & 0.7386 (10) & 4.23 (28) \\ C(33) & -0.441 (9) & 0.1309 (9) & 0.7386 (10) & 4.23 (28) \\ C(33) & -0.451 (9) & 0.1250 (9) & 0.8925 (9) & 3.80 (27) \\ C(33) & -0.331 (9) & 0.1114 (10) & 0.7920 (10) & 4.49 (30) \\ C(35) & -0.351 (9) & 0.1250 (9) & 0.8925 (9) & 3.55 (27) \\ C(33) & 0.3224 (10) & 0.1284 (10) & 0.7517 (9) & 4.03 (28) \\ C(44) & 0.2607 (9) & -0.2400 (9) & 0.7162 (8) & 2.90 (25) \\ C(43) & 0.3560 (11) & -0.4044 (10) & 0.8014 (10) & 5.15 (32) \\ C(44) & 0.2702 (10) & -0.3413 (11) & 0.7956 (10) & 5.19 (32) \\ C(44) & 0.2702 (10) & -0.3413 (11) & 0.7637 (9) & 4.54 (30) \\ C(47) & 0.4510 (9) & -0.3036 (10) & 0.7217 (9) & 3.66 (27) \\ \end{array}$	C(10)	0.2902(9)	0.2072(9)	0.7421(9)	2.95 (25)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(11)	0.3844(8)	-0.1190(9)	0.3033(0)	3.25 (25)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(12)	0.2362(0)	0.3070(10)	0.3917(3)	2.90 (23)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(13)	0.2302(9) 0.2435(9)	0.3070(10) 0.4017(12)	0.2917(10) 0.2619(11)	$4^{\circ}31(29)$
$\begin{array}{cccccc} C(16) & 0.3444 & (10) & 0.5005 & (11) & 0.4338 & (10) & 5.11 & (32) \\ C(17) & 0.3394 & (9) & 0.4063 & (10) & 0.44388 & (10) & 5.11 & (32) \\ C(18) & 0.0575 & (10) & 0.2090 & (10) & 0.44868 & (9) & 4.24 & (29) \\ C(19) & -0.0419 & (11) & 0.2593 & (12) & 0.4974 & (11) & 6.31 & (35) \\ C(20) & -0.0474 & (12) & 0.3709 & (13) & 0.5590 & (12) & 7.02 & (38) \\ C(21) & 0.0386 & (12) & 0.4353 & (11) & 0.6127 & (11) & 6.13 & (35) \\ C(22) & 0.1409 & (9) & 0.3878 & (10) & 0.6061 & (9) & 4.44 & (29) \\ C(23) & 0.1460 & (8) & 0.2760 & (9) & 0.5413 & (8) & 3.01 & (25) \\ C(24) & 0.2009 & (8) & 0.3554 & (9) & 0.8762 & (8) & 2.64 & (24) \\ C(25) & 0.1059 & (9) & 0.4105 & (10) & 0.8647 & (9) & 3.54 & (26) \\ C(26) & 0.1002 & (10) & 0.5220 & (11) & 0.9386 & (10) & 5.02 & (31) \\ C(27) & 0.1832 & (10) & 0.5723 & (10) & 1.0118 & (10) & 4.73 & (30) \\ C(28) & 0.2757 & (9) & 0.5193 & (9) & 1.0192 & (9) & 3.82 & (28) \\ C(29) & 0.2865 & (8) & 0.4053 & (9) & 0.95224 & (8) & 3.02 & (25) \\ C(30) & 0.0351 & (9) & 0.1606 & (8) & 0.9083 & (8) & 2.78 & (24) \\ C(32) & -0.0318 & (9) & 0.1598 & (9) & 0.9573 & (9) & 3.92 & (28) \\ C(33) & -0.1295 & (9) & 0.1250 & (9) & 0.8886 & (10) & 4.23 & (28) \\ C(34) & -0.1453 & (9) & 0.1114 & (10) & 0.7920 & (10) & 4.49 & (30) \\ C(35) & -0.0641 & (9) & 0.1309 & (9) & 0.7386 & (9) & 3.80 & (27) \\ C(36) & 0.3393 & 0.0101 & (8) & 0.8244 & (8) & 2.54 & (24) \\ C(37) & 0.4361 & (9) & 0.0581 & (9) & 0.8757 & (9) & 3.55 & (27) \\ C(38) & 0.4599 & (10) & 0.1244 & (10) & 0.9778 & (10) & 5.22 & (32) \\ C(39) & 0.3824 & (10) & 0.1285 & (10) & 1.0447 & (10) & 4.84 & (30) \\ C(40) & 0.2854 & (10) & 0.7937 & (10) & 1.0043 & (10) & 4.44 & (29) \\ C(41) & 0.2607 & (9) & -0.2566 & (10) & 0.7517 & (9) & 3.56 & (26) \\ C(44) & 0.2702 & (10) & -0.3897 & (10) & 0.7637 & (9) & 4.54 & (30) \\ C(44) & 0.2702 & (10) & -0.3897 & (10) & 0.7637 & (9) & 4.54 & (30) \\ C(44) & 0.475 & (10) & -0.3897 & (10) & 0.7637 & (9) & 4.54 & (30) \\ C(47) & 0.4510 & (9) & -0.2306 & (10) & 0.7217 & (9) & 3.566 & (27) \\ C(46) & 0.4475 & (10) & -0.3897 & (10) $	C(15)	0.2988(11)	0.4935(11)	0.2019(11) 0.3359(11)	5.52(33)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(16)	0.3444(10)	0.5005(11)	0.4338(10)	5.11(32)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$\mathbf{C}(17)$	0.3394(9)	0.4063(10)	0.4654(9)	3.88(28)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$\mathbf{C}(18)$	0.0575(10)	0.2090(10)	0.4868(9)	4.24(29)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$\tilde{C}(19)$	-0.0419(11)	0.2593(12)	0.4974(11)	6.31(35)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$\tilde{C}(20)$	-0.0474(12)	0.3709(13)	0.5590(12)	7.02(38)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(21)	0.0386 (12)	0.4353(11)	0.6127(11)	6.13(35)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(22)	0·1409 (9)	0.3878 (10)	0.6061(9)	4.44(29)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(23)	0.1460 (8)	0.2760 (9)	0.5413 (8)	3.01(25)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(24)	0.2009 (8)	0.3554 (9)	0.8762 (8)	2.64(24)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(25)	0.1059 (9)	0.4105 (10)	0.8647 (9)	3.54 (26)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(26)	0.1002 (10)	0.5220 (11)	0.9386 (10)	5.02 (31)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(27)	0.1832 (10)	0.5723 (10)	1.0118 (10)	4.73 (30)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(28)	0.2757 (9)	0.5193 (9)	1.0192 (9)	3.82 (28)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(29)	0.2865 (8)	0.4053 (9)	0.9524 (8)	3.02 (25)
$\begin{array}{ccccc} C(31) & 0.0522 \ (8) & 0.1796 \ (8) & 0.9083 \ (8) & 2.78 \ (24) \\ C(32) & -0.0318 \ (9) & 0.1598 \ (9) & 0.9573 \ (9) & 3.92 \ (28) \\ C(33) & -0.1295 \ (9) & 0.1250 \ (9) & 0.8986 \ (10) & 4.23 \ (28) \\ C(34) & -0.1453 \ (9) & 0.1114 \ (10) & 0.7920 \ (10) & 4.49 \ (30) \\ C(35) & -0.0641 \ (9) & 0.1309 \ (9) & 0.7386 \ (9) & 3.80 \ (27) \\ C(36) & 0.3393 \ (8) & 0.0101 \ (8) & 0.8244 \ (8) & 2.54 \ (24) \\ C(37) & 0.4361 \ (9) & 0.0581 \ (9) & 0.8657 \ (9) & 3.55 \ (27) \\ C(38) & 0.4399 \ (10) & 0.1214 \ (10) & 0.9778 \ (10) & 5.22 \ (32) \\ C(39) & 0.3824 \ (10) & 0.1285 \ (10) & 1.0447 \ (10) & 4.84 \ (30) \\ C(40) & 0.2854 \ (10) & 0.0793 \ (10) & 1.0043 \ (10) & 4.44 \ (29) \\ C(41) & 0.2607 \ (9) & -0.2400 \ (9) & 0.7162 \ (8) & 2.90 \ (25) \\ C(43) & 0.2720 \ (9) & -0.2566 \ (10) & 0.7517 \ (9) & 4.03 \ (28) \\ C(44) & 0.2702 \ (10) & -0.3413 \ (11) & 0.7956 \ (10) & 5.19 \ (32) \\ C(45) & 0.3560 \ (11) & -0.4044 \ (10) & 0.8014 \ (10) & 5.15 \ (32) \\ C(46) & 0.4475 \ (10) & -0.3897 \ (10) & 0.7637 \ (9) & 4.54 \ (30) \\ C(47) & 0.4510 \ (9) & -0.3036 \ (10) & 0.7217 \ (9) & 3.66 \ (27) \\ \end{array}$	C(30)	0.0351 (9)	0.1606 (8)	0.8001 (9)	3.06 (25)
$\begin{array}{cccccc} C(32) & & -0 \cdot 0318 \ (9) & & 0 \cdot 1598 \ (9) & & 0 \cdot 9573 \ (9) & & 3 \cdot 92 \ (28) \\ C(33) & & -0 \cdot 1295 \ (9) & & 0 \cdot 1250 \ (9) & & 0 \cdot 8986 \ (10) & & 4 \cdot 23 \ (28) \\ C(34) & & -0 \cdot 1453 \ (9) & & 0 \cdot 1114 \ (10) & & 0 \cdot 7920 \ (10) & & 4 \cdot 49 \ (30) \\ C(35) & & -0 \cdot 0641 \ (9) & & 0 \cdot 1309 \ (9) & & 0 \cdot 7386 \ (9) & & 3 \cdot 80 \ (27) \\ C(36) & & 0 \cdot 3393 \ (8) & & 0 \cdot 0101 \ (8) & & 0 \cdot 8244 \ (8) & & 2 \cdot 54 \ (24) \\ C(37) & & 0 \cdot 4361 \ (9) & & 0 \cdot 0581 \ (9) & & 0 \cdot 8657 \ (9) & & 3 \cdot 55 \ (27) \\ C(38) & & 0 \cdot 4599 \ (10) & & 0 \cdot 1214 \ (10) & & 0 \cdot 9778 \ (10) & & 5 \cdot 22 \ (32) \\ C(39) & & 0 \cdot 3824 \ (10) & & 0 \cdot 1285 \ (10) & & 1 \cdot 0447 \ (10) & & 4 \cdot 84 \ (30) \\ C(40) & & 0 \cdot 2854 \ (10) & & 0 \cdot 0793 \ (10) & & 1 \cdot 0043 \ (10) & & 4 \cdot 44 \ (29) \\ C(41) & & 0 \cdot 2607 \ (9) & & -0 \cdot 2400 \ (9) & & 0 \cdot 7162 \ (8) & & 2 \cdot 90 \ (25) \\ C(43) & & 0 \cdot 2720 \ (9) & & -0 \cdot 2566 \ (10) & & 0 \cdot 7517 \ (9) & & 4 \cdot 03 \ (28) \\ C(44) & & 0 \cdot 2702 \ (10) & & -0 \cdot 3413 \ (11) & & 0 \cdot 7956 \ (10) & & 5 \cdot 19 \ (32) \\ C(45) & & 0 \cdot 3560 \ (11) & & -0 \cdot 4044 \ (10) & & 0 \cdot 8014 \ (10) & & 5 \cdot 15 \ (32) \\ C(46) & & 0 \cdot 4475 \ (10) & & -0 \cdot 3897 \ (10) & & 0 \cdot 7637 \ (9) & & 4 \cdot 54 \ (30) \\ C(47) & & 0 \cdot 4510 \ (9) & & -0 \cdot 3036 \ (10) & & 0 \cdot 7217 \ (9) & & 3 \cdot 66 \ (27) \\ \end{array}$	C(31)	0.0522 (8)	0.1796 (8)	0.9083 (8)	2.78 (24)
$\begin{array}{cccccc} C(33) & -0.1295 \ (9) & 0.1250 \ (9) & 0.8986 \ (10) & 4.23 \ (28) \\ C(34) & -0.1453 \ (9) & 0.1114 \ (10) & 0.7920 \ (10) & 4.49 \ (30) \\ C(35) & -0.0641 \ (9) & 0.1309 \ (9) & 0.7386 \ (9) & 3.80 \ (27) \\ C(36) & 0.3393 \ (8) & 0.0101 \ (8) & 0.8244 \ (8) & 2.54 \ (24) \\ C(37) & 0.4361 \ (9) & 0.0581 \ (9) & 0.8657 \ (9) & 3.55 \ (27) \\ C(38) & 0.4599 \ (10) & 0.1214 \ (10) & 0.9778 \ (10) & 5.22 \ (32) \\ C(39) & 0.3824 \ (10) & 0.1285 \ (10) & 1.0447 \ (10) & 4.84 \ (30) \\ C(40) & 0.2854 \ (10) & 0.0793 \ (10) & 1.0043 \ (10) & 4.44 \ (29) \\ C(41) & 0.2607 \ (9) & 0.0187 \ (9) & 0.8925 \ (9) & 3.56 \ (26) \\ C(42) & 0.3640 \ (9) & -0.2400 \ (9) & 0.7162 \ (8) & 2.90 \ (25) \\ C(43) & 0.2720 \ (9) & -0.2566 \ (10) & 0.7517 \ (9) & 4.03 \ (28) \\ C(44) & 0.2702 \ (10) & -0.3413 \ (11) & 0.7956 \ (10) & 5.19 \ (32) \\ C(45) & 0.3560 \ (11) & -0.4044 \ (10) & 0.8014 \ (10) & 5.15 \ (32) \\ C(46) & 0.4475 \ (10) & -0.3397 \ (10) & 0.7217 \ (9) & 4.54 \ (30) \\ C(47) & 0.4510 \ (9) & -0.3036 \ (10) & 0.7217 \ (9) & 3.66 \ (27) \\ \end{array}$	C(32)	-0.0318(9)	0.1598(9)	0.9573 (9)	3.92 (28)
$\begin{array}{cccc} C(34) & -0 \cdot 1453 \ (9) & 0 \cdot 1114 \ (10) & 0 \cdot 7920 \ (10) & 4 \cdot 49 \ (30) \\ C(35) & -0 \cdot 0641 \ (9) & 0 \cdot 1309 \ (9) & 0 \cdot 7386 \ (9) & 3 \cdot 80 \ (27) \\ C(36) & 0 \cdot 3393 \ (8) & 0 \cdot 0101 \ (8) & 0 \cdot 8244 \ (8) & 2 \cdot 54 \ (24) \\ C(37) & 0 \cdot 4361 \ (9) & 0 \cdot 0581 \ (9) & 0 \cdot 8657 \ (9) & 3 \cdot 55 \ (27) \\ C(38) & 0 \cdot 4599 \ (10) & 0 \cdot 1214 \ (10) & 0 \cdot 9778 \ (10) & 5 \cdot 22 \ (32) \\ C(39) & 0 \cdot 3824 \ (10) & 0 \cdot 1285 \ (10) & 1 \cdot 0447 \ (10) & 4 \cdot 84 \ (30) \\ C(40) & 0 \cdot 2854 \ (10) & 0 \cdot 0793 \ (10) & 1 \cdot 0043 \ (10) & 4 \cdot 44 \ (29) \\ C(41) & 0 \cdot 2607 \ (9) & 0 \cdot 0187 \ (9) & 0 \cdot 8925 \ (9) & 3 \cdot 56 \ (26) \\ C(42) & 0 \cdot 3640 \ (9) & -0 \cdot 2400 \ (9) & 0 \cdot 7162 \ (8) & 2 \cdot 90 \ (25) \\ C(43) & 0 \cdot 2702 \ (9) & -0 \cdot 2566 \ (10) & 0 \cdot 7517 \ (9) & 4 \cdot 03 \ (28) \\ C(44) & 0 \cdot 2702 \ (10) & -0 \cdot 3413 \ (11) & 0 \cdot 7956 \ (10) & 5 \cdot 19 \ (32) \\ C(45) & 0 \cdot 3560 \ (11) & -0 \cdot 4044 \ (10) & 0 \cdot 8014 \ (10) & 5 \cdot 15 \ (32) \\ C(46) & 0 \cdot 4475 \ (10) & -0 \cdot 3397 \ (10) & 0 \cdot 7217 \ (9) & 4 \cdot 54 \ (30) \\ C(47) & 0 \cdot 4510 \ (9) & -0 \cdot 3036 \ (10) & 0 \cdot 7217 \ (9) & 3 \cdot 66 \ (27) \\ \end{array}$	C(33)	-0.1295(9)	0.1250(9)	0.8986 (10)	4.23 (28)
$\begin{array}{ccccc} C(35) & -0.0641 (9) & 0.1309 (9) & 0.7386 (9) & 3.80 (27) \\ C(36) & 0.3393 (8) & 0.0101 (8) & 0.8244 (8) & 2.54 (24) \\ C(37) & 0.4361 (9) & 0.0581 (9) & 0.8657 (9) & 3.55 (27) \\ C(38) & 0.4599 (10) & 0.1214 (10) & 0.9778 (10) & 5.22 (32) \\ C(39) & 0.3824 (10) & 0.1285 (10) & 1.0447 (10) & 4.84 (30) \\ C(40) & 0.2854 (10) & 0.0793 (10) & 1.0043 (10) & 4.44 (29) \\ C(41) & 0.2607 (9) & 0.0187 (9) & 0.8925 (9) & 3.56 (26) \\ C(42) & 0.3640 (9) & -0.2400 (9) & 0.7162 (8) & 2.90 (25) \\ C(43) & 0.2720 (9) & -0.2566 (10) & 0.7517 (9) & 4.03 (28) \\ C(44) & 0.2702 (10) & -0.3413 (11) & 0.7956 (10) & 5.19 (32) \\ C(45) & 0.3560 (11) & -0.4044 (10) & 0.8014 (10) & 5.15 (32) \\ C(46) & 0.4475 (10) & -0.3336 (10) & 0.7217 (9) & 4.54 (30) \\ C(47) & 0.4510 (9) & -0.3036 (10) & 0.7217 (9) & 3.66 (27) \\ \end{array}$	C(34)	-0.1453(9)	0.1114(10)	0.7920(10)	4.49 (30)
$\begin{array}{ccccc} C(36) & 0&3393 \ (8) & 0&0101 \ (8) & 0&8244 \ (8) & 2&54 \ (24) \\ C(37) & 0&4361 \ (9) & 0&0581 \ (9) & 0&8657 \ (9) & 3&55 \ (27) \\ C(38) & 0&4399 \ (10) & 0&1214 \ (10) & 0&9778 \ (10) & 5&22 \ (32) \\ C(39) & 0&3824 \ (10) & 0&1285 \ (10) & 1&0447 \ (10) & 4&84 \ (30) \\ C(40) & 0&2854 \ (10) & 0&0793 \ (10) & 1&0447 \ (10) & 4&84 \ (30) \\ C(41) & 0&2607 \ (9) & 0&0187 \ (9) & 0&8925 \ (9) & 3&56 \ (26) \\ C(42) & 0&3640 \ (9) & -0&2400 \ (9) & 0&7162 \ (8) & 2&90 \ (25) \\ C(43) & 0&2720 \ (9) & -0&2566 \ (10) & 0&7517 \ (9) & 4&03 \ (28) \\ C(44) & 0&2702 \ (10) & -0&3413 \ (11) & 0&7956 \ (10) & 5&19 \ (32) \\ C(45) & 0&3560 \ (11) & -0&4044 \ (10) & 0&8014 \ (10) & 5&15 \ (32) \\ C(46) & 0&4475 \ (10) & -0&3897 \ (10) & 0&7637 \ (9) & 4&54 \ (30) \\ C(47) & 0&4510 \ (9) & -0&3036 \ (10) & 0&7217 \ (9) & 3&66 \ (27) \\ \end{array}$	C(35)	-0.0641(9)	0.1309(9)	0.7386(9)	3.80 (27)
$\begin{array}{ccccccc} C(37) & 0.4351 \ (9) & 0.0581 \ (9) & 0.98657 \ (9) & 3.55 \ (27) \\ C(38) & 0.4599 \ (10) & 0.1214 \ (10) & 0.9778 \ (10) & 5.22 \ (32) \\ C(39) & 0.3824 \ (10) & 0.1285 \ (10) & 1.0447 \ (10) & 4.84 \ (30) \\ C(40) & 0.2854 \ (10) & 0.0793 \ (10) & 1.0043 \ (10) & 4.44 \ (29) \\ C(41) & 0.2607 \ (9) & 0.0187 \ (9) & 0.8925 \ (9) & 3.56 \ (26) \\ C(42) & 0.3640 \ (9) & -0.2400 \ (9) & 0.7162 \ (8) & 2.90 \ (25) \\ C(43) & 0.2720 \ (9) & -0.2566 \ (10) & 0.7517 \ (9) & 4.03 \ (28) \\ C(44) & 0.2702 \ (10) & -0.3413 \ (11) & 0.7956 \ (10) & 5.19 \ (32) \\ C(45) & 0.3560 \ (11) & -0.4044 \ (10) & 0.8014 \ (10) & 5.15 \ (32) \\ C(46) & 0.4475 \ (10) & -0.3897 \ (10) & 0.7637 \ (9) & 4.54 \ (30) \\ C(47) & 0.4510 \ (9) & -0.3036 \ (10) & 0.7217 \ (9) & 3.66 \ (27) \\ \end{array}$	C(36)	0.3393(8)	0.0101(8)	0.8244(8)	2.54(24)
$\begin{array}{ccccc} C(33) & 0^{*}4399(10) & 0^{*}1214(10) & 0^{*}9778(10) & 5^{*}22(32) \\ C(39) & 0^{*}3824(10) & 0^{*}1285(10) & 1^{*}0447(10) & 4^{*}84(30) \\ C(40) & 0^{*}2854(10) & 0^{*}0793(10) & 1^{*}0043(10) & 4^{*}44(29) \\ C(41) & 0^{*}2607(9) & 0^{*}0187(9) & 0^{*}8925(9) & 3^{*}56(26) \\ C(42) & 0^{*}3640(9) & -0^{*}2400(9) & 0^{*}7162(8) & 2^{*}90(25) \\ C(43) & 0^{*}2720(9) & -0^{*}2566(10) & 0^{*}7517(9) & 4^{*}03(28) \\ C(44) & 0^{*}2702(10) & -0^{*}3413(11) & 0^{*}7956(10) & 5^{*}19(32) \\ C(45) & 0^{*}3560(11) & -0^{*}4044(10) & 0^{*}8014(10) & 5^{*}15(32) \\ C(46) & 0^{*}4475(10) & -0^{*}3897(10) & 0^{*}7637(9) & 4^{*}54(30) \\ C(47) & 0^{*}4510(9) & -0^{*}3036(10) & 0^{*}7217(9) & 3^{*}66(27) \\ \end{array}$	C(37)	0.4361(9)	0.0581(9)	0.865/(9)	3.55 (27)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(30)	0.4399(10)	0.1214(10) 0.1285(10)	0.9778(10)	5.22(32)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(39)	0.3624(10) 0.2854(10)	0.1203(10) 0.0703(10)	1.044/(10)	4.84 (30)
$\begin{array}{ccccccc} C(42) & 0.3640 \ (9) & -0.2400 \ (9) & 0.7162 \ (8) & 2.90 \ (25) \\ C(43) & 0.2720 \ (9) & -0.2566 \ (10) & 0.7517 \ (9) & 4.03 \ (28) \\ C(44) & 0.2702 \ (10) & -0.3413 \ (11) & 0.7956 \ (10) & 5.19 \ (32) \\ C(45) & 0.3560 \ (11) & -0.4044 \ (10) & 0.8014 \ (10) & 5.15 \ (32) \\ C(46) & 0.4475 \ (10) & -0.3897 \ (10) & 0.7637 \ (9) & 4.54 \ (30) \\ C(47) & 0.4510 \ (9) & -0.3036 \ (10) & 0.7217 \ (9) & 3.66 \ (27) \\ \end{array}$	C(40)	0.2634 (10)	0.0193(10)	0.8025 (0)	4144 (29)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(42)	0.26/0 (0)	-0.2400(0)	0.7167 (9)	3.00 (20)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(42)	0.2720 (0)	-0.2400 (9)	0.7517(0)	2.90 (23)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(44)	0.2720(3) 0.2702(10)	-0.3413(11)	0.7956 (10)	4·03 (28) 5·10 (22)
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	C(45)	0.3560(11)	-0.4044(10)	0.8014(10)	5.15 (32)
C(47) 0.4510 (9) -0.3036 (10) 0.7217 (9) 3.66 (27)	C(46)	0.4475(10)	-0.3897(10)	0.7637(9)	4.54 (30)
	C(47)	0.4510 (9)	-0.3036(10)	0.7217(9)	3.66(27)

Table 1 (cont.)

H(C13)	0.193	0.232	0.236
H(C14)	0.207	0.401	0.183
H(C15)	0.305	0.564	0.313
H(C16)	0.387	0.576	0.488
H(C17)	0.375	0.408	0.544
H(C18)	0.065	0.120	0.437
H(C19)	-0.115	0.209	0.460
H(C20)	-0.125	0.407	0.565
H(C21)	0.029	0.523	0.657
H(C22)	0.212	0.437	0.652
H(C25)	0.040	0.370	0.803
H(C26)	0.028	0.568	0.935
H(C27)	0.176	0.659	1.066
H(C29)	0.358	0.360	0.961
H(C31)	0.128	0.209	0.956
H(C33)	-0.194	0.110	0.938
H(C34)	-0.222	0.084	0.746
H(C35)	-0.017	0.124	0.653
H(C37)	0.496	0.047	0.812
H(C38)	0.535	0.162	1.011
H(C39)	0.399	0.176	1.131
H(C40)	0.227	0.086	1.058
H(C41)	0.184	-0.053	0.859
H(C43)	0.204	-0.506	0.746
H(C44)	0.200	-0.358	0.824
H(C45)	0.354	-0.467	0.836
H(C46)	0.515	-0.444	0.765
H(C47)	0.523	-0.582	0.693
H'(C11)	0.423	-0.106	0.289
H"(C11)	0.453	-0.199	0.357

* *B* values of Br(1) and Br(2) are the equivalent isotropic temperature factors proposed by Hamilton (1959). The anisotropic temperature factors of Br(1) and Br(2) in the form $T = \exp \{-(h^2\beta_{11} + k^2\beta_{22} + l^2\beta_{33} + 2hk\beta_{12} + 2hl\beta_{13} + 2kl\beta_{23}) \times 10^{-4}\}$ are as follows:

	β_{11}	\$22	β33	β_{12}	β_{13}	β_{23}
$B_1(1)$	168 (2)	71 (1)	76 (1)	-30(1)	-3(1)	3 (1)
Br(2)	92 (1)	186 (2)	103 (2)	18 (1)	44 (1)	71 (2)

flexions had intensities above background. Standard deviations on intensities were calculated according to the expression used by Doedens & Ibers (1967): $\sigma(I) = [P + 0.25(B_1 + B_2) (T_P/T_B)^2 + (0.03 I)^2]^{1/2}$ where P is the total peak count in a scan of time T_P , B_1 and B_2 are the background counts each in a time T_B , I is the intensity, equal to $P - 0.5 (T_P/T_B) (B_1 + B_2)$; reflexions with $I < 3\sigma(I)$ were considered unobserved and excluded from further computations. The Lorentz-polarization correction was applied. The absorption correction was neglected owing to the small dimensions of the crystal and the low absorption coefficient ($\mu = 22.4 \text{ cm}^{-1}$ for Mo $K\alpha$).

Structure determination and refinement

The coordinates for the two independent Br atoms in the cell were readily deduced from a three-dimensional Patterson function. In this way only about 15% of the electrons in the cell were located (conventional $R \simeq$ 0.56) and Fourier and difference Fourier syntheses, computed with the signs from bromide atoms, failed to show any groups of maxima which had any chemical significance. An α -synthesis was then computed (Raman, 1961) from which it was possible to find 16 atoms. These atoms were considered to be carbon atoms ($R \simeq 0.48$). A Fourier synthesis was computed using as coefficients the structure factors phased from this fragment of the molecule, and with amplitudes multiplied by $w = \tanh(F_{obs} \cdot F_P \ge f_{MJ}^2)$ where F_P is the

contribution to the amplitude by the partially known structure and f_{MJ} is the scattering factor of the missing *j*th atom (Woolfson, 1956). This map, which was very well resolved, gave the locations of 25 new atoms (R = 0.39). Successive Fourier syntheses revealed the whole structure and improved the model to give an R value of 0.26.

At this stage the refinement of the structure was continued by the least-squares method with the fullmatrix program *ORFLS*. The quantity minimized was: $w||F_o| - |F_c||^2$, with weights w assigned as $1/[\sigma(F)]^2$. In three cycles, with individual isotropic thermal parameters, R was lowered to 0.13.

Because of the unfavourable observations-to-parameters ratio, the possibility of refining all the atoms anisotropically was ruled out.

Two more cycles performed with anisotropic thermal parameters for the bromine atoms alone, reduced R to 0.08. At this stage the contribution of the hydrogen atoms at theoretical positions computed assuming the value 1.075 Å for the C-H bond length and thermal parameters equal to those of the carbon atoms to which they are bonded was introduced. A final cycle, refining only the non-hydrogen atoms, yielded the coordinates, thermal parameters and relative standard deviations listed in Table 1. The R value reached the final value 0.067 for 2230 observed reflexions. The atomic scattering factors used were those listed for Br, O, N, C and H in International Tables for X-ray Crystallography (1962); an anomalous dispersion correction was applied to the bromine scattering curve. Observed and calculated structure factors are given in Table 2.



Fig. 1. The cell contents projected along the a axis.

Table 2. Observed and calculated structure factors (\times 10)

m. 1.11-1 m.				1 111111 1 1111111 1 1111111 1 1111111 1	191977 – 1929292944997449 – 122999727428 – 90922200000 – 909241950 – 9000 Å 4000 Å 4000 Å 4000 Å 4000000 Å 400000000		2007-00420042000 - 4007000000 - 4007000000 - 400700000 - 4007000 - 4007000 - 400700 - 400700000 - 40070000000 - 4007000000 - 40070000000 - 40070000000 - 40070000000 -		1977 - 1979-2072-2072 - 2072-2023 - 2072-202	 	1999-1997-1997 - 1997-1997 - 1997-1997 - 1997-1997 - 1998 - 1997 - 1997 - 1997 - 1997-199
-4 299 -349 Hi 1, -3 -8 297 278 -6 261 271	H, 9, -2 -7 167 -200 -6 291 -229 -5 261 -262 -6 289 -257	U 135-132 1 1159-1182 2 279 273 4 745 -724 5 679 -657 6 125 144 8 212 -226	-1 227 -229 U 310 -315 +7. 1 -V 1/1 -243 -8 244 191	-3 7C3 -7C9 -2 102 -06 -1 1019-1005 C 124 13C 1 109 -14C 2 637 -629 4 434 -452	0 280 290 1 374 -380 +, -6, 2 -3 131 126 -2 249 293	7 337 -345 H. 4, 2 -11 188 180 -7 576 577 -5 235 245	0 144 139 2 256 -318 H5, 3 -12 149 -171 -8 262 -305	-4 419 433 -3 368 -283 -2 327 353 -1 658 620 1 173 763 2 128 -146 3 763 725	-0 200 324 -7 185 -163 -0 277 -286 -3 455 -445 -4 236 -231 -2 506 -511 0 547 -558 2 608 -635	0 324 326 1 341 345 2 347 -332 3 413 380 5 402 380 8 380 358 P, 5, 4	4 129 111

276

Table 2 (cont.)

5 5 101015151515151515151515151515151515	H, 4, 5 - 4,		11777777777777777777777777777777777777			H, -12, 0 H, -11, 0 H, -12, 0 H, -11, 0 H, -12, 0	1417-1717-1017-1017-1017-1017-1017-1017-		- 4 222 227 2 4 4 5 5 5 5 5 5 5 5 5 5 5 5 5 5 5 5 5		b, -1, 11 r, -1, 11 <td< th=""></td<>
--	---	--	--	--	--	--	--	--	---	--	---

Results and discussion

The structure of 4'-methylene-1,2-di-*m*-bromophenyl-1', 2', 6', 7' - tetraphenyl - spiro {pyrazolidine - 4,8' - [8'H, 4'H]-benzo[1,2-c:4,5-c']dipyrazoline}-3,5,3',5' - tetraone projected along the *a* axis is shown in Fig. 1. The conformation of the molecule is represented in Figs. 2 and 3. Bond lengths and angles are listed in Tables 3 and 4. Chemically equivalent bond lengths and angles are averaged and shown in Fig. 4, together with the atomic numbering scheme.

The most important feature of the molecular structure is the presence of cross conjugation involving the system O(1), C(7), C(5), C(6), C(4), C(11), C(3), C(2), C(8) and O(2): these atoms are roughly coplanar. The equation of the best plane (plane A) and atomic deviations from this plane are listed in Table 5. Because of conjugation, C(7)-O(1) and C(8)-O(2) bonds, 1.25 Å long, are significantly longer than the other two carbonylic bonds of the molecule, C(9)-O(3) and C(10)-O(4), which are 1.20 Å long; furthermore the C(2)-C(3), C(5)-C(6) and C(4)-C(11) double bonds are slightly longer than normal localized double bonds, while the single C(5)-C(7), C(4)-C(5), C(3)-C(4) and C(3)-C(8) bonds have a certain double bond character. Probably the two phenyl rings linked to N(1) and N(4) contribute to the resonating system, since the relative C-N bond lengths are significantly shorter than the other analogous C-N distances: 1.42 against 1.46 Å. Other interatomic distances in the molecule are in good agreement with those reported in literature.

The pyrazolinic and pyrazolidinic rings are not per-

fectly planar, with the two nitrogen atoms in each ring displaced on opposite sides of the ring plane; equations of the best planes and atomic deviations from planarity are reported in Table 5 (planes B, C and D). Phenyl rings are planar within the limits of experimental error (see Table 5, planes E to J).



Fig. 2. The molecule as viewed along the a axis.



Fig. 3. The molecule as viewed along a direction lying in the plane of the three condensed rings.

Table 3. Bond distances

Estimated standard	deviations	are between 0.012	and 0·018 Å
Br(1) - C(28)	1.88 Å	C(13) - C(14)	1·40 Å
Br(2) - C(32)	1.87	C(14) - C(15)	1.36
O(1) - C(7)	1.25	C(15) - C(16)	1.32
O(2) - C(8)	1.25	C(16) - C(17)	1.41
O(3) - C(9)	1.20	C(18) - C(19)	1.41
O(4) - C(10)	1.19	C(18) - C(23)	1.38
N(1) - N(2)	1.43	C(19) - C(20)	1.36
N(1) - C(7)	1.38	C(20) - C(21)	1.34
N(1) - C(12)	1.41	C(21) - C(22)	1.43
N(2) - C(6)	1.40	C(22) - C(23)	1.37
N(2) - C(23)	1.46	C(24) - C(25)	1.41
N(3)N(4)	1.44	C(24) - C(29)	1.37
N(3) - C(2)	1.39	C(25) - C(26)	1.40
N(3)C(36)	1.46	C(26)-C(27)	1.33
N(4)C(8)	1.36	C(27)-C(28)	1.36
N(4)C(42)	1.42	C(28)-C(29)	1.41
N(5)—N(6)	1.43	C(30) - C(31)	1.34
N(5) - C(9)	1.37	C(30)-C(35)	1.39
N(5) - C(30)	1.44	C(31)-C(32)	1.39
N(6) - C(10)	1.39	C(32) - C(33)	1.37
N(6)—C(24)	1.45	C(33)–C(34)	1.34
C(1) - C(2)	1.52	C(34) - C(35)	1.40
C(1) - C(6)	1.48	C(36) - C(37)	1.35
C(1)—C(9)	1.51	C(36) - C(41)	1.41
C(1) - C(10)	1.54	C(37)–C(38)	1.39
C(2) - C(3)	1.35	C(38) - C(39)	1.39
C(3) - C(4)	1.44	C(39)-C(40)	1.35
C(3) - C(8)	1.46	C(40) - C(41)	1.39
C(4) - C(5)	1.45	C(42) - C(43)	1.37
C(4) - C(11)	1.34	C(42)–C(47)	1.37
C(5) - C(6)	1.35	C(43) - C(44)	1.40
C(5) - C(7)	1.42	C(44) - C(45)	1.35
C(12)-C(13)	1.36	C(45) - C(46)	1.37
C(12)-C(17)	1.38	C(46)-C(47)	1.40

Table 4. Bond angles

Estimated standard deviations are between 0.8 an	d l	l·l°	
--	-----	------	--

C(2) - C(1) - C(6)	105·9°	C(2) - N(3) - N(4)	104·9°
C(6) - C(1) - C(10)	110.1	C(2) - N(3) - C(36)	120.2
C(10)-C(1)-C(9)	102.1	N(4) - N(3) - C(36)	111.8
C(9) - C(1) - C(2)	108.5	N(3) - N(4) - C(8)	109.3
C(2) - C(1) - C(10)	114.8	N(3) - N(4) - C(42)	120.1
C(6) - C(1) - C(9)	115.8	C(8) - N(4) - C(42)	127.4
C(1) - C(2) - C(3)	126.1	C(9) - N(5) - N(6)	109.9
C(1) - C(2) - N(3)	121.9	C(9) - N(5) - C(30)	122.2
C(3) - C(2) - N(3)	111.6	N(6) - N(5) - C(30)	120.0
C(2) - C(3) - C(4)	1 2 4·7	N(5) - N(6) - C(10)	109.7
C(2) - C(3) - C(8)	106.6	N(5) - N(6) - C(24)	119.4

C(4) - C(3) - C(8)	128.4	C(10) - N(6) - C(24)	122.6
C(3) - C(4) - C(5)	111.2	N(1) - C(12) - C(13)	118.1
C(3) - C(4) - C(11)	122.5	N(1) - C(12) - C(17)	119.5
C(5) = C(4) = C(11)	126.3	C(13)-C(12)-C(17)	122.4
C(4) - C(5) - C(6)	124.3	C(12)-C(13)-C(14)	118.9
C(4) - C(5) - C(7)	127.5	C(13)-C(14)-C(15)	117.3
C(6) - C(5) - C(7)	108.1	C(14) - C(15) - C(16)	124.8
C(1) - C(6) - C(5)	127.5	C(15) = C(16) = C(17)	119.1
C(1) - C(6) - N(2)	121.6	C(16) - C(17) - C(12)	117.4
C(5) - C(6) - N(2)	110.9	N(2) - C(23) - C(18)	119.4
C(5) - C(7) - O(1)	130.9	N(2) - C(23) - C(22)	117.7
C(5) - C(7) - N(1)	106.9	C(18)-C(23)-C(22)	122.9
O(1) - C(7) - N(1)	122.2	C(23)-C(18)-C(19)	118.0
C(3) - C(8) - O(2)	128.9	C(18) - C(19) - C(20)	119.4
C(3) - C(8) - N(4)	107.2	C(19) - C(20) - C(21)	122.4
O(2) - C(8) - N(4)	123.9	C(20) - C(21) - C(22)	120.3
C(1) - C(9) - O(3)	125.4	C(21) - C(22) - C(23)	116.9
C(1) - C(9) - N(5)	109.6	N(3) - C(36) - C(37)	122.2
O(3) - C(9) - N(5)	124.8	N(3) - C(36) - C(41)	116.3
C(1) - C(10) - O(4)	127.3	C(41) - C(36) - C(37)	121.6
C(1) - C(10) - N(5)	108.2	C(36) - C(37) - C(38)	120.4
O(4) - C(10) - N(5)	124.4	C(37) - C(38) - C(39)	117.7
C(7) - N(1) - N(2)	109.2	C(38) - C(39) - C(40)	122.3
C(7) - N(1) - C(12)	127.3	C(39) - C(40) - C(41)	120.1
N(2) - N(1) - C(12)	119.2	C(40) - C(41) - C(36)	117.9
C(6) - N(2) - N(1)	104.6	N(4) - C(42) - C(43)	119.8
C(6) - N(2) - C(23)	122.4	N(4) - C(42) - C(47)	117.8
N(1) - N(2) - C(23)	111.2	C(47) - C(42) - C(43)	122.4
C(42)-C(43)-C(44)	117.1	C(33) - C(34) - C(35)	121.6
C(43)-C(44)-C(45)	120.9	C(34)-C(35)-C(30)	117.0
C(44)-C(45)-C(46)	122.1	N(6) - C(24) - C(25)	119.1
C(45)-C(46)-C(47)	117.5	N(6) - C(24) - C(29)	116.9
C(46)-C(47)-C(42)	120.0	C(29)-C(24)-C(25)	123.9
N(5) - C(30) - C(31)	120.9	C(24)-C(25)-C(26)	116.3
N(5) - C(30) - C(35)	116.6	C(25)-C(26)-C(27)	120.6
C(35)-C(30)-C(31)	122.5	C(26)-C(27)-C(28)	122.4
C(30)-C(31)-C(32)	117.7	C(27)-C(28)-Br(1)	121.7
C(31)-C(32)-Br(2)	119.4	C(27)-C(28)-C(29)	120.9
C(31)-C(32)-C(33)	121.6	Br(1)-C(28)-C(29)	117.4
Br(2)-C(32)-C(33)	119.1	C(28)-C(29)-C(24)	115.8
C(32)-C(33)-C(34)	119.3		

The three condensed rings are slightly twisted. The extent of torsional buckling is shown in Fig. 3, where the molecule is projected along a direction lying in the best plane through the rings (plane K, Table 5). Some short intramolecular contacts between atoms not directly bonded are reported in Fig. 2. The packing of the molecules is shown in Fig. 1. Pairs of mole-

Table 5. Equations of least-squares planes

Equations are in the form lx + my + nz = p, where x, y and z are triclinic fractional atomic coordinates referred to the cell axes.

Plane	Atoms defining the plane	l	m	n	р
A	O(1), O(2), C(2), C(3), C(4), C(5), C(6), C(7),				_
	C(8), C(11)	10.353	4.679	2.670	4.860
В	N(1), N(2), C(5), C(6), C(7)	10.549	3.277	3.667	5.219
С	N(4), N(3), C(2), C(3), C(8)	10.290	4.465	3.002	5.106
D	N(5), N(6), C(1), C(9), C(10)	-2.176	10.774	-9.769	- 5.680
E	C(12), C(13), C(14), C(15), C(16), C(17)	11.397	-2.509	- 5.292	0.383
F	C(18), C(19), C(20), C(21), C(22), C(23)	-2.430	-7.302	12.742	4.538
G	C(36), C(37), C(38), C(39), C(40), C(41)	-3.656	11.950	- 5.744	-5.860
H	C(42), C(43), C(44), C(45), C(46), C(47)	2.782	4.519	9.031	6.393
Ι	C(30), C(31), C(32), C(33), C(34), C(35)	-3.336	11.812	-1.138	0.898
J	C(24), C(25), C(26), C(27), C(28), C(29)	- 5.691	-7.742	11.685	6.342
Κ	O(1), O(2), N(1), N(2), N(3), N(4), C(1), C(2),				
	C(3), C(4), C(5), C(6), C(7), C(8), C(11)	10.488	4.482	2.643	4.917

Table 4 (cont.)



Table 5 (cont.)

cules related by the inversion centre at $\frac{1}{2}$, 0, $\frac{1}{2}$ are

located in such a way that the system of the three con-

densed rings and methylene and keto groups are partially overlapped. The distance between the best planes

through two adjacent triple ring systems is 3.30 Å. In-

termolecular distances are normal; those shorter than

the sum of the van der Waals radii, assuming for Br, O,

N, and C the values 1.85, 1.50, 1.55, 1.75 Å respectively, are listed in Table 6. The contact Br(1)-O(4'), 3.35 Å long, with the C(28)-Br(1) bond approximately pointing towards O(4) [angle C(28)-Br(1)···O(4')= 166°] is perhaps indicative of some 'charge transfer' interaction between oxygen and halogen (Gaultier, Hauw & Schvoerer, 1971).



Fig. 4. Molecular dimensions and atomic numbering scheme.

Table 6. Shortest intermolecular approaches

Br(1) - O(4')	(1 - x,	1 - y, 2 - z	3.35 Å
O(2) - C(16')	(x, \cdot)	-1+y, z	3.19
O(3) - C(18')	(-x,	-y, 1-z	3.17
C(3) - C(11')	(1 - x,	-y, 1-z	3.48
C(4) - C(4')	(1 - x,	-y, 1-z	3.42
C(7) - C(8')	(1 - x)	-y, 1-z	3.49
C(26) - C(26')	(-x,	1 - y, 2 - z	3.36
C(38) - C(38')	(1 - x)	-y, 2-z	3.45

The authors thank Prof. L. Sacconi for the use of the automatic diffractometer at the Istituto di Chimica Generale of Florence University, Prof. P. Orioli for his valuable suggestions during data collection and Prof. G. Adembri for helpful discussions on the molecular structure.

References

- DOEDENS, R. J. & IBERS, J. A. (1967). Inorg. Chem. 6, 204-210.
- GAULTIER, J., HAUW, C. & SCHVOERER, M. (1971). Acta Cryst. B27, 2199-2204.
- HAMILTON, W. C. (1959). Acta Cryst. 12, 609-610.
- International Tables for X-ray Crystallography (1962). Vol. III. Birmingham: Kynoch Press.
- KEMPSTER, C. J. E. & LIPSON, H. (1972). Acta Cryst. B28, 3674.
- MANGIAVACCHI, S. (1968). Atti Accad. Fisiocritici, 17, 2234–2240.
- MANGIAVACCHI, S., LAZZI, L. & RIDI, M. (1966). Atti Accad. Fisiocritici, 15, 1027–1044.
- RAMAN, S. (1961). Acta Cryst. 14, 148-150.
- WOOLFSON, M. M. (1956). Acta Cryst. 9, 804-810.