BULLETIN OF THE CHEMICAL SOCIETY OF JAPAN VOL. 43 1962—1966 (1970)

Crystal Structure of Bis(nitrosocyclohexane)*1

Masamitsu TANIMURA,*2 Kiyotomi KOBORI,*3 Michio KASHIWAGI and Yukio KINOSHITA*3

Basic Research Laboratory, Toray Industries Inc., Tebiro, Kamakura

(Received December 1, 1969)

The crystal structure of bis(nitrosocyclohexane), $(C_6H_{11}NO)_2$, has been determined by means of three-dimensional X-ray analysis by the photographic method. The crystal belongs to the triclinic space group $P\bar{1}$, with cell parameters, $a=10.133$ A, $b=6.758$ A, $c=5.583$ A, $\alpha=121°42'$, $\beta=96°50'$, $\gamma=78°06'$. The unit cell contains one molecule. The nitrogen and oxygen atoms and the carbon atoms bound to nitroso-groups, CNO–O'N'C', are almost coplanar, and the oxygen atoms take a trans configuration around the N–N' bond. The cyclohexane ring takes a chair form and the nitrogen atom is bound to an equatorial bond. The CNO–O'N'C' plane is twisted from the cyclohexane ring by 73°.

Bis(nitrosocyclohexane), which is easily converted into cyclohexanone oxime by light, is a very important material from an industrial point of view. However, its structure has not been determined. The present investigation was carried out in order to determine a detailed structure by means of X-ray diffration.

Experimental

Single crystals of bis(nitrosocyclohexane), $(C_6H_{11}NO)_2$, were obtained by slow evaporation from methanol solution as triclinic plates tabular on (100). The cell dimensions were determined from the oscillation and Weissenberg photographs taken around the three axes using Cu$K\alpha$ radiation. The space group was determined from the measurement of piezo-electricity and from the Wilson ratio test. The results are listed in Table 1.

Intensity data were recorded on the equi-inclination Weissenberg photographs taken around b-axis up to the fifth layer and around c-axis up to the fourth layer, using the multiple film technique. The relative intensities, estimated visually with a set of standard scales, were corrected for the Lorentz-polarization factors. No absorption corrections were made. A total of 1096 independent reflections were observed.

TABLE 1. CRYSTAL DATA

Triclinic	
$a=10.133$ A	$\gamma=78°06'$
$b=6.758$	$Z=1$
$c=5.583$	$p\bar{1}$
$\alpha=121°42'$	$\mu=0.0794\ \mathrm{cm}^{-1}$ (Cu$K\alpha$)
$\beta=96°50'$	

Structure Determination

The space group was assumed to be $P\bar{1}$. Choice of the space group was corroborated by structure determination.

An approximate structure was readily deduced from the three dimensional Patterson syntheses. The structure thus obtained was refined by Fourier

*1 Presented at the Symposium of Molecular Structure, Nagoya, October, 1965.

*2 Present address: Pioneering Research and Development Laboratories, Toray Industries Inc, Sonoyama, Otsu.

*3 Present address: Aichi Laboratory, Toray Industries Inc, Nishiku, Nagoya.

Table 2a. Atomic coordinates

Atom	$x(10^3\sigma_x)$	$y(10^3\sigma_y)$	$z(10^3\sigma_z)$	Atom	$x(10^3\sigma_x)$	$y(10^3\sigma_y)$	$z(10^3\sigma_z)$
C(1)	0.1306(4)	0.1343(5)	−0.0576(5)	H(2e)	0.0710(56)	0.5085(65)	0.2417(64)
C(2)	0.1557(5)	0.3854(5)	0.1571(6)	H(3a)	0.1793(56)	0.4580(65)	−0.1932(64)
C(3)	0.2416(6)	0.4579(6)	0.0143(7)	H(3e)	0.2524(56)	0.6503(65)	0.1859(64)
C(4)	0.3752(6)	0.2885(8)	−0.0829(7)	H(4a)	0.4218(55)	0.2877(65)	0.1004(64)
C(5)	0.3520(5)	0.0372(7)	−0.2855(7)	H(4e)	0.4279(56)	0.2931(65)	−0.2378(64)
C(6)	0.2637(4)	−0.0394(6)	−0.1519(6)	H(5a)	0.2943(56)	0.0160(65)	−0.4743(64)
N	0.0479(3)	0.0609(4)	0.0760(4)	H(5e)	0.4398(56)	−0.0661(65)	−0.3561(64)
O	0.0689(4)	0.1113(5)	0.3296(4)	H(6a)	0.3172(56)	−0.0546(65)	0.0187(64)
H(1a)	0.0860(55)	0.1213(65)	−0.2324(64)	H(6e)	0.2354(56)	−0.1914(65)	−0.3354(64)
H(2a)	0.2144(56)	0.3585(56)	0.3282(64)				

Table 2b. Anisotropic thermal parameters in the form $\exp(-(B_{11}h^2 + B_{22}k^2 + B_{33}l^2 + B_{12}hk + B_{13}hl + B_{23}kl))$

Atom	$B_{11}(10^3\sigma)$	$B_{22}(10^3\sigma)$	$B_{33}(10^3\sigma)$	$B_{12}(10^3\sigma)$	$B_{13}(10^3\sigma)$	$B_{23}(10^3\sigma)$
C(1)	0.0055 (0)	0.0228 (1)	0.0298 (2)	−0.0085 (1)	−0.0014 (1)	0.0314 (3)
C(2)	0.0129 (1)	0.0202 (2)	0.0431 (2)	−0.0090 (2)	0.0112 (2)	0.0268 (3)
C(3)	0.0179 (1)	0.0283 (2)	0.0550 (3)	−0.0236 (2)	0.0074 (2)	0.0333 (4)
C(4)	0.0108 (1)	0.0586 (3)	0.0620 (3)	−0.0332 (2)	−0.0054 (2)	0.0644 (5)
C(5)	0.0078 (1)	0.0399 (2)	0.0601 (3)	−0.0063 (2)	0.0156 (2)	0.0477 (4)
C(6)	0.0076 (0)	0.0275 (2)	0.0475 (2)	−0.0012 (1)	0.0104 (2)	0.0388 (3)
N	0.0060 (0)	0.0266 (1)	0.0243 (1)	−0.0101 (1)	−0.0041 (1)	0.0316 (2)
O	0.0137 (0)	0.0602 (2)	0.0298 (1)	−0.0385 (1)	−0.0166 (1)	0.0588 (3)
	$B(\sigma)$					
all H	3.0 (1.3)					

syntheses projected along the *b*- and *c*-axes, and later by a three dimensional isotropic least-squares method with ERBR1 program.[1] After a few cycles of this refinement, the structure was further refined by a block-diagonal anisotropic least-squares method with HBLS program.[2] Finally the difference Fourier syntheses were carried out in order to find the positions of the hydrogen atoms.

The final discrepancy factor, $R=\sum||F_o|-|F_c||/\sum|F_o|$ became 0.105 for all the observed reflections. Atomic scattering factors were taken from the International Tables for X-ray Crystallography. Final atomic parameters and their standard deviations are listed in Tables 2a and 2b. The observed and calculated structure factors are listed in Table 3.

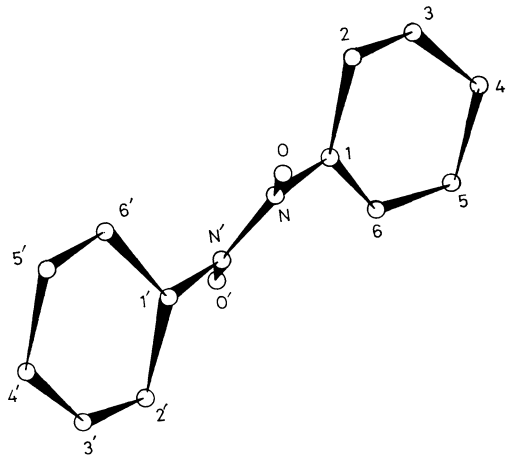

Fig. 1. Schematic diagram projected along c-axis.

Results and Discussion

The interatomic distances and bond angles are listed in Table 4. A schematic diagram of the molecule along the *c*-axis is shown in Fig. 1.

The atoms C(1), O, N, C′(1), O′ and N′ are almost coplanar, and oxygen atoms are placed *trans* with regard to the N–N′ bond. The cyclohexane ring takes a chair form, and the C(1)–N bond is equatorial.

The least-squares plane through C(1), O, N, N′, O′ and C′(1) is given by the equation.

$$0.9532X - 1.3248Y + 0.1805Z = 0 \quad (1)$$

Deviations of the atoms from this plane are apparently not significant as shown in Table 5.

The planes through C(1), C(3), C(5) and through C(2), C(4) and C(6) are given by the fol-

1) J. H. Van den Hende, "Crystallographic Structure Factor and Least-Squares Refinement Program" (1961).

2) T. Ashida, "Block-Diagonal Matrix Least-Squares Refinement Program" (1964).

lowing equations.

$$0.3534X - 0.3270Y + 0.7918Z = -0.0837 \quad (2)$$

$$0.4569X - 0.2034Y + 0.7720Z = 0.7862 \quad (3)$$

Planes (2) and (3) are almost parallel, and these planes make an average angle of 73° against plane (1).

The distances of N–N bond (1.32Å) and N–O bond (1.27 Å) suggest their double-bond characters. The N–C(1) bond (1.49 Å) has a single-bond character.

The C–C bond distances and C–C–C angles of the cyclohexane ring are normal. The bond distances and bond angles including the hydrogen atoms do not considerably deviate from the normal values as shown in Table 4. The values of other bond distances and bond angles including hydrogen are of the same order as those listed in Table 4.

All the intermolecular atomic distances observed are within the limits of experimental error, equal to or greater than the sum of the van der Waals radii of the corresponding atoms.

TABLE 3. OBSERVED AND CALCULATED STRUCTURE FACTORS

The running indices are *hl*, the value for *k* immediately precedes the group. The central column is $|F_o|$, the right hand column F_c.

	0			1	2	9.7	9.2	-5	1	1.1	0.9	2	-3	7.8	-6.9	0	-1	8.6	7.6	-4	3	2.4	-2.6	2	3	5.6	-6.1
0	1	33.4	39.7	-1	2	19.2	19.0	-5	-1	0.9	-0.8	2	4	3.3	-3.6	0	-2	1.1	0.9	-4	-3	0.9	-0.7	-2	3	1.9	1.6
0	2	1.3	0.3	-1	-2	1.0	1.0	-5	-1	2.4	2.9	-2	4	2.9	1.2	0	-2	3.3	-2.0	4	-3	1.3	-0.6	-2	-3	0.9	-0.2
0	3	7.0	6.7	-1	-2	6.0	-5.3	5	2	3.6	-3.2	-2	-4	1.3	-1.2	0	3	3.0	2.3	4	-4	4.3	-4.2	2	-3	1.3	0.0
0	4	3.0	-2.9	1	3	2.0	1.6	-5	2	2.1	1.2	2	-4	7.8	7.9	0	-3	4.7	4.7	-4	5	3.9	-4.3	2	4	1.2	-1.1
0	5	2.8	-3.2	-1	3	10.7	11.2	5	-2	6.7	6.0	-2	5	5.1	-5.0	0	4	1.4	-1.7	4	-5	2.3	-2.5	-2	4	5.5	-5.1
1	0	33.4	29.1	-1	-3	4.3	4.3	-5	3	2.6	2.0	2	-5	1.9	1.1	0	5	1.1	0.9	-4	6	1.6	-1.5	-2	-4	1.9	1.0
1	1	9.3	-7.9	-1	-3	5.4	5.3	-5	-3	2.9	-2.7	-2	6	2.2	-1.8	0	-5	4.5	-5.0	4	-6	0.9	0.7	2	-4	4.0	-3.8
-1	1	2.4	4.6	1	4	3.2	-3.4	-5	4	7.3	7.5	2	-6	1.1	-0.9	1	0	4.0	-3.8	5	0	1.9	-1.9	-2	5	2.1	-2.3
-1	2	12.3	-13.0	-1	4	1.5	-1.9	5	-4	0.7	-0.4	3	0	17.3	-18.9	-1	0	37.0	37.4	5	1	0.8	-0.4	2	-5	4.8	4.4
1	3	5.1	4.5	-1	-4	2.3	-1.7	-5	5	4.0	4.5	-3	0	9.1	9.8	1	1	12.6	-10.8	-5	-1	0.8	-0.2	-2	6	1.3	0.9
-1	3	3.9	5.3	1	-4	3.7	2.8	5	-5	6.1	7.0	3	1	10.5	-9.9	-1	1	3.1	-2.9	5	-1	2.1	-2.4	3	0	8.4	-8.3
-1	4	2.2	-3.5	-1	5	4.8	-6.0	-5	6	0.9	-0.8	-3	1	7.5	7.0	-1	-1	18.1	18.0	5	2	1.2	-0.7	-3	0	3.3	3.5
-1	5	7.9	-9.0	-1	-5	0.6	-0.8	5	-6	3.0	2.1	-3	-1	10.6	11.0	1	-1	2.6	2.5	-5	2	1.2	-1.1	3	1	4.1	3.8
-1	6	2.2	-1.8	1	-5	2.5	-2.7	6	0	1.2	0.8	3	-1	6.1	5.7	1	2	3.5	-2.8	-5	-2	0.8	-0.6	-3	1	7.4	6.7
2	0	23.8	22.3	-1	6	1.8	-1.7	-6	0	2.3	-2.6	3	2	1.4	-0.6	-1	2	10.9	-10.4	5	-2	9.5	10.3	-3	-1	2.7	-3.0
2	1	4.0	2.5	1	-6	1.4	-1.3	-6	-1	2.8	-3.4	-3	2	1.6	-1.5	-1	-2	1.1	-0.2	-5	3	1.4	-1.3	3	-1	2.7	-3.0
-2	1	13.9	13.3	2	0	12.9	12.8	-6	-1	2.3	2.9	-3	-2	8.5	8.6	1	-2	3.8	-3.6	5	-3	9.6	10.2	3	2	1.7	-1.2
2	2	3.7	3.4	-2	0	5.0	5.4	-6	2	1.2	1.0	3	-2	10.8	10.2	1	3	6.6	-6.2	-5	4	0.9	-0.9	-3	2	2.3	1.5
-2	2	10.2	-9.4	2	1	11.7	10.6	-6	-2	5.8	5.7	3	3	1.4	-1.4	-1	3	1.3	-0.9	5	-4	1.6	0.8	-3	-2	1.2	0.4
-2	3	3.7	4.2	-2	1	13.3	14.2	-6	3	1.8	1.4	-3	3	3.3	2.4	-1	-3	2.6	2.0	-5	5	2.9	-3.0	3	-2	7.2	6.2
2	4	0.8	-1.0	-2	-1	3.2	-3.4	6	-3	3.5	2.7	-3	-3	1.6	1.5	1	-3	4.3	3.4	5	-5	3.4	2.7	3	3	4.1	-4.4
-2	4	5.6	5.7	-2	-1	17.1	18.5	-6	4	3.5	2.9	3	-3	8.7	9.4	1	4	3.4	-3.2	-5	6	2.5	-2.1	-3	3	3.8	2.9
-2	5	3.7	-3.6	2	2	7.6	6.3	6	-4	2.3	1.4	-3	4	2.0	1.5	-1	4	4.6	-4.5	5	-6	1.7	1.2	-3	-3	0.7	0.7
3	0	5.1	4.8	-2	2	7.7	-6.6	7	0	1.8	0.2	-3	-4	0.8	-0.9	-1	-4	2.9	-2.3	5	-7	1.5	-1.3	3	-3	8.1	-8.4
3	1	5.4	-5.9	-2	-2	8.7	7.4	7	-1	1.3	-1.2	3	-4	13.6	17.6	1	-4	6.9	6.5	6	0	2.0	-2.3	3	4	1.1	-1.0
-3	1	13.6	14.0	2	-2	3.3	-3.0	-7	2	2.2	2.3	-3	5	3.8	-3.3	1	5	0.8	0.7	-6	0	1.1	1.4	3	-4	1.4	1.0
3	2	6.7	-6.5	2	3	5.1	-5.0	7	-2	3.1	-2.8	3	-5	8.0	7.5	-1	5	1.1	-0.6	6	-1	6.6	-7.2	-3	5	2.7	-3.3
-3	2	2.7	2.2	-2	3	1.3	1.0	-7	3	5.4	5.2	-3	6	2.0	-1.7	-1	-5	3.7	-3.5	6	-2	3.2	-3.2	3	-5	8.9	8.9
3	3	5.4	-5.5	-2	-3	5.3	5.4	7	-3	3.6	-3.6	-3	-6	1.7	-1.3	-1	6	1.6	1.5	-6	3	1.2	-0.6	3	-6	2.9	1.9
-3	3	1.9	-1.7	2	-3	3.9	-3.4	-7	4	5.8	5.6	-4	0	1.8	-1.7	+1	-6	1.8	1.4	6	-4	3.8	-3.3	-4	0	5.4	6.7
3	4	1.4	-1.5	2	4	4.3	-4.6	7	-4	2.6	-2.2	-4	0	4.2	-3.7	2	0	14.3	-14.4	7	-1	1.0	-1.2	-4	1	9.2	9.1
-3	4	1.8	0.8	-2	4	1.2	1.0	8	-3	1.3	-1.2	-4	1	1.0	0.9	-2	0	19.9	20.0	7	-4	3.7	-3.7	-4	-1	1.7	1.6
-3	5	5.6	-5.2	2	-4	6.0	5.5	-8	4	1.7	1.4	4	-1	2.0	2.0	2	1	17.0	-16.6	8	-2	2.6	-2.7	4	-1	5.8	5.3
-3	6	1.9	-1.5	-2	5	7.1	-7.6	8	-4	2.3	-2.1	4	2	1.7	-1.8	-2	1	6.5	4.8					4	2	3.1	-2.9
4	0	3.2	-2.8	2	-5	1.6	1.0					-4	2	0.6	-0.6	-2	1	3.1	4.8	4				-4	2	3.7	3.3
4	1	1.3	-1.4	-2	6	5.0	-4.2	2				-4	-2	0.8	0.6	-2	-1	3.7	3.0	0	0	9.1	-8.0	-4	-2	1.2	1.1
-4	1	1.3	-1.5	2	-6	1.2	0.6	0	0	12.1	-10.7	4	-4	2.5	2.1	2	-1	12.8	12.5	0	1	6.4	-6.8	4	-2	12.9	13.1
4	2	2.1	-1.6	3	0	17.7	-17.1	0	1	34.9	-36.9	4	3	2.8	-3.0	2	2	12.9	-11.8	0	-1	2.8	2.5	-4	3	2.1	1.9
4	2	5.5	5.6	-3	0	2.8	1.9	0	-1	17.6	17.6	-4	3	1.3	-0.9	-2	2	0.7	-0.4	0	2	5.8	5.3	4	-3	2.9	-2.4
4	3	1.3	-1.4	3	1	5.8	-5.2	0	2	2.7	-1.1	-4	-3	1.1	-0.7	-2	-2	0.8	-1.0	0	-2	1.3	-0.1	-4	4	1.5	1.4
-4	3	3.1	2.5	-3	1	14.2	12.6	0	-2	1.1	-1.0	4	-3	2.0	1.5	2	-2	18.5	18.2	0	3	5.1	-5.0	4	-4	5.3	-4.7
-4	4	5.0	4.6	-3	-1	3.9	-3.7	0	3	4.8	4.4	-4	4	1.9	0.9	2	3	9.6	-9.4	0	-3	2.9	1.9	-4	5	2.2	-2.5
-4	5	0.9	0.4	3	-1	1.4	2.2	0	-3	5.1	4.5	4	-4	9.2	11.0	-2	3	12.1	11.1	0	4	5.7	-5.7	4	-5	4.3	4.3
-4	6	1.4	-1.3	3	2	1.6	-1.0	0	4	4.7	-4.5	-4	5	2.4	-2.2	-2	-3	1.9	1.9	0	-4	4.6	4.4	-4	6	1.3	-1.5
-4	7	1.8	1.8	-3	2	1.2	-0.3	0	-4	1.1	0.7	4	-5	9.0	9.8	2	-3	7.3	6.1	0	5	1.0	1.0	4	-6	1.6	1.3
5	1	1.0	-1.3	-3	-2	2.0	1.5	0	-5	4.1	-4.7	-4	6	2.4	-2.0	2	4	1.8	-1.6	0	-5	2.3	2.7	5	0	0.8	-1.1
-5	1	1.4	-0.8	3	-2	2.4	2.4	1	0	4.7	4.1	4	-6	0.8	0.2	-2	-4	0.7	-0.5	1	0	8.1	-7.1	-5	0	1.2	1.6
5	2	3.7	-3.7	3	3	4.5	-4.7	-1	0	22.4	22.3	-5	0	4.5	-4.5	2	-4	6.4	5.9	-1	0	1.3	0.9	-5	1	5.7	5.9
-5	2	3.2	3.7	-3	3	0.8	0.4	1	1	3.4	-3.0	-5	1	3.0	-2.2	-2	5	2.5	-3.1	1	1	7.1	-6.1	-5	-1	1.7	-1.9
-5	3	1.3	1.1	3	-3	9.0	-0.0	-1	1	3.1	3.2	5	2	1.1	-0.9	-2	6	1.6	1.2	-1	1	3.4	-3.3	5	-1	1.6	-1.8
-5	4	3.2	2.3	3	4	1.7	-1.8	-1	-1	13.9	13.2	-5	2	0.8	0.0	2	-6	0.8	-0.6	-1	-1	2.4	-1.4	-5	2	3.8	3.8
-5	5	4.8	5.4	-3	4	5.8	5.3	1	-1	20.5	20.1	-5	-2	1.2	1.8	3	0	2.4	-1.6	1	1	17.3	16.4	-5	-2	0.8	-0.6
-5	6	3.7	3.1	3	-4	1.8	1.8	1	2	15.3	14.3	5	-2	2.9	3.4	-3	0	11.0	11.5	1	2	5.7	-5.2	5	-2	3.7	3.3
-5	7	2.6	2.4	-3	5	2.8	-2.7	-1	2	12.4	12.5	-5	3	0.8	0.3	3	1	4.2	4.1	-1	2	1.1	0.6	-5	3	1.3	0.9
6	1	2.2	-2.2	3	-5	2.5	1.3	-1	-2	1.2	0.0	-5	-3	2.1	2.1	-3	1	5.1	5.2	-1	-2	4.0	-3.0	5	-3	1.2	0.3
-6	1	1.7	1.9	-3	6	2.7	-2.1	1	-2	5.8	-5.2	-5	4	4.8	4.9	-3	-1	5.0	4.9	1	-2	6.7	5.3	5	-4	4.3	-4.0
-6	2	7.1	7.1	-3	-6	1.0	-0.8	1	3	5.5	4.2	-5	-4	2.0	1.8	3	-1	5.0	4.2	1	3	15.8	-16.5	-5	5	4.0	-4.0
-6	3	6.4	5.2	4	0	6.1	-6.3	-1	3	14.6	14.2	-5	5	3.0	3.4	3	2	1.6	-1.5	-1	3	3.9	-3.5	5	-5	1.4	0.8
-6	4	1.7	1.2	-4	-0	8.3	-8.6	-1	-3	3.1	2.9	+5	-5	3.9	3.4	-3	2	4.0	-3.9	-1	-3	2.5	-2.6	-5	6	3.0	-3.3
-7	2	2.4	2.4	4	1	2.7	2.2	1	-3	5.0	4.9	-5	6	1.1	-0.9	-3	-2	2.6	2.3	1	-3	4.5	-3.9	5	-6	0.8	0.1
-7	3	3.6	3.4	-4	1	3.7	3.5	1	4	3.3	-3.4	5	-6	0.8	-0.2	3	-2	22.3	24.2	1	4	8.1	-8.1	5	-7	1.8	-1.7
-7	4	1.8	1.6	-4	-1	8.5	-9.5	-1	4	1.1	-1.6	5	-7	1.2	-0.6	3	3	2.7	-3.1	-1	4	5.9	-5.7	6	1	0.8	-0.6
-8	4	2.8	-2.4	4	-1	0.4	0.5	1	-4	8.4	8.5	6	-1	4.9	-4.7	-3	3	5.5	4.2	-1	-4	3.8	-3.5	6	-1	3.0	-2.8
				-4	2	3.4	2.8	-1	5	1.1	-0.4	6	-2	2.9	-2.6	3	-3	15.0	14.6	1	-4	3.3	3.5	6	-2	7.0	-6.2
	1			-4	-2	2.7	-2.4	-1	-5	0.7	-0.9	-6	3	1.6	-1.5	-3	4	3.7	2.7	1	5	0.7	-0.4	6	-3	8.2	-7.5
0	1	14.2	15.4	4	-2	6.7	7.2	-1	6	1.9	1.6	6	-3	4.1	2.7	-3	-4	1.5	-1.5	-1	5	0.9	0.8	6	-4	7.7	-7.4
0	-1	0.7	0.8	4	3	2.8	-2.8	1	-6	2.0	-1.9	-6	4	2.2	1.8	3	-4	8.9	9.8	-1	-5	1.9	-1.9	7	-4	1.2	-0.9
0	2	6.9	5.9	-4	3	2.0	1.7	2	0	21.4	-21.3	6	-4	5.4	4.1	-3	5	3.7	-3.8	1	-5	2.9	3.4	8	-2	2.4	-2.5
0	-2	3.7	-2.7	-4	-3	0.9	-0.1	-2	0	2.6	3.4	7	-1	1.9	-2.0	3	-5	5.7	5.6	-1	6	0.9	0.7	8	-3	1.9	-1.4
0	3	5.9	5.4	4	-3	1.1	-0.1	2	1	19.6	-18.8	7	-2	4.6	-4.5	-3	6	1.2	-0.8	1	-6	2.1	2.0				
0	-3	4.6	3.4	-4	4	6.0	5.3	-2	1	0.7	1.3	-7	3	1.1	1.0	3	-6	0.8	-0.3	2	0	8.7	-7.9	5			
0	4	2.6	-2.0	+4	-4	9.9	11.2	-2	-1	1.6	1.5	7	-3	2.9	-2.6	4	0	3.3	-3.3	-2	0	18.5	17.6	0	0	3.7	-2.8
0	-4	3.4	-3.1	-4	5	2.6	2.4	2	-1	3.9	3.5	-7	4	2.0	1.9	-4	0	10.1	11.6	2	1	3 5	3.3	0	1	6.5	-6.2
0	-5	2.5	-3.0	4	-5	11.5	13.4	2	2	2.5	-2.1	7	-4	2.3	-2.1	4	1	1.2	-0.3	-2	1	18.7	16.4	0	-1	9.1	9.6
1	0	15.3	-12.8	-4	6	5.2	-4.6	-2	2	7.1	6.0	8	-2	1.6	-1.7	-4	1	8.6	8.7	-2	-1	2.7	2.3	0	2	2.9	2.2
-1	0	34.2	34.7	4	-6	3.6	3.1	-2	-2	4.1	3.7	-8	4	2.1	2.5	-4	-1	5.0	5.7	2	-1	24.0	23.2	0	-2	1.1	-0.8
1	1	0.7	-0.6	4	-7	1.8	1.6	2	-2	16.6	-15.7					4	-1	5.7	-6.1	2	2	1.7	1.5	0	3	7.1	-6.7
-1	1	49.2	52.7	5	0	1.0	-0.9	2	3	5.0	-4.9	3				4	2	4.4	-4.1	-2	2	2.9	7.2	0	-3	3.1	2.6
-1	-1	7.7	-6.8	-5	0	2.0	-1.5	-2	3	15.3	15.1	0	0	10.2	8.6	-4	2	1.2	0.4	-2	-2	0.9	-0.7	0	4	8.1	-8.4
1	-1	8.9	-9.6	5	1	0.8	-0.8	-2	-3	2.0	2.4	0	1	13.6	-12.0	4	-2	5.5	5.9	2	-2	27.2	26.6	0	-4	8.1	8.8

TABLE 3. (continued)

0	6	2.5	-2.1	5	-6	3.4	2.8	-4	-2	0.6	-0.7	-3	2	3.8	-3.8	-2	4	3.7	-3.9	3	2	1.3	-1.2	5	-3	2.6	-2.0				
0	-5	4.4	4.2	6	0	1.9	2.0	4	-2	5.0	-4.6	3	-2	3.7	3.9	2	-4	1.7	-1.8	-3	2	2.5	-2.6	5	-4	1.0	0.8				
1	0	7.5	-6.7	6	1	0.8	-0.9	4	3	2.0	2.2	3	3	1.6	1.6	2	-5	1.8	1.4	3	-2	7.9	-8.1	5	-5	2.8	-2.8				
-1	0	13.0	-11.8	6	-1	1.5	-1.2	-4	3	3.4	3.3	-3	3	5.8	-6.1	3	1	3.7	-3.7	-3	3	1.7	-1.5	6	-2	2.7	-2.4				
1	1	4.6	-3.1	-6	2	1.3	1.4	4	-3	2.5	2.3	-3	-3	0.7	-0.7	-3	1	1.7	-1.7	3	-3	1.0	-0.7	6	-3	3.4	-3.0				
-1	1	17.0	-15.5	6	-2	5.8	-5.6	-4	4	2.1	1.7	3	-3	1.9	-1.3	-3	-1	0.8	1.3	-3	4	1.4	-1.7	6	-4	2.6	2.0				
-1	-1	1.3	0.6	-6	3	1.7	1.5	4	-4	4.2	-3.7	-3	4	5.0	-4.9	-3	-1	5.5	-5.3	3	-4	2.5	-2.5								
1	-1	10.0	9.4	7	-1	2.5	-2.8	4	-5	1.0	0.9	3	-4	4.9	-4.5	3	2	5.4	-5.3	3	-5	1.2	0.7		11						
1	2	1.3	0.3	7	-2	4.4	-4.2	4	-6	3.0	2.3	-3	5	0.9	-1.0	-3	2	1.2	-0.9	4	0	0.8	0.8	0	0	1.1	-1.0				
-1	-2	1.0	-0.9	8	-2	2.2	-2.1	5	0	2.6	-2.4	3	-6	1.5	1.3	-3	-2	0.8	-0.6	-4	1	1.3	-1.4	0	1	1.5	-1.5				
1	-2	1.2	-0.3	8	-3	2.0	-1.2	-5	0	3.5	4.1	3	-6	1.5	1.4	3	-2	4.2	-3.9	4	-1	4.2	-4.1	0	-1	1.6	-2.4				
1	3	6.1	-6.2					5	1	1.2	-1.0	4	0	6.0	-5.9	3	3	0.5	0.6	4	2	0.5	-0.1	0	2	2.0	-1.9				
-1	3	1.1	-0.6	6				-5	1	1.6	-1.8	-4	0	0.7	0.5	-3	3	4.3	-3.0	-4	2	1.9	-1.7	0	-2	3.4	-3.8				
-1	-3	2.8	2.5	0	0	6.2	-6.4	-5	-1	2.2	-2.8	4	1	5.3	-5.9	3	-3	1.0	0.3	4	-2	4.1	-4.2	0	-3	2.2	-2.5				
1	-3	3.3	-3.0	0	1	4.6	-4.1	5	-1	9.7	-10.8	-4	1	3.3	1.4	-3	4	4.2	-4.7	-4	3	1.1	-1.1	1	0	0.9	-0.6				
1	4	3.2	-3.3	0	-1	1.1	0.8	5	2	1.1	-1.0	4	-1	8.8	-8.6	3	-4	2.0	-2.0	4	-3	1.7	1.6	-1	0	0.7	-0.4				
-1	4	5.4	-5.4	0	2	2.0	-2.0	5	-2	7.3	-7.5	4	2	3.2	-2.9	-3	5	1.2	-1.7	-4	4	0.9	-1.8	1	1	3.8	-3.4				
-1	-4	4.1	4.0	0	-2	1.2	1.0	-5	3	1.2	1.0	-4	-2	1.0	0.1	-3	-5	0.9	-0.9	4	-4	1.4	0.8	-1	1	2.1	-2.1				
1	-4	5.5	5.3	0	3	6.9	-7.0	5	-3	2.1	1.0	4	-2	3.9	-3.4	4	0	2.8	2.9	4	-5	4.1	3.1	-1	-1	0.6	0.6				
1	5	0.7	-0.6	0	-3	3.7	2.8	-5	4	0.9	0.9	-4	3	0.8	-0.4	4	1	0.8	-0.8	5	1	0.9	-0.8	1	-1	1.7	-2.1				
1	-5	2.2	2.7	0	4	2.9	-2.7	5	-4	1.4	-1.5	4	-3	1.2	0.5	-4	-1	0.7	-0.4	5	-1	0.8	1.0	1	2	3.9	-3.9				
2	0	10.4	-10.4	0	-4	3.2	3.2	-5	5	0.7	-0.6	-4	4	0.7	-0.6	4	-1	5.3	-5.8	5	-3	1.6	1.5	-1	2	2.0	-1.9				
-2	0	1.0	-0.5	1	0	9.7	-9.2	5	-5	2.3	-1.7	4	-4	1.7	-2.1	4	2	1.2	-1.0	5	-4	1.1	-0.7	-1	-2	1.0	-0.9				
-2	1	4.6	-4.1	1	1	2.8	-2.3	5	-6	5.1	4.6	4	-6	1.0	0.7	-4	2	0.8	-1.2	5	-5	2.3	-1.8	1	-2	3.0	-2.9				
-2	-1	1.0	1.3	-1	1	6.4	-5.8	6	1	1.0	-1.5	5	0	1.9	1.9	4	-2	9.8	-10.5	6	-1	2.2	2.3	1	-3	1.5	-1.4				
2	-1	1.8	-0.5	-1	-1	4.2	4.3	6	-1	1.8	-1.7	-5	0	1.7	2.0	-4	3	0.7	-0.6	6	-2	1.2	-0.9	2	0	0.8	-0.1				
2	2	1.6	1.3	1	-1	2.3	2.3	-6	3	1.3	-1.4	5	1	1.3	-1.6	4	-3	2.4	-2.4	6	-3	2.2	-1.9	-2	0	0.8	0.8				
-2	2	4.0	3.4	1	2	3.6	-3.7	6	-3	3.1	2.4	-5	1	2.0	2.4	-4	4	2.9	-3.1	7	-2	1.8	-1.6	2	1	4.3	-4.2				
-2	-2	2.9	-3.5	-1	2	2.2	-1.6	7	-1	4.1	-3.9	5	-1	1.7	1.8	4	-4	2.4	-2.4	7	-3	5.6	-5.6	-2	1	0.5	-0.3				
+2	-2	4.8	3.8	-1	-2	4.3	4.9	7	-2	4.4	-4.2	+5	-2	2.1	-1.8	4	-6	2.9	3.2	7	-4	4.2	-3.9	-2	-1	0.9	1.0				
2	3	1.3	-0.8	1	-2	13.6	13.4	7	-3	1.4	-1.0	-5	3	0.7	-0.4	5	0	1.5	1.5					2	-1	1.6	-1.6				
-2	3	3.2	2.7	1	3	3.8	-3.7	+8	-4	2.3	1.2	5	-4	0.8	-1.3	-5	0	0.7	-0.7		10			2	2	2.9	-3.5				
-2	-3	1.9	-1.9	-1	3	4.0	-3.5					5	-5	3.4	2.8	5	1	3.2	-3.0	0	0	5.8	-7.0	-2	2	0.5	-0.5				
2	-3	3.9	3.0	-1	-3	7.1	8.2		7			6	0	3.5	-3.7	5	-1	0.8	0.2	0	1	2.9	-3.0	2	-2	1.5	-1.5				
-2	4	5.3	-4.4	-1	-8	10.4	9.5	0	0	4.4	-4.3	6	-1	2.0	-1.8	-5	2	0.6	0.8	0	-1	2.6	-3.2	2	-3	2.4	2.1				
2	-4	10.7	10.8	1	4	2.3	2.4	0	1	2.7	-2.3	-6	2	1.6	2.0	5	-2	4.6	-4.5	0	2	1.7	-1.9	2	-4	1.2	0.8				
-2	5	1.0	-0.5	-1	4	3.7	-3.8	0	-1	8.3	8.0	6	-2	3.0	-3.1	-5	3	0.5	0.7	0	-2	1.5	1.7	3	0	5.2	-5.4				
2	-5	8.5	7.7	-1	-4	4.4	4.6	0	2	3.6	-3.6	6	-4	1.2	0.8	5	-3	0.9	0.5	1	0	3.2	-3.4	3	1	0.9	0.6				
-2	6	1.2	1.1	1	-4	5.8	5.2	0	-2	9.3	8.8	7	-1	1.8	1.9	5	-5	2.5	-1.9	-1	0	2.6	-3.2	3	-1	2.7	3.1				
-3	0	1.0	0.7	-1	5	0.9	-0.7	0	3	8.1	-9.0	7	-3	1.8	1.3	5	-6	3.6	3.8	1	1	1.3	-0.8	3	-2	1.0	-1.0				
3	1	2.6	2.2	1	-5	1.0	-1.4	0	-3	1.0	-0.6	7	-4	5.5	4.7	-6	-1	1.1	1.0	-1	1	1.4	-1.2	3	-3	2.0	1.8				
-3	1	3.8	2.8	2	0	7.3	-6.1	0	4	2.0	-1.9	8	-3	0.2	-1.1	6	-2	1.4	-1.3	-1	-1	1.0	0.8	3	-4	0.7	0.4				
3	-1	4.5	-3.9	-2	0	0.8	-0.0	1	0	4.3	-3.9					6	-3	2.6	2.6	1	-1	2.5	-2.9	4	0	1.9	2.0				
3	2	3.6	-3.7	2	1	4.2	4.2	-1	0	2.6	-2.6		8			6	-4	2.2	2.1	1	2	1.4	-1.4	4	-1	1.2	1.6				
-3	2	8.0	7.7	-2	1	11.7	-11.1	1	1	2.5	2.4	0	0	3.1	2.9	7	-3	0.9	-0.8	-1	-2	3.0	2.8	4	-2	3.1	-2.7				
3	-2	5.1	-4.2	-2	-1	2.3	2.2	-1	1	1.4	-2.0	0	1	2.7	2.9					1	3	0.7	0.7	4	-3	1.2	-0.8				
3	3	2.8	-2.5	2	-1	9.9	-8.8	-1	-1	2.5	3.1	0	-1	6.9	7.0		9			1	-3	2.4	-2.4	4	-4	2.7	-2.2				
-3	3	8.3	7.9	2	2	2.5	-2.7	1	-1	7.7	7.5	0	-2	2.2	2.2	0	0	4.5	-4.5	1	-4	1.6	-1.5	5	-1	1.5	-1.4				
3	-3	5.5	-4.8	-2	2	9.3	-9.2	-1	-2	3.2	3.7	0	3	1.9	-2.0	0	1	1.2	1.0	2	0	5.5	-5.9	5	-2	1.1	-0.8				
-3	4	1.1	-0.7	-2	-2	1.4	0.7	-1	-2	14.4	14.8	0	-3	1.3	-1.0	0	-1	5.6	-6.1	-2	0	0.9	-1.2	5	-3	1.1	1.0				
3	-4	1.9	1.5	2	-2	7.9	7.3	1	3	3.1	-3.1	0	4	0.7	0.4	0	-2	1.0	-0.1	2	1	4.6	-5.0	5	-4	1.3	1.3				
-3	5	1.9	-2.1	2	3	2.8	-2.6	-1	3	6.0	-6.5	0	-4	1.6	1.3	0	3	1.0	1.1	-2	-1	0.9	1.0								
3	-5	5.4	5.2	-2	3	5.1	-4.4	-1	-3	1.6	1.1	1	0	3.2	-3.2	0	-3	2.2	2.2	2	-1	5.3	-6.0		12						
4	0	4.6	4.5	-2	-3	2.3	2.1	-1	-3	3.0	2.3	-1	0	0.8	0.6	0	4	3.4	3.7	2	2	2.6	-2.4	0	0	1.5	-1.6				
4	1	3.3	2.9	2	-3	7.8	6.6	1	4	1.6	1.6	1	1	1.6	1.4	0	-4	1.4	0.6	-2	-2	1.5	1.9	0	-1	3.1	-4.6				
-4	1	0.7	0.4	2	4	1.1	1.2	-1	4	3.8	-4.1	-1	1	1.2	-0.9	1	0	5.2	-5.1	2	-2	1.7	-1.4	0	-2	1.6	-2.6				
-4	-1	2.0	2.3	-2	4	4.1	-3.9	-1	-4	1.8	1.7	-1	-1	3.4	3.5	-1	0	1.2	0.5	-2	3	1.1	-1.4	1	0	0.5	-0.1				
4	-1	0.6	-0.5	2	-4	3.9	3.1	-1	-5	1.4	1.4	1	-1	5.6	-5.5	-1	1	2.5	2.7	2	-3	1.1	-0.9	-1	0	1.7	-2.2				
-4	2	2.2	1.6	2	-5	2.3	2.6	2	0	5.6	-5.0	-1	2	1.1	-0.8	1	-1	10.3	-11.4	2	-4	0.8	-0.9	1	1	0.7	-0.5				
-4	-2	2.1	1.9	2	-6	0.8	-0.5	-2	0	0.8	-0.2	-1	-2	1.2	-1.3	1	2	2.9	-2.9	3	0	2.5	-2.8	1	1	2.1	-2.3				
4	-2	2.2	1.7	-3	0	3.3	3.0	2	1	2.8	2.9	1	-2	7.2	-7.0	1	-2	4.8	-4.6	3	1	4.0	-3.9	2	0	2.5	-2.5				
4	3	2.4	2.1	3	1	5.5	5.6	-2	1	2.3	-2.5	1	3	0.8	0.6	1	3	0.7	-0.5	-3	1	0.6	-0.9	2	1	4.1	2.2				
-4	3	3.7	3.4	-3	1	1.5	-1.7	-2	-1	2.9	3.4	-1	3	4.7	-5.4	-1	3	1.2	-1.0	-3	-1	1.1	2.1	2	-1	0.8	-1.0				
4	-3	1.8	1.2	-3	-1	9.0	-8.4	2	-1	5.9	-5.6	-1	-3	0.9	-0.9	-1	-3	1.6	1.4	3	-1	3.2	-3.3	2	-2	0.7	-0.8				
4	-4	3.0	-2.7	3	2	1.0	-0.8	2	2	0.8	-0.6	1	-3	5.2	-5.3	-1	4	0.8	0.7	3	2	0.8	-0.7	3	0	1.7	2.2				
-4	5	1.3	-1.6	-3	2	1.4	-0.9	-2	2	3.3	-2.8	1	4	2.6	2.5	2	0	0.9	-0.5	-3	2	1.0	-1.0	3	-2	1.9	-1.8				
4	-5	1.5	1.0	3	-2	3.5	3.3	-2	-2	3.4	4.5	-1	4	3.8	-4.1	2	1	1.1	1.2	3	-2	2.0	-1.8	3	-3	2.4	-2.1				
5	0	2.6	-2.4	3	3	1.5	1.3	2	-2	0.8	0.4	-1	-4	2.3	2.7	2	-1	7.2	-7.2	-3	3	1.7	-2.1	4	-1	1.5	1.6				
-5	0	1.1	1.3	-3	3	2.7	3.0	-2	3	6.3	-5.9	1	-4	0.8	0.3	2	2	3.0	-2.8	3	-3	2.0	2.0	4	-3	1.4	-0.9				
5	1	1.3	-1.0	-3	-3	0.9	1.0	-2	-3	1.3	1.3	1	-5	1.4	1.2	-2	2	0.8	-0.6	3	-4	3.8	3.2								
-5	1	3.1	3.5	3	-4	6.4	-6.3	2	-3	8.6	-8.8	-2	0	1.4	-1.1	-2	-2	0.8	1.4	3	-5	2.6	2.5								
-5	-1	1.1	1.2	3	-5	0.8	-0.4	2	4	2.4	2.4	2	1	0.9	-0.5	2	-2	1.6	-0.7	4	0	2.6	2.4								
5	-1	12.9	-13.9	3	-6	0.7	0.5	-2	4	4.5	-4.8	-2	1	2.7	-2.4	-2	3	1.7	-1.8	4	1	1.3	2.4								
-5	2	4.2	4.4	4	0	7.9	-8.2	2	-4	6.4	-6.2	-2	-1	1.8	2.1	2	-3	1.2	1.3	4	-1	3.6	4.3								
5	-2	13.6	-16.1	-4	0	1.5	2.1	2	-5	1.0	1.1	2	-1	5.4	-5.2	-2	4	1.5	-1.7	-4	2	1.2	-2.2								
-5	3	3.2	3.2	4	1	2.5	-2.4	3	0	5.3	-5.2	2	2	5.0	-4.7	2	-4	2.1	-2.5	4	-2	0.8	0.9								
5	-3	6.2	-5.9	-4	1	0.9	-1.1	-3	0	2.1	1.8	-2	-2	0.8	0.7	2	-5	0.8	0.6	4	-3	1.2	1.1								
-5	4	1.0	1.1	-4	-1	1.0	1.3	3	1	3.9	-3.9	2	-2	6.7	-5.9	3	0	3.7	-3.9	4	-4	2.1	1.5								
5	-4	5.0	-4.6	4	-1	17.3	-17.6	-3	-1	0.8	0.6	-2	3	2.2	-2.0	3	1	1.4	-1.6	5	0	0.7	0.4								
-5	5	1.4	-1.2	4	2	1.6	-1.6	3	-1	5.2	-4.9	-2	-3	1.7	1.5	-3	1	2.2	-2.5	5	-1	1.5	1.6								
-5	6	0.7	-1.5	-4	2	1.2	-1.2	3	2	3.6	-3.6	-2	-3	5.2	-5.1	3	-1	11.3	-12.8	5	-2	3.2	-2.5								

TABLE 4. INTERATOMIC DISTANCES AND BOND ANGLES

N-N	1.319±0.006A	N'-N-O	120.2±0.4°
N-O	1.272±0.006	O-N-C(1)	121.4±0.4
N-C(1)	1.488±0.006	C(1)-N-N'	118.4±0.4
C(1)-C(2)	1.532±0.007	N-C(1)-C(2)	108.8±0.4
C(2)-C(3)	1.544±0.007	N-C(1)-C(6)	108.6±0.4
C(3)-C(4)	1.523±0.007	C(1)-C(2)-C(3)	107.9±0.5
C(4)-C(5)	1.517±0.007	C(2)-C(3)-C(4)	110.6±0.5
C(5)-C(6)	1.542±0.007	C(3)-C(4)-C(5)	110.9±0.5
C(6)-C(1)	1.530±0.007	C(4)-C(5)-C(6)	111.6±0.5
C(1)-H(1a)	0.99±0.07	C(5)-C(6)-C(1)	109.3±0.5
C(2)-H(2a)	1.11±0.07	N-C(1)-H(1a)	111±4
C(2)-H(2e)	1.02±0.07	H(2a)-C(2)-H(2e)	109±6
		C(1)-C(2)-H(2a)	101±4
		C(1)-C(2)-H(2e)	115±4

Table 5. Departure from the least-squares plane (1)

O	0.002 Å
N	0.006
C (1)	0.001

Kurita *et al.*[3] determined the configuration of the C(1)–H(1a), C(1)–C(2) and C(1)–C(6) bonds around the N–C(1) bond from ESR analysis of the irradiated single crystal. The results show that the plane through C(1), N, and O makes an angle of 40° against the plane through C(2), C(1) and N. The result agrees within 5° with the value calculated from the structure of the molecule.

The authors are grateful to Dr. Y. Kurita for kindly supplying the specimen and for valuable discussions.

3) Y. Kurita, M. Kashiwagi and H. Nakata, Preprints for the Symposium of Molecular Structure, (Nagoya, 1965), p. 165.