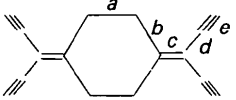


Table 2. Comparison of the mean bond lengths (Å) in the TCNQ anion, TCNQ<sup>0</sup> and TCNQ<sup>-</sup>


	a	b	c	d	b-c	c-d	α*	f†	k*
TCNQ (A)	1.364 (4)	1.425 (5)	1.417 (5)	1.421 (5)	0.008	-0.004	0.486	0.4	0.4
TCNQ (B)	1.355 (5)	1.437 (5)	1.395 (5)	1.432 (5)	0.042	-0.037	0.498	0.9	0.9
TCNQ <sup>0</sup> ‡	1.344 (3)	1.442 (4)	1.373 (3)	1.435 (4)	0.069	-0.062	0.476	0.0	0.0
TCNQ <sup>-</sup> §	1.363 (5)	1.422 (3)	1.420 (3)	1.417 (3)	0.002	0.003	0.500	1.0	1.0

\* Kistenmacher, Emge, Bloch &amp; Cowan (1982).

† Flandrois &amp; Chasseau (1977).

‡ Long, Sparks &amp; Trueblood (1965).

§ Ashwell &amp; Wallwork (1979).

are sensitive to the charge state of the TCNQ molecule (about 2200 and 1382 cm<sup>-1</sup>) a weak multiplet structure has been observed (Semkin, Vlasova, Kartenko, Prieu, Usov, Yartsev, Agroskin, Petrov, Abashev & Russkikh, 1989).

In the MT-TCNQ salt hydrogen bonds join every H atom of cation NH<sub>2</sub> groups with the N atom of anion cyanomethylene groups. In the (MT)<sub>2</sub>-(TCNQ)<sub>3</sub>·2H<sub>2</sub>O salt two H atoms of the cation take part in a hydrogen bond with the O atom of the water molecule. In the centrosymmetric A molecule all N atoms are involved in hydrogen bonds; N(13) with the cation and N(14) with the water molecule (see Table 3). In the non-centrosymmetric B molecule hydrogen bonds are found only for N(24) and N(26) atoms lying on one diagonal of the TCNQ molecule, atom N(26) being connected with an H atom of the water molecule, atom N(24) with an H atom of the cation. The crystallographic arrangement and the interaction between anion, cation and the water molecule, strongly suggest an important role for water molecules in the packing of TCNQ molecules, the water molecule being distorted: the HOH angle is 98 (3)°, two hydrogen bonds are

Table 3. Hydrogen-bond lengths (Å) and angles (°)

X—H...Y	X—H	X—Y	H...Y	∠X—H...Y	r <sub>H</sub> + r <sub>Y</sub> - 0.2*
N(2)—H(9)...N(24)	0.89 (3)	3.033 (5)	2.39 (3)	151 (3)	2.55
N(2)—H(10)...N(13)	0.90 (3)	2.999 (5)	2.10 (3)	180 (3)	2.55
O(43)—H(44)...N(14)	0.96 (3)	3.045 (5)	2.10 (3)	168 (3)	2.55
O(43)—H(45)...N(26)	0.83 (3)	3.067 (4)	2.32 (3)	150 (3)	2.55
N(3)—H(11)...O(43)	0.93 (3)	2.889 (4)	1.98 (3)	164 (3)	2.50
N(3)—H(12)...O(43)	0.93 (3)	2.889 (4)	2.11 (3)	153 (3)	2.50

\* Hamilton & Ibers (1968) criterion. Van der Waals radii: r<sub>H</sub> = 1.20, r<sub>N</sub> = 1.55, r<sub>O</sub> = 1.50 Å.

0.96 (3) for O(43)—H(44) and 0.83 (3) Å for O(43)—H(45).

Specific distribution of charge in the TCNQ molecules may reduce electron correlations (Semkin, Vlasova, Kartenko, Prieu, Usov, Yartsev, Agroskin, Petrov, Abashev & Russkikh, 1989) and this contributes to the high electrical conductivity of the salt. The observation of crystal water molecules in a 2/3 TCNQ salt and the unusual effects on structure and physical properties seems to be unique.

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## Structures of 3-Amino-4-nitro-1,2-dihydro-5-pyrazolone and its Acidic Ammonium Salt

BY PETER KLINDT MOGENSEN AND OLE SIMONSEN

Department of Chemistry, Odense University, DK-5230 Odense M, Denmark

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**Abstract.** C<sub>3</sub>H<sub>4</sub>N<sub>4</sub>O<sub>3</sub> (1), *M<sub>r</sub>* = 144.09, monoclinic, *P*2<sub>1</sub>/*n*, *a* = 5.277 (1), *b* = 8.110 (2), *c* = 12.335 (1) Å, β = 98.526 (8)°, *V* = 522.1 (4) Å<sup>3</sup>, *Z* = 4, *D<sub>m</sub>* = 1.83 (1), *D<sub>x</sub>* = 1.833 Mg m<sup>-3</sup>, λ(Mo *K*α) = 0.71073 Å, μ = 0.135 mm<sup>-1</sup>, *F*(000) = 296, *T* =

295 K, *R* = 0.038 for 942 unique observed reflections. Acidic ammonium salt of (1): NH<sub>4</sub><sup>+</sup>·C<sub>3</sub>H<sub>3</sub>N<sub>4</sub>O<sub>3</sub><sup>-</sup> (2), *M<sub>r</sub>* = 305.21, triclinic, *P*1̄, *a* = 6.998 (2), *b* = 7.203 (1), *c* = 12.116 (1) Å, α = 77.899 (7), β = 84.05 (1), γ = 85.75 (1)°, *V* = 593.1 (6) Å<sup>3</sup>, *Z* = 2, *D<sub>m</sub>*

$= 1.71$  (1),  $D_x = 1.710 \text{ Mg m}^{-3}$ ,  $\lambda(\text{Mo } K\alpha) = 0.71073 \text{ \AA}$ ,  $\mu = 0.11 \text{ mm}^{-1}$ ,  $F(000) = 288$ ,  $T = 295 \text{ K}$ ,  $R = 0.037$  for 2872 unique observed reflections. (1) is an —N—H acid with a nearly planar five-membered ring. The anion in (2) has a planar five-membered ring with a maximum deviation of  $0.004 \text{ \AA}$  from the least-squares plane. Delocalization of  $\pi$ -electron density from the  $\text{H}_2\text{N}-\text{C}-\text{NH}^*$ — part to the nitro group makes the  $\text{H}^*$  atom the acid proton. All the H atoms both in (1) and (2) participate in hydrogen bonds.

**Introduction.** The present investigation is part of a study of aliphatic nitro compounds, nitronate salts and tautomeric forms of these compounds. The title compounds (1) and (2) were first synthesized and described by Darapsky & Hillers (1915). They mention that 2-nitropropanenitrilehydrazide crystallizes with one mole of water, which can be lost by heating, giving an anhydrous form of 2-nitropropanenitrilehydrazide. Later it has been shown that this anhydride is equivalent with (1) (Ridi, Papini & Checchi, 1961). The present work has been undertaken to investigate the acid-base properties of (1) and (2) in relation to their structures.

**Experimental.** (1) and (2) were prepared from ethyl cyanoacetate as described by Darapsky & Hillers (1915). Pale yellow needles of (1) were obtained by recrystallization from methoxyethanol; red-brown prisms of (2) separated from an ammonia/water solution of (1) by evaporation at room temperature. The densities of (1) and (2) were determined by flotation in  $[\text{H}_2\text{O}, \text{KBr}(\text{aq})]$  and  $(\text{CHCl}_3, \text{CH}_2\text{BrCl})$ , respectively.

Crystals of dimensions  $0.13 \times 0.20 \times 0.23$  (1) and  $0.26 \times 0.23 \times 0.23 \text{ mm}$  (2) were investigated on an Enraf-Nonius CAD-4F diffractometer with graphite-monochromatized  $\text{Mo } K\alpha$  radiation. The lattice parameters were determined from the setting angles of 24 reflections (1) with  $8.14 < \theta < 12.15^\circ$  and 25 reflections (2) with  $9.93 < \theta < 14.18^\circ$ . The intensities of 1285 unique reflections were measured for (1) ( $h: 0 \rightarrow 7, k: 0 \rightarrow 11, l: -17 \rightarrow 17$ ) with  $2.0 < \theta < 30.0^\circ$ . 1104 reflections with  $I > 2.5\sigma(I)$  were used in the refinement process. The intensities of 3452 unique reflections were measured for (2) ( $h: 0 \rightarrow 9, k: -10 \rightarrow 10, l: -16 \rightarrow 16$ ) with  $2.0 < \theta < 30.0^\circ$ , 2872 with  $I > 2.5\sigma(I)$  were used in the refinement process.  $\omega/2\theta$  scan technique was used with scan angle  $\Delta\omega = (1.00 + 0.35\tan\theta)^\circ$  for (1).  $\Delta\omega = (1.20 + 0.35\tan\theta)^\circ$  for (2). Three reflections were used for orientation control every 100 reflections (1):  $1\bar{2}\bar{6}, \bar{2}22, 211$ ; (2):  $243, \bar{2}4\bar{1}, \bar{2}\bar{2}\bar{6}$ . An intensity stability check was made every third hour of exposure time using  $1\bar{2}\bar{6}$  (1) and  $\bar{2}0\bar{7}$  (2), standard intensity variations  $< 3.0\%$  of

Table 1. Fractional coordinates ( $\times 10^4$ ) and equivalent isotropic thermal parameters of non-H atoms with e.s.d.'s in parentheses

	$B_{\text{eq}} = \frac{1}{3} \sum_i \sum_j B_{ij}(a_i, a_j)$			
(1)	<i>x</i>	<i>y</i>	<i>z</i>	$B_{\text{eq}}(\text{\AA}^2)$
O(5)	-3298 (3)	809 (2)	3872 (1)	2.6 (1)
O(41)	952 (3)	2495 (2)	3146 (1)	3.3 (1)
O(42)	3261 (3)	4058 (2)	4344 (1)	2.8 (1)
N(1)	-2814 (3)	1310 (2)	5733 (1)	2.5 (1)
N(2)	-1095 (3)	2151 (2)	6515 (1)	2.4 (1)
N(3)	2305 (3)	4001 (2)	6526 (1)	2.4 (1)
N(4)	1487 (3)	3056 (2)	4093 (1)	2.0 (1)
C(3)	593 (4)	2984 (3)	6011 (1)	1.9 (1)
C(4)	63 (4)	2545 (3)	4881 (1)	1.8 (1)
C(5)	-2143 (4)	1478 (3)	4716 (1)	2.0 (1)
(2), N(00) is the N atom of the $\text{NH}_4^+$ ion				
O(5)	3514 (2)	4804 (2)	-1816 (1)	2.5 (1)
O(41)	3422 (2)	717 (2)	-1521 (1)	4.0 (1)
O(42)	2381 (3)	-807 (2)	160 (1)	4.3 (1)
N(00)	5213 (2)	1748 (2)	3173 (1)	2.7 (1)
N(1)	2508 (2)	5371 (2)	-35 (1)	2.3 (1)
N(2)	1865 (2)	4402 (2)	1057 (1)	2.3 (1)
N(3)	1443 (3)	1203 (2)	1900 (1)	3.3 (1)
N(4)	2800 (2)	718 (2)	-520 (1)	2.6 (1)
C(3)	1903 (2)	2597 (2)	989 (1)	2.1 (1)
C(4)	2572 (2)	2381 (2)	-134 (1)	2.0 (1)
C(5)	2940 (2)	4237 (2)	-785 (1)	1.9 (1)
O(95)	-4550 (2)	2323 (2)	5441 (1)	2.9 (1)
O(941)	-1158 (2)	1799 (2)	3906 (1)	3.1 (1)
O(942)	1577 (2)	2364 (2)	4439 (1)	3.1 (1)
N(91)	-3357 (2)	3311 (2)	6911 (1)	2.6 (1)
N(92)	-1561 (2)	3508 (2)	7267 (1)	2.3 (1)
N(93)	1643 (2)	3461 (2)	6494 (1)	2.7 (1)
N(94)	-213 (2)	2255 (2)	4616 (1)	2.1 (1)
C(93)	-200 (3)	3225 (2)	6450 (1)	1.9 (1)
C(94)	-1126 (2)	2664 (2)	5597 (1)	1.9 (1)
C(95)	-3178 (2)	2718 (2)	5907 (1)	2.1 (1)

mean value. The intensities were corrected for Lorentz and polarization effects. Absorption was ignored. The structures were solved by direct methods, and refined by full-matrix least-squares refinement of anisotropic non-H atoms. Positional H-atom parameters from  $\Delta\rho$  maps. Refinements of  $\text{H}(x, y, z)$  with fixed isotropic temperature factors ( $B = 3.0 \text{ \AA}^2$ ). The function  $\sum w(\Delta|F_i|)^2$  was minimized. The weighting scheme used was  $w = K/[\sigma(F)]^2$  with (1):  $K = 3.976$  and (2):  $K = 1.00$ . The refinement converged to final  $R = 0.039$ ,  $wR = 0.037$ ,  $S = 0.54$  and  $(\Delta/\sigma)_{\text{max}} = 0.02$  for (1) and to final  $R = 0.040$ ,  $wR = 0.038$ ,  $S = 0.42$  and  $(\Delta/\sigma)_{\text{max}} = 0.09$  for (2). Min., max. values in final  $\Delta\rho$  maps were  $-0.27, 0.23 \text{ e \AA}^{-3}$  for (1) and  $-0.28, 0.34 \text{ e \AA}^{-3}$  for (2). Scattering factors from *International Tables for X-ray Crystallography* (1974, Vol. IV, pp. 99, 149). Computer programs from *SHELX76* (Sheldrick, 1976), *XRAY76* (Stewart, Machin, Dickinson, Ammon, Heck & Flack, 1976) and *MULTAN80* (Main, Fiske, Hull, Lessinger, Germain, Declercq & Woolfson, 1980). The positional and thermal parameters are given in Table 1.\*

\* Lists of structure factors, anisotropic thermal parameters and H-atom parameters have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 53928 (24 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

**Discussion.** Bond distances and bond angles are shown in Fig. 1 with the atomic numbering scheme. Figs. 2 and 3 show projections of the crystal structures. The maximum deviations from a least-squares plane defined by the five pyrazolone ring atoms are small [(1), N(2):  $-0.029$ ; (2)-acid, N(92):  $-0.030$  and (2)-anion, C(5):  $-0.004$  Å]. Salt formation apparently

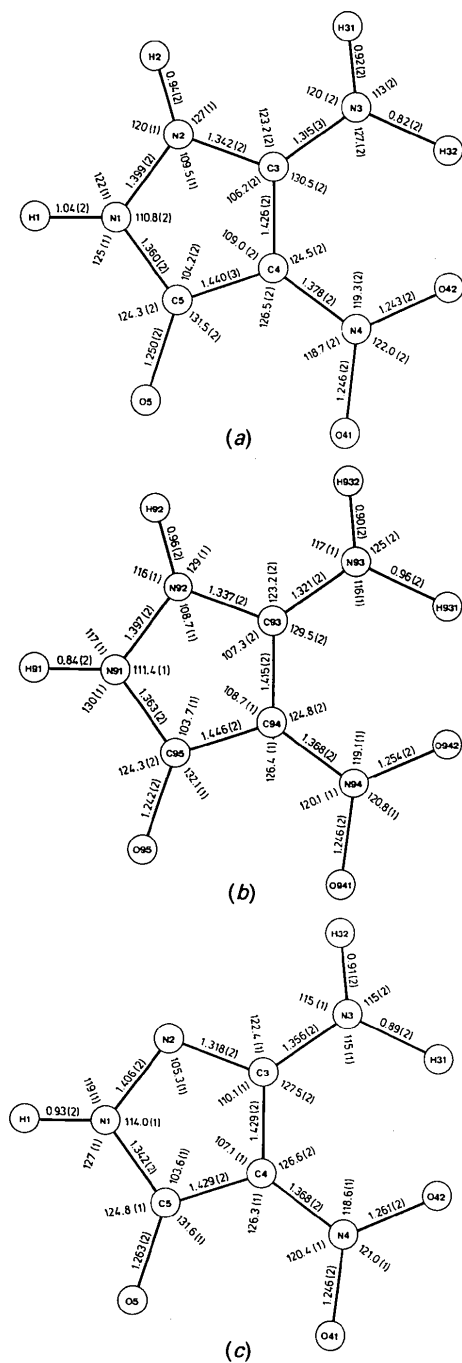


Fig. 1. Bond distances (Å), angles ( $^{\circ}$ ) and atomic numbering scheme for the compounds (a) (1), (b) (2) in acid form and (c) (2) as anion. E.s.d.'s are in parentheses.

promotes a more planar structure. The general trend in the torsion angles associated with the nitro group support this point of view [(1), O(41)—N(4)—C(4)—C(3):  $174.9$  (3), O(42)—N(4)—C(4)—C(5):  $175.9$  (3); (2)-acid, O(941)—N(94)—C(94)—C(93):  $-179.0$  (2), O(942)—N(94)—C(94)—C(95):  $176.1$  (2) and (2)-anion, O(41)—N(4)—C(4)—C(3):  $-179.2$  (2), O(45)—N(4)—C(4)—C(5):  $-178.9$  (2) $^{\circ}$ ].

The C—NO<sub>2</sub> bond distances in (1) and (2) are much shorter than a C—NO<sub>2</sub> single bond [ $1.481$  (3) Å in nitroacetamide] (Thorup, Dreier & Simonsen, 1981), but characteristic for a partial double bond. The C—NO<sub>2</sub> bond distance lies between the corresponding bonds in nitromalonamide,  $1.397$  (4) Å (Simonsen & Thorup, 1979) and the nitromalonamide anion,  $1.327$  (4) Å (Simonsen, 1981).

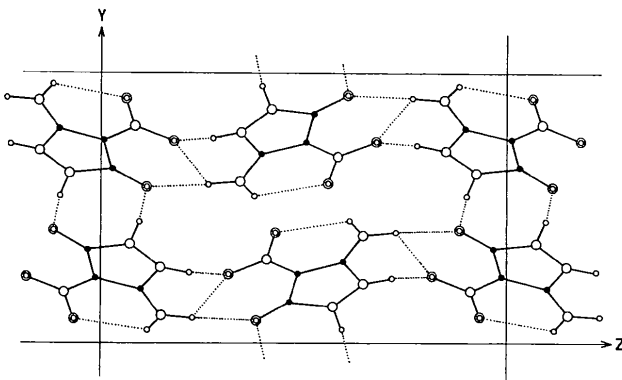


Fig. 2. The crystal structure of (1) projected down the *a* axis. Double circles: O atoms, large single circles: N atoms, black circles: C atoms, small circles: H atoms. The dotted lines represent the hydrogen-bonding system.

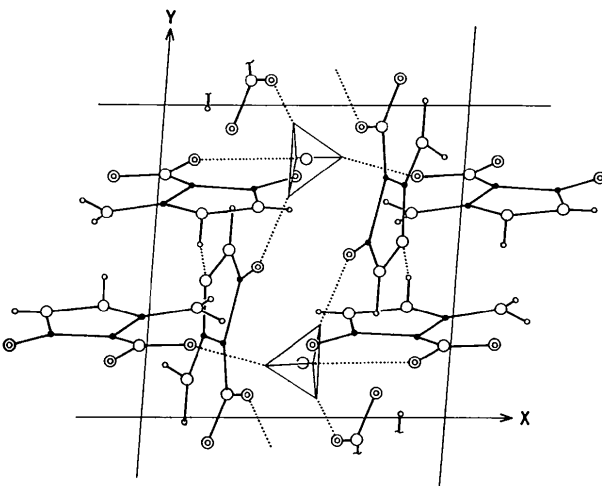
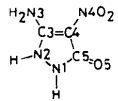
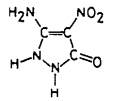
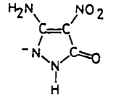
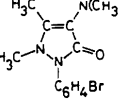
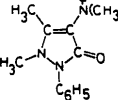
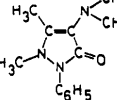


Fig. 3. The crystal structure of (2) projected down the *z* axis. Double circles: O atoms, large single circles: N atoms, black circles: C atoms, small circles: H atoms. Ammonium ions are symbolized by tetrahedra. The dotted lines represent hydrogen bonds. Only the strongest hydrogen bond (N—H...N) and those which originate from the NH<sub>4</sub><sup>+</sup> ion are shown.

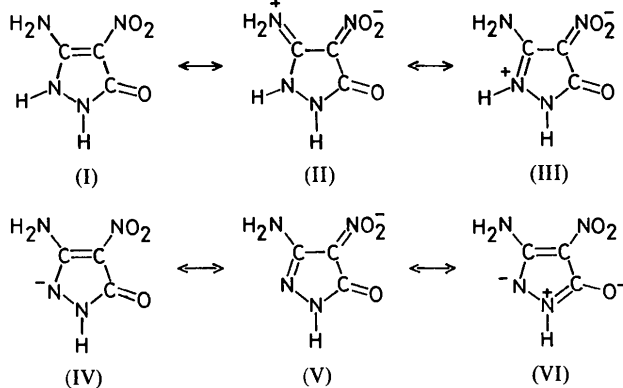
Table 2. Bond lengths (Å) in (1) and (2) compared with corresponding bonds in related compounds

Compound	Ref.	N(1)—N(2)	N(2)—C(3)	C(3)—C(4)	C(4)—C(5)	C(5)—N(1)	C(3)—N(3)	C(4)—N(4)	C(5)—O(5)
	(1)	1.399 (2)	1.342 (2)	1.426 (2)	1.440 (3)	1.360 (2)	1.315 (3)	1.378 (2)	1.250 (2)
	(2)	1.397 (2)	1.337 (2)	1.415 (2)	1.446 (2)	1.363 (2)	1.321 (2)	1.368 (2)	1.242 (2)
	(2)	1.406 (2)	1.318 (2)	1.429 (2)	1.429 (2)	1.342 (2)	1.356 (2)	1.368 (2)	1.263 (2)
	(3)	1.427 (8)	1.388 (7)	1.34 (1)	1.445 (8)	1.382 (8)		1.419 (8)	1.235 (9)
	(4)	1.394 (3)	1.360 (3)	1.380 (3)	1.428 (3)	1.400 (2)		1.478 (2)	1.232 (3)
	(5)	1.41 (1)	1.41 (1)	1.35 (1)	1.44 (1)	1.39 (1)		1.41 (1)	1.22 (1)

References: (3) Shimizu & Uno (1980), (4) Argay, Kálmán, Ribár, Vladimirov & Zirvanov-Stakić (1980) and (5) Krishna Murti, Vijayan & Brehm (1979).

In Table 2 a comparison is made between bond distances in (1), (2) and other pyrazolone compounds. The nitropyrazolones have longer C(5)—O(5) bonds and shorter N(2)—C(3) and N(1)—C(5) bonds than the other compounds.

Significant changes in bond distances associated with the formation of the ammonium salt are: an elongation of C(3)—C(4), C(5)—O(5) and especially C(3)—N(3), whereas shortenings are seen for N(2)—C(3) and C(5)—N(1). We propose the resonance formulae (I)→(III) and (IV)→(VI) in the scheme below to describe some of the principal features associated with the  $\pi$ -electron distribution in (1) and (2)-anion, respectively.



All the H atoms both in (1) and (2) participate in N—H $\cdots$ O hydrogen bonds, apart from one N—H $\cdots$ N bond in (2). The arrangement of two N—H $\cdots$ O centrosymmetrically related hydrogen bonds observed in the crystal structures of the N—H acid 4-oxypyrido[1,2-*a*]pyrimidin-1-ium-2-olate (Thorup & Simonsen, 1985) and the 3-nitro analogue (Simonsen, 1986) is also observed in the crystal structure of (1) where the N(1) $\cdots$ O(5,  $-x-1, -y, -z+1$ ) distance is 2.773 (4) Å. Not unexpectedly the acidic H atom is involved in the shortest hydrogen bond, both in (1) and (2). These hydrogen bonds are: (1), N(2) $\cdots$ O(41,  $x-\frac{1}{2}, -y+\frac{1}{2}, z+\frac{1}{2}$ ) = 2.737 (4) Å and (2), N(92) $\cdots$ N(2,  $-x, -y+1, -z+1$ ) = 2.751 (2) Å. The N(92) $\cdots$ N(2) distance seems to be rather short compared to the mean value of 2.98 (16) Å quoted for such a bond by Kuleshova & Zorkii (1981). This is probably caused by negative charge located on N(2). The ammonium ion in (2) stabilizes the crystal structure by knitting the acid and the anion together through hydrogen bonds in the range 2.781 (2)–3.100 (2) Å (Fig. 3).

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## Structure of Hexamethylenetetratellurafulvalene Diiodide

BY NING-HAI HU AND ZHONG-SHENG JIN

*Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, Changchun, People's Republic of China*

AND ZHU-SHI LI

*Department of Chemistry, Yanbian University, Yanji, Jilin, People's Republic of China*

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**Abstract.** C<sub>12</sub>H<sub>12</sub>I<sub>2</sub>Te<sub>4</sub>,  $M_r = 920.44$ , monoclinic,  $P2_1/n$ ,  $a = 10.942$  (2),  $b = 14.924$  (2),  $c = 11.415$  (2) Å,  $\beta = 104.32$  (1)°,  $V = 1806.0$  (5) Å<sup>3</sup>,  $Z = 4$ ,  $D_x = 3.38$  g cm<sup>-3</sup>,  $\lambda(\text{Mo } K\alpha) = 0.71069$  Å,  $\mu = 100.7$  cm<sup>-1</sup>,  $F(000) = 1592$ ,  $T = 294$  K,  $R = 0.033$  for 1828 observed reflections. One of the Te atoms is bonded to the two I atoms, which are on either side of the molecular plane. The Te—I distances are 2.963 (1) and 2.961 (1) Å, which means oxidation at the Te atom instead of at the C=C bonds.

**Introduction.** In the exploration of organic conductors much attention has been paid to hexamethylenetetratellurafulvalene (HMTTeF) as an interesting electron donor (Wudl & Aharon-Shalom, 1982). The larger van der Waals radius and lower electronegativity of the tellurium compared to those of sulfur and selenium are expected to enhance the metallic character of the organic radical salts. The electrical conductivity and the structures for several charge-transfer complexes of HMTTeF with TCNQF<sub>4</sub> (Li, Matsuzaki, Onomichi, Sano & Saito, 1986) and PF<sub>6</sub> (Kikuchi, Yakushi, Kuroda, Ikemoto, Kobayashi, Honda, Katayama & Tanaka, 1985; Li, Matsuzaki, Kato, Kobayashi, Kobayashi & Sano, 1986) have been studied. The plate-like crystals of

HMTTeF.X<sub>n</sub> (X = Cl, Br and I;  $n < 1$ ) have been found to show metallic character in the range from 100 to 300 K and to remain fairly conductive at 5 K (Matsuzaki, Li & Sano, 1986). It is well known that electrical conductivity is dependent on the crystal structure for organic radical salts. We report here the structure of a rhombic iodine derivative of HMTTeF and try to understand its conduction behaviour from a structural point of view.

**Experimental.** Single crystals of (HMTTeF)I<sub>2</sub> were prepared from a mixture of HMTTeF (10 mg) dissolved in CS<sub>2</sub> (20 ml) and (*n*-Bu)<sub>4</sub>N.I<sub>3</sub> (70 mg) in CH<sub>3</sub>CN (20 ml) by an electrochemical method. The black rhombic crystals were obtained after passing a current of 0.5–1.0 μA through the mixture, under a nitrogen atmosphere, for one week. The conductivity value measured for a compressed pellet of (HMTTeF)I<sub>2</sub> powder was  $4.1 \times 10^{-3}$  S m<sup>-1</sup> at room temperature.

Crystal 0.2 × 0.2 × 0.2 mm. Nicolet R3m/E four-circle diffractometer, graphite-monochromated Mo Kα ( $\lambda = 0.71069$  Å) radiation; cell parameters by a least-squares fit for 25 centred reflections ( $10 < 2\theta < 26^\circ$ ); reflection data  $2\theta_{\text{max}} = 45^\circ$ ,  $\omega$ -scan mode, scan speed 4.88° min<sup>-1</sup>, scan width 1.2°. Intensity