Table 2. *Fractional atomic coordinates and equivalent isotropic thermal parameters* (Å^2) *for compound* (2*a*)

$$U_{eq} = \tfrac{1}{3}\Sigma_i\Sigma_j U_{ij}a_i^*a_j^*\mathbf{a}_i.\mathbf{a}_j.$$

	x	y	z	U_{eq}
O13	0.26412 (3)	0.0909 (1)	0.10907 (6)	0.0527 (3)
O15	0.35304 (4)	0.0766 (2)	0.18216 (6)	0.0665 (4)
O17	0.43736 (4)	0.7242 (1)	0.18797 (6)	0.0536 (3)
O19	0.35191 (4)	0.6521 (2)	0.09910 (7)	0.0693 (4)
O27	0.46132 (4)	0.1365 (2)	0.30774 (7)	0.0709 (5)
N1	0.38070 (4)	0.3531 (2)	0.30264 (6)	0.0388 (4)
N2	0.36267 (4)	0.4391 (2)	0.22953 (6)	0.0375 (4)
C3	0.40191 (5)	0.5237 (2)	0.23404 (8)	0.0373 (5)
C4	0.45400 (5)	0.5009 (2)	0.31541 (8)	0.0459 (6)
C5	0.43856 (5)	0.3773 (2)	0.36341 (8)	0.0370 (6)
C6	0.34428 (4)	0.2616 (2)	0.31621 (7)	0.0337 (5)
C7	0.30666 (5)	0.1552 (2)	0.25318 (8)	0.0370 (5)
C8	0.26743 (5)	0.0809 (2)	0.26480 (9)	0.0465 (5)
C9	0.26693 (5)	0.1032 (2)	0.33853 (9)	0.0525 (6)
C10	0.30591 (5)	0.2007 (2)	0.40194 (8)	0.0504 (6)
C11	0.34394 (5)	0.2812 (2)	0.39050 (8)	0.0416 (5)
C12	0.31148 (5)	0.1072 (2)	0.17946 (8)	0.0440 (5)
C14	0.26550 (6)	0.0243 (3)	0.03675 (9)	0.0675 (6)
C16	0.39280 (5)	0.6370 (2)	0.16533 (8)	0.0435 (5)
C18	0.43452 (6)	0.8489 (2)	0.1294 (1)	0.0627 (6)
C20	0.46699 (5)	0.2073 (2)	0.38216 (8)	0.0437 (5)
C21	0.52604 (5)	0.2218 (2)	0.44692 (9)	0.0464 (6)
C22	0.56362 (6)	0.2597 (2)	0.4238 (1)	0.0607 (6)
C23	0.61709 (6)	0.2762 (3)	0.4840 (1)	0.0798 (9)
C24	0.63428 (7)	0.2512 (2)	0.5677 (1)	0.0835 (10)
C25	0.59808 (7)	0.2118 (3)	0.5919 (1)	0.0771 (9)
C26	0.54353 (6)	0.1976 (2)	0.5311 (1)	0.0580 (7)

Table 3. *Comparison of selected bond distances* (Å) *and angles* (°) *in compounds* (1) *and* (2*a*)

	(1)	(2*a*)
O13—C12	1.332 (1)	1.330 (1)
O13—C14	1.446 (2)	1.452 (2)
O15—C12	1.203 (1)	1.204 (2)
O17—C16	1.326 (1)	1.333 (1)
O17—C18	1.449 (2)	1.438 (2)
O19—C16	1.205 (1)	1.197 (1)
O27—C20	1.206 (2)	1.411 (2)
N1—N2	1.354 (1)	1.356 (1)
N1—C5	1.467 (1)	1.480 (1)
N1—C6	1.388 (1)	1.406 (2)
N2—C3	1.286 (1)	1.288 (2)
C3—C4	1.497 (2)	1.501 (1)
C3—C16	1.465 (1)	1.465 (2)
C4—C5	1.541 (1)	1.534 (2)
C5—C20	1.530 (1)	1.533 (2)
C7—C12	1.488 (1)	1.482 (2)
C20—C21	1.489 (2)	1.512 (1)
C12—O13—C14	115.6 (1)	115.6 (1)
C16—O17—C18	116.2 (1)	116.7 (1)
C5—N1—C6	125.3 (1)	126.7 (1)
N2—N1—C6	121.4 (1)	119.6 (1)
N2—N1—C5	113.1 (1)	113.6 (1)
N1—N2—C3	108.7 (1)	108.6 (1)
N2—C3—C16	122.2 (1)	120.2 (1)
N2—C3—C4	114.3 (1)	114.0 (1)
C4—C3—C16	123.4 (1)	125.7 (1)
C3—C4—C5	101.3 (1)	102.3 (1)
N1—C5—C4	102.0 (1)	101.5 (1)
C4—C5—C20	110.6 (1)	114.7 (1)
N1—C5—C20	110.9 (1)	109.1 (1)
O15—C12—C7	124.3 (1)	124.9 (1)
O13—C12—C7	111.6 (1)	112.7 (1)
O13—C12—O15	123.8 (1)	122.3 (1)
O19—C16—C3	122.5 (2)	126.2 (2)
O17—C16—C3	113.2 (1)	109.4 (1)
O17—C16—O19	124.3 (1)	124.4 (2)
O27—C20—C5	119.8 (1)	110.6 (1)
C5—C20—C21	118.4 (1)	111.9 (1)
O27—C20—C21	121.8 (1)	108.8 (1)

For both compounds, data collection, cell refinement and data reduction were performed using *SDP* (Frenz, 1983). Both structures were solved using *MULTAN*11/82 (Main, Fiske, Hull, Lessinger, Germain, Declercq & Woolfson, 1982) and refined with *SDP*. Molecular graphics were prepared using *ORTEP*II (Johnson, 1976); the material for publication was produced using *PARST* (Nardelli, 1983).

We thank the Servizio Italiano Diffusione Dati Cristallografici del CNR, Parma, for the access to the Cambridge Structural Database. Thanks are due to Professor L. Garanti for supplying the crystals.

Lists of structure factors, anisotropic thermal parameters, H-atom coordinates and complete geometry have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 71465 (44 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England. [CIF reference: NA1050]

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Acta Cryst. (1994). **C50**, 91-93

N-Methylnitrilotriacetamide, $C_7H_{14}N_4O_3$

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(*Received* 4 *March* 1993; *accepted* 6 *October* 1993)

Abstract

The title compound is one of the possible chelating ligands used in our study of metal complexes with the various derivatives of amidated nitrilotriacetic acid. The molecules form a three-dimensional hydrogen-bonding

network utilizing the O and NH_2 atoms from the amide groups. The lengths of the hydrogen bonds vary from 2.81 to 2.98 Å.

Comment

As part of our program to design and study novel amide chelating ligands, we were interested in asymmetrical alkylated derivatives of nitrilotriacetamide. Our interest arose, not only from the potentially greater solubility of the ligands in organic solvents, but also from the increased electron donation of the methyl group to the amides. We have reported previously the divergent synthesis of nitrilotriacetamide and its *N*-methyl, *N*,*N'*-dimethyl, and *N*,*N'*,*N''*-trimethyl derivatives from nitrilotriacetic acid *via* transamidation and cyclization to the dioxopiperazineacetamide, followed by nucleophilic ring opening with primary amines (Smith, Cramer, Sucheck & Skrzypczak-Jankun, 1992). However, only the crystal structure of the symmetrical and unalkylated nitrilotriacetamide was reported there. We have also reported the first isolable crystalline complex of nitrilotriacetamide with a metal and the first example of both a completely acyclic ligand and a 2:1 ligand:metal ratio in a ten-coordinate lead complex (Smith, Sucheck & Pinkerton, 1992). This paper describes the crystal structure of the mono-*N*-methylnitrilotriacetamide and evaluates the effects of the hydrogen bonding on the structure and the crystal morphology.

The crystal structure revealed that the molecule adopts a conformation which allows it to utilize all potential donors and acceptors in the hydrogen-bonding interactions. Two molecules related by the center of symmetry form a dimer through two hydrogen bonds N5—H51···O4$^{\rm i}$. These dimers are further linked by another hydrogen bond, N5—H52···O8$^{\rm ii}$; these two bonds are approximately parallel to the *yz* plane. The hydrogen bonds N9—H92···O8$^{\rm ii}$ and N13—H131···O8$^{\rm ii}$ make a chain of molecules along the *y* direction, while the hydrogen bond N9—H91···O12 between identical molecules shifted along **a** is responsible for joining the layers of molecules together. This type of interaction explains the shape of crystals that grow as plates with the smallest dimension corresponding to the shortest parameter of the unit cell. Comparison of the observed bond lengths and the tabulated values (Allen *et al.*, 1987) indicates that some bonds might be shortened (N1—C2, N9—C7) and some elongated (C11—O12, C7—O8); the changes are within 2σ of the mean value, however, and could be related to the packing of molecules and the hydrogen bonds involved (Table 3, Fig. 2).

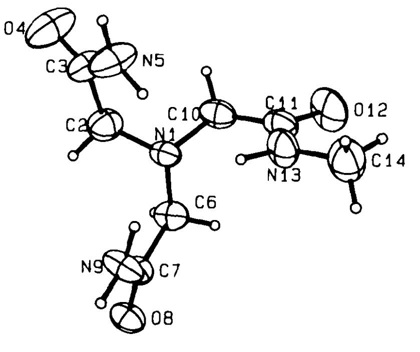

Fig. 1. An *ORTEP* drawing of the molecule with 50% probability thermal ellipsoids.

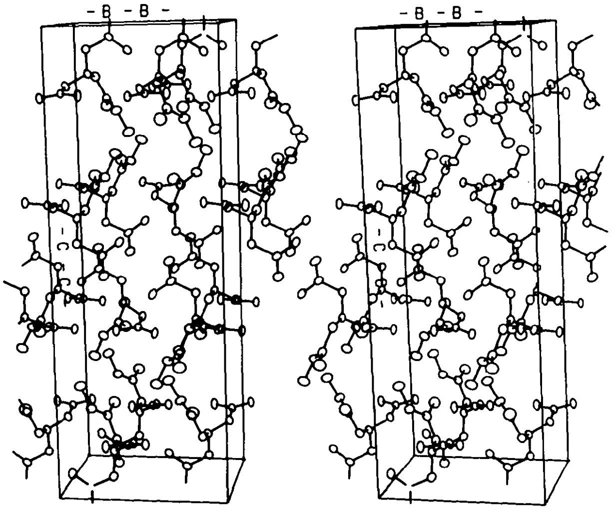

Fig. 2. Packing diagram of the unit cell.

Experimental

Crystal data

$C_7H_{14}N_4O_3$
M_r = 202.21
Orthorhombic
Pbca
a = 8.107 (2) Å
b = 9.757 (2) Å
c = 26.891 (6) Å
V = 2127 (1) Å^3
Z = 8
D_x = 1.26 Mg m^{-3}

Mo $K\alpha$ radiation
λ = 0.71073 Å
Cell parameters from 20 reflections
θ = 9–15°
μ = 0.093 mm^{-1}
T = 294 K
Plate
0.40 × 0.11 × 0.05 mm
Transparent

Data collection

Enraf–Nonius CAD-4 diffractometer
$\theta/2\theta$ scans
Absorption correction: empirical
$T_{\min}$ = 0.745, $T_{\max}$ = 0.998
2423 measured reflections
2423 independent reflections

1170 observed reflections [$I > 3.0\sigma(I)$]
$\theta_{\max}$ = 25.97°
$h = 0 \rightarrow 10$
$k = 0 \rightarrow 12$
$l = 0 \rightarrow 33$
3 standard reflections
frequency: 60 min
intensity variation: −0.61%

Refinement

Refinement on F
$R = 0.055$
$wR = 0.073$
$S = 1.710$
1170 reflections
128 parameters
H atoms riding
$w = 4F_o^2/[\sigma(F_o^2)+0.0036F_o^4]$
$(\Delta/\sigma)_{\max} = 0.001$
$\Delta\rho_{\max} = 0.23$ e Å^{-3}
$\Delta\rho_{\min} = -0.29$ e Å^{-3}
Extinction correction: isotropic (Zachariasen, 1963)
Extinction coefficient: 0.46×10^{-6}
Atomic scattering factors from *International Tables for X-ray Crystallography* (1974, Vol. IV)

Data collection: CAD-4 (Enraf–Nonius, 1977). Backgrounds were obtained from analysis of the scan profile (Blessing, Coppens & Becker, 1974). Cell refinement: CAD-4. Data reduction: *MolEN PROCESS* (Fair, 1990). Program(s) used to solve structure: direct-methods *MULTAN* (Main *et al.*, 1980). Program(s) used to refine structure: *MolEN LSFM*. Molecular graphics: *ORTEP* (Johnson, 1976). Software used to prepare material for publication: *MolEN CIF*.

Table 1. *Fractional atomic coordinates and equivalent isotropic thermal parameters* (Å^2)

$$U_{\rm eq} = \tfrac{1}{3}\Sigma_i\Sigma_j U_{ij}a_i^*a_j^*\mathbf{a}_i.\mathbf{a}_j.$$

	x	y	z	$U_{\rm eq}$
N1	−0.0083 (3)	−0.1644 (2)	0.60548 (9)	0.0376 (6)
C2	0.0293 (4)	−0.1646 (3)	0.5528 (1)	0.0514 (9)
C3	0.0104 (4)	−0.3028 (3)	0.5274 (1)	0.0553 (9)
O4	−0.0159 (4)	−0.3072 (2)	0.48279 (8)	0.0855 (9)
N5	0.0232 (4)	−0.4114 (3)	0.5562 (1)	0.0663 (9)
C6	0.0529 (4)	−0.0408 (3)	0.6303 (1)	0.0429 (8)
C7	0.2341 (3)	−0.0419 (2)	0.6429 (1)	0.0337 (6)
O8	0.3040 (3)	0.0688 (2)	0.65097 (8)	0.0454 (5)
N9	0.3109 (3)	−0.1594 (2)	0.6451 (1)	0.0481 (7)
C10	−0.1875 (4)	−0.1778 (3)	0.6141 (1)	0.0485 (8)
C11	−0.2315 (3)	−0.2360 (3)	0.6640 (1)	0.0460 (8)
O12	−0.3698 (3)	−0.2140 (3)	0.6814 (1)	0.0731 (8)
N13	−0.1217 (3)	−0.3136 (3)	0.68684 (9)	0.0517 (7)
C14	−0.1533 (4)	−0.3783 (4)	0.7344 (1)	0.071 (1)

Table 2. *Bond lengths* (Å) *and angles* (°)

N1—C2	1.448 (4)	C7—O8	1.239 (3)
N1—C6	1.466 (3)	C7—N9	1.307 (3)
N1—C10	1.477 (4)	C10—C11	1.499 (5)
C2—C3	1.520 (4)	C11—O12	1.234 (4)
C3—O4	1.219 (4)	C11—N13	1.321 (4)
C3—N5	1.318 (4)	N13—C14	1.450 (4)
C6—C7	1.507 (4)		
C2—N1—C6	112.0 (2)	C6—C7—O8	118.6 (2)
C2—N1—C10	111.1 (2)	C6—C7—N9	118.8 (2)
C6—N1—C10	109.6 (2)	O8—C7—N9	122.6 (3)
N1—C2—C3	114.8 (2)	N1—C10—C11	114.1 (2)
C2—C3—O4	119.5 (3)	C10—C11—O12	119.4 (3)
C2—C3—N5	116.1 (3)	C10—C11—N13	118.2 (3)
O4—C3—N5	124.4 (3)	O12—C11—N13	122.3 (3)
N1—C6—C7	115.3 (2)	C11—N13—C14	122.8 (3)

Table 3. *Hydrogen-bond geometry* (Å, °)

D	*A*	*D*···*A*	*D*—H···*A*
N5	O4$^{\rm i}$	2.940 (4)	173.0 (2)
N5	O8$^{\rm ii}$	2.914 (4)	149.4 (2)
N9	O8$^{\rm ii}$	2.815 (3)	155.3 (2)
N13	O8$^{\rm ii}$	2.980 (3)	157.0 (2)
N9	O12$^{\rm iii}$	2.817 (4)	161.9 (2)

Symmetry codes: (i) $-x, -y-1, -z+1$; (ii) $-x+\frac{1}{2}, y-\frac{1}{2}, z$; (iii) $1+x, y, z$.

This work was supported in part by a grant to DAS from the University of Toledo Undergraduate Summer Research Participation program. We thank the College of Arts and Sciences of the University of Toledo for generous financial support of the X-ray diffraction facility.

Lists of structure factors, anisotropic thermal parameters, H-atom coordinates and least-squares-planes data have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 71472 (32 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England. [CIF reference: HH1070]

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Acta Cryst. (1994). C**50**, 93–95

2,2,6,6-Tetramethylpiperidinium Thiocyanate, $2C_9H_{20}N^+.2SCN^-$

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(*Received* 31 *March* 1993; *accepted* 17 *June* 1993)

Abstract

The synthesis and crystal structure of bis(2,2,6,6-tetramethylpiperidinium) dithiocyanate are reported. Geometric parameters for the component species are typical.