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N,N-Bis(2-oxopyrrolidin-1-ylmethyl)glycine

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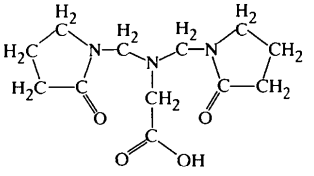
Abstract

The molecular structure of the title compound, $C_{12}H_{19}N_3O_4$, shows that the three moieties attached to the central, neutral N atom are each nearly planar. The molecules are linked by O—H···O hydrogen bonds between the COOH group and one of the pyrrolidone groups of an adjacent molecule, and C—H···O hydrogen bonds between one methylene of another pyrrolidone group and the C=O of the same pyrrolidone group of an adjacent molecule.

Comment

Derivatives of 2-pyrrolidone and polyaminocarboxylates have been used widely to form transition metal or lanthanide complexes (Doyle *et al.*, 1993, 1995; Finnen & Pinkerton, 1997; Goodgame *et al.*, 1996) with interesting structures and properties. Recently we have synthesized the title compound, (I), which contains 2-pyrrolidone as well as aminocarboxylate groups, and found that some lanthanide complexes with this ligand exhibited interesting properties such as photochromicity (Jin *et al.*, unpublished work). In the process of investigating the structures and properties of these complexes, we have determined the structure of the title compound and report the result here.



(I)

The two pyrrolidone groups are distinguished from each other by their different conformations. The pyrrolidone group, defined by N2, O3, C4–C7, has an envelope form with C6 at the flap position displaced from the plane of the other five non-H atoms by 0.352 (3) Å, which is similar to the value (0.31 Å) found in the 2-pyrrolidone unit of *N*-[2-(*N*,*N*-diisopropylamino)-ethyl]-2-oxo-1-pyrrolidineacetamide sulfate (PMRCT) (Bandoli *et al.*, 1987). The other pyrrolidone group is nearly planar [maximum deviation 0.035 (2) Å], which is better than is found in the planar 2-pyrrolidone group in the monoclinic phase of (2-oxo-1-pyrrolidinyl)-acetamide (Admiraal *et al.*, 1982). The dihedral angle between planes N2, O3, C4–C7 and C3, and N3, O4, C9–C12 and C8 is 65.57 (6)°. The planar carboxylmethyl group is approximately parallel to the plane defined by N2, O3, C3, C4–C7 with a dihedral angle of 10.7 (1)°, while it is slanted to the plane defined by N3, O4, C8, C9–C12 with a dihedral angle of 57.61 (8)°.

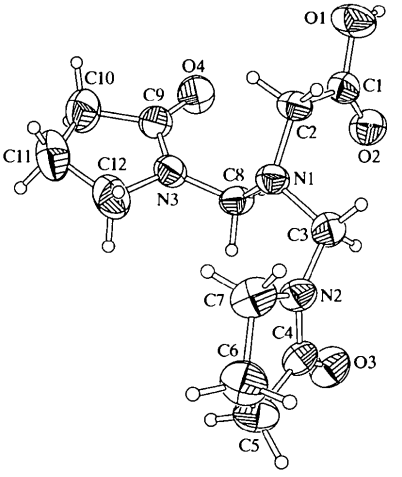


Fig. 1. The molecular structure of (I) showing 50% probability displacement ellipsoids.

The bond lengths and angles in the molecule are in good agreement with corresponding values previously reported (2-oxo-1-pyrrolidinyl)acetamide (Admiraal *et al.*, 1982; Louër *et al.*, 1995), PMRCT (Bandoli *et al.*, 1987), *N*,*N*,*N*′,*N*″,*N*‴,*N*‴′-triethylenetetraminehexacetic acid (TTHA) (Finnen & Pinkerton, 1997) and *N*-(hydroxyethyl)ethylenediaminetriacetic acid (H3heedta) (Kettmann *et al.*, 1993). In the molecule, the nine C—N bonds can be divided into two groups, of which seven C—N bonds lengths, falling in the range 1.440 (2)–1.459 (2) Å, characterize C—N single bonds showing that the central N1 atom is a neutral one (Billing *et al.*, 1991; Finnen & Pinkerton, 1997). The two shorter C—N bonds, C4—N2 [1.353 (2) Å] and C9—N3 [1.339 (2) Å], together with the coplanarity of the non-H atoms around N2 or N3 atoms, indicate the conjugation of the lone pair of electrons of N with the adjacent C=O group (Admiraal *et al.*, 1982; Billing *et al.*, 1991). The seven C—C single bond distances from 1.494 (3) to 1.519 (3) Å, and the two C=O distances of the 2-pyrrolidone group, 1.220 (2) and 1.235 (2) Å, are

all comparable with those bond distances found in the literature (Admiraal *et al.*, 1982; Bandoli *et al.*, 1987; Billing *et al.*, 1991; Finnen & Pinkerton, 1997; Louër *et al.*, 1995; Müller *et al.*, 1996). The two dissymmetric C—O bonds [1.201 (2) and 1.319 (2) Å] of the carboxyl group agree with those found in TTHA (Finnen & Pinkerton, 1997), H3heedta (Kettmann *et al.*, 1993), and some derivatives or complexes of betaine (Ilczyszyn, Barnes *et al.*, 1995; Ilczyszyn, Lis & Ratajczak, 1995; Ratajczak *et al.*, 1994), and show that the carboxyl group is in an acid form.

There is one intermolecular hydrogen bond between O1 and O4, and two C—H···O intermolecular hydrogen bonds (Table 2). The geometry of the latter are similar to those found in *N*-methyl-2-pyrrolidone (Müller *et al.*, 1996) and the solid complex of betaine and maleic acid (Ilczyszyn, Lis & Ratajczak *1995*).

Experimental

A mixture of 2-pyrrolidone (5.67 g, 66.7 mmol), glycine (2.5 g, 33.3 mmol) and methanal (10 ml) (37%) was stirred at room temperature for 4 h. The mixture was filtered and crystals were obtained after slow evaporation of the filtrate at room temperature (yield 55%). Elemental analysis: calculated for $C_{12}H_{19}N_3O_4$ (%): C 53.52, H 7.11, N 15.60; found (%): C 52.73, H 7.29, N 16.07. ^{1}H NMR (DMSO-d_6, p.p.m.): 1.9 (*m*, 4H), 2.2 (*t*, 4H), 3.3 (*s*, 2H), 3.4 (*t*, 4H), 4.2 (*s*, 4H).

Crystal data

$C_{12}H_{19}N_3O_4$
M_r = 269.30
Monoclinic
$P2_1/c$
a = 13.7310 (6) Å
b = 9.1357 (5) Å
c = 10.8227 (5) Å
β = 98.852 (4)°
V = 1341.45 (11) Å^3
Z = 4
D_x = 1.333 Mg m^{-3}
D_m not measured
Mo $K\alpha$ radiation
λ = 0.71073 Å
Cell parameters from 25 reflections
θ = 9.98–13.56°
μ = 0.101 mm^{-1}
T = 293 (2) K
Plate
0.40 × 0.37 × 0.15 mm
Colourless

Data collection

Nonius CAD-4 MACH3 diffractometer
$\theta/2\theta$ scans
Absorption correction: none
3097 measured reflections
2936 independent reflections
2072 reflections with $I > 2\sigma(I)$
R_{int} = 0.012
θ_{max} = 27.04°
$h = -17 \rightarrow 17$
$k = 0 \rightarrow 11$
$l = 0 \rightarrow 13$
3 standard reflections
frequency: 60 min
intensity decay: 3.3%

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)]$ = 0.045
$wR(F^2)$ = 0.119
S = 1.044
2936 reflections
249 parameters
All H-atom parameters refined
$w = 1/[\sigma^2(F_o^2) + (0.0537P)^2 + 0.2819P]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{max}$ = 0.004
$\Delta\rho_{max}$ = 0.249 e Å^{-3}
$\Delta\rho_{min}$ = −0.145 e Å^{-3}
Extinction correction: *SHELX*97 (Sheldrick, 1997)
Extinction coefficient: 0.0053 (15)
Scattering factors from *International Tables for Crystallography* (Vol. C)

Table 1. *Selected geometric parameters (Å, °)*

O1—C1	1.319 (2)	N3—C9	1.339 (2)
O2—C1	1.201 (2)	N3—C8	1.446 (2)
O3—C4	1.220 (2)	N3—C12	1.453 (3)
O4—C9	1.235 (2)	C1—C2	1.518 (2)
N1—C8	1.449 (2)	C4—C5	1.508 (3)
N1—C2	1.449 (2)	C5—C6	1.502 (3)
N1—C3	1.459 (2)	C6—C7	1.519 (3)
N2—C4	1.353 (2)	C9—C10	1.494 (3)
N2—C3	1.440 (2)	C10—C11	1.508 (4)
N2—C7	1.455 (2)	C11—C12	1.514 (4)
C8—N1—C2	113.8 (1)	O3—C4—N2	125.6 (2)
C8—N1—C3	112.9 (1)	O3—C4—C5	126.7 (2)
C2—N1—C3	114.2 (1)	N2—C4—C5	107.8 (2)
C4—N2—C3	124.5 (2)	C6—C5—C4	104.7 (2)
C4—N2—C7	113.3 (2)	C5—C6—C7	104.6 (2)
C3—N2—C7	122.0 (2)	N2—C7—C6	103.5 (2)
C9—N3—C8	124.4 (2)	N3—C8—N1	111.8 (1)
C9—N3—C12	113.7 (2)	O4—C9—N3	123.9 (2)
C8—N3—C12	121.9 (2)	O4—C9—C10	126.6 (2)
O2—C1—O1	124.4 (2)	N3—C9—C10	109.5 (2)
O2—C1—C2	125.6 (2)	C9—C10—C11	105.3 (2)
O1—C1—C2	110.0 (2)	C10—C11—C12	106.7 (2)
N1—C2—C1	116.4 (1)	N3—C12—C11	104.6 (2)
N2—C3—N1	110.6 (2)		

Table 2. *Hydrogen-bonding geometry (Å, °)*

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
O1—H1···O4^{i}	0.93 (3)	1.70 (3)	2.596 (2)	162 (2)
C6—H8···O3^{ii}	0.99 (3)	2.56 (3)	3.532 (3)	167 (2)
C6—H9···O3^{iii}	1.06 (3)	2.44 (3)	3.453 (3)	160 (2)

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

All H atoms were located by difference Fourier synthesis, and refined isotropically.

Data collection: *CAD*-4 *ARGUS Software* (Nonius, 1996). Cell refinement: *CAD*-4 *ARGUS Software*. Data reduction: *MolEN* (Fair, 1990). Program(s) used to solve structure: *SHELX*97 (Sheldrick, 1997). Program(s) used to refine structure: *SHELX*97. Molecular graphics: *SHELXTL/PC* (Sheldrick, 1990). Software used to prepare material for publication: *SHELX*97.

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Supplementary data for this paper are available from the IUCr electronic archives (Reference: TA1237). Services for accessing these data are described at the back of the journal.

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7-Deaza-2′-deoxyisoguanosine adopts the form of an N1—H,2-keto tautomer

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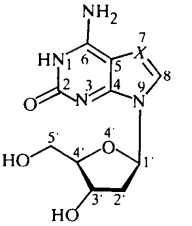
Abstract

The structure of the title hydrate [4-amino-7-(2-deoxy-β-D-*erythro*-pentofuranosyl)-3,7-dihydro-2*H*-pyrrolo-[2,3-*d*]pyrimidin-2-one hydrate, $C_{11}H_{14}N_4O_4\cdot H_2O$], an analogue of 2′-deoxyisoguanosine, was determined by single-crystal X-ray diffraction. The glycosylic torsion angle [$\chi = -118.3(3)°$] is in the *anti* conformation range and the sugar ring adopts a C1′-*exo* pucker. The molecule shows the N1—H,2-keto-6-amino tautomeric form in the solid state.

Comment

Parallel-stranded (ps) DNA duplexes formed by oligonucleotides containing isoguanine and/or 5-methylisocytosine have been realised (Seela *et al.*, 1993; Sugiyama *et al.*, 1996; Seela & Wei, 1997*a*). Also, DNA quartets have been constructed from oligodeoxyribonucleotides containing short runs of isoguanine residues (Seela *et al.*, 1996). More recently, the solution structure of a ps-DNA duplex containing isoguanine and 5-methylisocytosine bases has been determined by NMR analysis (Yang *et al.*, 1998). It was demonstrated that the structural characteristics of the refined ps-duplex are different from those of B-DNA and that most of the nucleosides, including 2′-deoxyisoguanosine, are in the *anti* conformation and show a C2′-*endo* pucker. Recently, an isosteric analogue of 2′-deoxyisoguanosine, namely 7-deaza-2′-deoxyisoguanosine, (1) (purine numbering is used throughout the manuscript), has been synthesized and incorporated into oligonucleotides. Similar thermodynamic base-pair stability and selectivity were observed in the case of 7-deaza-2′-deoxyisoguanosine compared to 2′-deoxyisoguanosine (Seela & Wei, 1999).



(1) X = CH
(2) X = N

Crystal structures related to compound (1) have been reported. They include isoguanine sulfate monohydrate (Subramanian & Marsh, 1971), 9-methylisoguanine hydrochloride dihydrate (Banerjee *et al.*, 1978), 1-methylisoguanine dihydrate (Wong & Nachman, 1984), and 1-allylisoguanosine (Liaw *et al.*, 1992). However, a search of the Cambridge Structural Database revealed that no crystal structures of 7-deazaisoguanine derivatives or of isoguanine 2′-deoxyribonucleosides have been determined. To obtain the conformation of the title compound, (1)·H_2O, its X-ray analysis was performed.

The three-dimensional structure of 7-deaza-2′-deoxyisoguanosine is shown in Fig. 1. Selected bond distances and angles of (1)·H_2O are presented in Table 1. It is apparent that the ring proton is localized at N1 and the molecule adopts the N1—H,2-keto-6-amino tautomeric form (Sepiol *et al.*, 1976). This is in line with the structure of 2′-deoxyisoguanosine, (2), in aqueous solution (Seela *et al.*, 1995). The same tautomeric species of 2′-deoxyisoguanosine has been detected in a crystal structure of a DNA duplex (Robinson *et al.*, 1998). In such an antiparallel duplex, the isoguanine