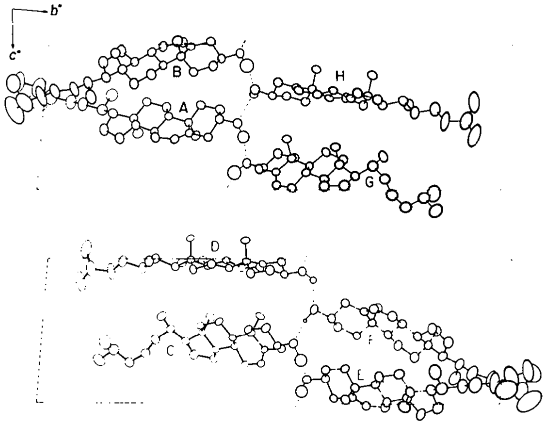

Fig. 4. One unit cell of the structure viewed along **a** (into page). The eight isotropically refined hydroxyl H atoms are shown as 25% spheres. For clarity, the two hydrogen-bonded chains are shown separately; the top figure is to be superposed on the bottom figure to give the entire cell.

$\pm 2{\cdot}18$ Å about the mean plane of the sheet. In this regard the structure differs from the monohydrate and the other solvates studied; in the latter structures the hydrophilic interactions are more narrowly confined to planar sheets. The hydrogen bonding is shown more clearly in Fig. 4, where the isotropically refined hydroxyl H atoms are shown.

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On Nitroguanidines.
II. The Structure of *N*-Methyl-*N'*-nitroguanidine

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Abstract

The crystal and molecular structure of *N*-methyl-*N'*-nitroguanidine, $C_2H_6N_4O_2$ (M_r = 118·096), have been determined by X-ray diffraction at 115 K. The space group is $P2_1/n$ with a = 4·6320 (8), b = 10·1265 (13), c = 11·2399 (13) Å and β = 100·164 (11)° (V = 518·94 Å^3), Z = 4 and $d_{\rm calc}$ = 1·511 Mg m^{-3} (115 K). The observed density at 295 K is $d_{\rm obs}$ = 1·50 Mg m^{-3}. Full-matrix least-squares refinement converged at R = 0·047 for 3294 observed reflexions. Modifications of the weighting scheme in order to reduce polarization errors gave small but significant changes in bonding distances. The molecule is planar with an unusually long formal double C—N bond of 1·377 Å, and is interpreted as a zwitterionic structure. One of the N—O bonds is 0·02 Å longer than the other, and the difference is explained by hydrogen bonding. The crystal is built of hydrogen-bonded sheets parallel to the $(\bar{1}03)$ plane, between which van der Waals interactions prevail.

Introduction

The highly conjugated guanidine moiety of the nitroguanidines forms an interesting bonding system, reflect-

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ing the 'Y-aromaticity' of guanidine (Gund, 1972). As might be expected the introduction of a nitro group into the guanidine molecule drastically alters its chemical properties, *e.g.* pK_a is $-0{\cdot}5$ for nitroguanidine (Amos, Cooper, Nishizawa & Wright, 1961) and 13·6 for guanidine (Gund, 1972).

The molecular structure of nitroguanidine has been known for some time (Bryden, Burkardt, Hughes & Donohue, 1956), and although the bond lengths of the guanidine moiety are somewhat longer than in guanidine (Adams & Ramdas, 1978), the high degree of delocalization is still present. The accuracy of the nitroguanidine structure is, however, rather poor, and any conclusions regarding the effect of the nitro group on the molecular structure are precluded by the high e.s.d.'s in bonding. In order to obtain more accurate data on nitroguanidines, and particularly to look for the sensitivity of this guanidine moiety to different substituents, a series of nitroguanidines are being analyzed by means of X-ray crystallography in our laboratory.

A previous paper in this series dealt with two crystalline modifications of the potent carcinogen, *N*-methyl-*N*-nitroso-*N'*-nitroguanidine (MNNG, Nordenson & Hvoslef, 1981). Both were in the nitrimino-amino form, as was nitroguanidine. The bonding distances were, however, considerably different from those in nitroguanidine, presumably due to the nitroso substituent.

The title compound, *N*-methyl-*N'*-nitroguanidine, is the denitrosated analog of MNNG, and is formed in appreciable amounts when the latter is metabolized (Lawley & Thatcher, 1970; Ioki, Imamura, Nagata & Nakadate, 1975). This transformation may be an essential step in the carcinogenesis, although recent results seem to favor a mechanism in which the active species is nitrocyanamide (Day & Ziolkowski, 1979). The title compound has no biological activity (Ioki *et al.*, 1975).

Experimental

The title compound was prepared from nitroguanidine and methylamine hydrochloride (Fluka), as described by McKay & Wright (1947), and recrystallized from methanolic solution by evaporation. The crystals thus obtained were in general large, elongated prisms of poor quality. As in nitroguanidine (Ripper & Krien, 1969), the crystals frequently had holes or channels along the longer morphological axes. A crystal specimen of satisfactory quality and the dimensions $0{\cdot}3 \times 0{\cdot}3 \times 0{\cdot}4$ mm was selected for the experiments.

The space group was uniquely determined as $P2_1/n$ from the systematic extinctions. The cell parameters were determined from a least-squares fit to 15 reflexions with $35° < 2\theta < 40°$, measured on an automatic Syntex $P\bar{1}$ diffractometer, using graphite-monochromated Mo $K\alpha$ radiation ($\lambda = 0{\cdot}71069$ Å). The conditions for data collection were as in the previous paper in this series (Nordenson & Hvoslef, 1981). All reflexions (1572) with $2\theta < 60°$ were considered, and 1352 of these had $I > 2{\cdot}5\sigma(I)$ and were treated as observed. In the region $60° < 2\theta < 100°$, only those reflexions with intensity larger than a preset threshold value were considered. The total data set thus comprises 3294 observed reflexions. The data were treated in the usual way by programs locally adapted to a CDC 6600 computer (Groth, 1973). Unless otherwise stated this reference covers all computer programs used. Absorption and extinction corrections were not applied.

Structure determination and refinement

The structure was solved by direct methods (Germain, Main & Woolfson, 1971). Full-matrix least-squares refinement with all non-hydrogen atoms gave a conventional R of 0·08. In the last two cycles the non-hydrogen atoms were given anisotropic thermal parameters. At this stage all the H atoms were located from a difference Fourier synthesis, and included in the refinement with isotropic thermal parameters. The refinement converged at $R = 0{\cdot}047$. The atomic scattering factors used were those of Doyle & Turner (1968) for C, N and O, and those of Stewart, Davidson & Simpson (1965) for H.

Several subset refinements were tried in an attempt to reduce the polarization errors inherent in the spherical-atom model (Cruickshank, 1956; Brown, 1969). A commonly used way of doing this is by neglecting the low-order reflexions. The parameters obtained by such a refinement, using all data with $\sin\theta/\lambda > 0{\cdot}7$ Å^{-1}, have been deposited.* This procedure usually excludes the H atoms from the refinement, and in order to include the H atoms and still reduce the polarization errors a different approach was adopted. In the final refinement only reflexions with $\sin\theta/\lambda > 0{\cdot}3$ Å^{-1} were included and, in addition, the weights were reduced for reflexions with $\sin\theta/\lambda < 0{\cdot}65$ Å^{-1}. This was done by multiplication with a sinusoidal function varying from 0 to 1 as $\sin\theta/\lambda$ varies from 0·0 to 0·65 Å^{-1}. Reflexions with $\sin\theta/\lambda > 0{\cdot}65$ Å^{-1} were given ordinary weights

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* A list of observed and calculated structure factors, with e.s.d.'s of the observations, tables of thermal parameters corresponding to the final coordinates, atomic parameters from the high-angle refinement, some data concerning the different subset refinements, the results for the rigid-body approximation (high-angle data) and least-squares plane data have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 35850 (16 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Table 1. *Fractional coordinates and isotropic thermal parameters obtained from the refinement with* $\sin\theta/\lambda > 0{\cdot}3$ Å^{-1} *and with reduced weights for* $\sin\theta/\lambda < 0{\cdot}65$ Å^{-1}

E.s.d.'s are in parentheses. For non-hydrogen atoms $B_{eq} = \frac{8}{3}\pi^2 \sum_i \sum_j U_{ij} a_i^* a_j^* \mathbf{a}_i.\mathbf{a}_j$.

	x	y	z	B_{eq}/B (Å^2)
C(1)	0·61816 (13)	0·88721 (6)	0·39363 (5)	1·16
C(2)	0·76391 (18)	0·67495 (7)	0·48969 (7)	1·71
N(1)	0·80099 (13)	0·81563 (6)	0·47280 (5)	1·38
N(2)	0·70379 (12)	1·01736 (5)	0·39236 (5)	1·28
N(3)	0·38159 (13)	0·83380 (6)	0·32854 (5)	1·48
N(5)	0·55288 (12)	1·10035 (5)	0·31381 (5)	1·30
O(2)	0·64292 (13)	1·21654 (5)	0·32009 (5)	1·74
O(3)	0·33425 (13)	1·06819 (6)	0·23494 (5)	1·90
H(N1)	0·959 (3)	0·853 (1)	0·509 (1)	1·3 (2)
H(N3)1	0·347 (4)	0·751 (2)	0·333 (2)	2·8 (3)
H(N3)2	0·258 (4)	0·882 (2)	0·283 (2)	2·7 (3)
H(C2)1	0·780 (4)	0·627 (2)	0·417 (2)	2·2 (3)
H(C2)2	0·925 (5)	0·646 (2)	0·546 (2)	2·9 (3)
H(C2)3	0·580 (5)	0·657 (2)	0·513 (2)	2·9 (3)

obtained from counting statistics modified for experimental instability. The results from this refinement are in better agreement with the high-angle refinement results than those using the full dataset, the weighted residual and goodness-of-fit are substantially improved, and the e.s.d.'s of the H atom positions are only slightly increased (from 0·014 to 0·019 Å). The coordinates obtained from this refinement are given in Table 1.* A difference Fourier synthesis revealed peaks of 0·4 to 0·7 e Å^{-3} at the expected positions of lone-pair and conjugated bonding electrons. Apart from these peaks the electron density has an absolute value of less than 0·25 e Å^{-3} throughout the cell.

A much better approach is, of course, to improve the model, *e.g.* by deformation refinement (Hirshfeld & Hope, 1980), but it is felt that the quality of the data does not justify the efforts of such a procedure.

Discussion

Molecular structure

The molecule, with thermal ellipsoids, atomic notation, bonding distances and angles, is shown in Fig. 1, whereas the torsion angles are given in Table 2. The molecule is almost planar, and all torsion angles (not including H atoms) are within 5° of 0° or 180°. This planarity necessitates a very short non-bonded contact between N(3) and O(3). The strain imposed on the molecule by this short contact is released by (1) an opening of the N(2)—N(5)—O(3) and N(2)—C(1)—N(3) angles and (2) establishing a hydrogen bond, N(3)—H···O(3), that stabilizes the molecule. The opening of these angles is accompanied by a closing of the N(2)—N(5)—O(2) and N(1)—C(1)—N(2) angles, whereas the remaining angles are close to those expected from the hybridizations.

The bonding distances reveal some interesting features. The formal double bond, C(1)—N(2), is 0·05 Å longer than the two other C—N bonds of the guanidine moiety, which are of approximately the same length as the C—N bonds in protonated guanidine moieties (Cotton, Day, Hazen & Larsen, 1973; Cotton, Day, Hazen, Larsen & Wang, 1974; Larsen, 1975; Berthou, Laurent & Nakajima, 1976; Bracuti, 1979), and only slightly longer than in the guanidinium ion (Adams & Ramdas, 1978). The nitrimino group closely resembles that in trimethylammonionitramidate (Cameron, Hair & Morris, 1972), where the zwitterionic structure is evident, and that in cyanonitramide,

Table 2. *Torsion angles* (°) *corresponding to the coordinates of Table* 1

E.s.d.'s are in the range 0·06 to 0·11° for non-hydrogen atoms and 1° for those involving H atoms.

N(2)–C(1)–N(1)–C(2)	−178·5	N(1)–C(1)–N(3)–H(N3)1	−4
N(3)–C(1)–N(1)–C(2)	2·9	N(2)–C(1)–N(3)–H(N3)1	178
N(1)–C(1)–N(2)–N(5)	176·5	N(1)–C(1)–N(3)–H(N3)2	175
N(3)–C(1)–N(2)–N(5)	−5·0	N(2)–C(1)–N(3)–H(N3)2	−4
C(1)–N(2)–N(5)–O(2)	179·0		
C(1)–N(2)–N(5)–O(3)	−2·2	C(1)–N(1)–C(2)–H(C2)1	63
N(2)–C(1)–N(1)–H(N1)	−6	C(1)–N(1)–C(2)–H(C2)2	176
N(3)–C(1)–N(1)–H(N1)	175	C(1)–N(1)–C(2)–H(C2)3	−59

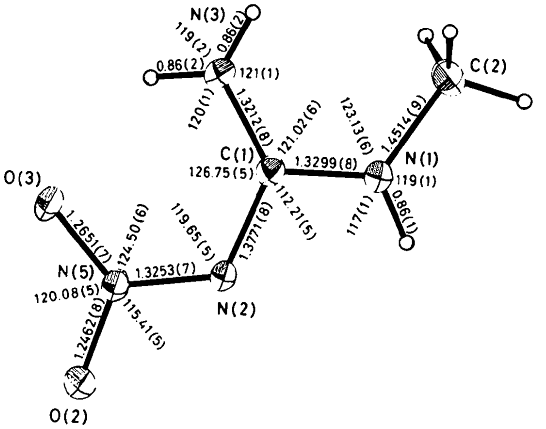

Fig. 1. The *N*-methyl-*N'*-nitroguanidine molecule with the thermal ellipsoids scaled to 50% probability (Johnson, 1976), the atomic notation, and bonding distances (Å) and angles (°). The e.s.d.'s, referring to the last figure, are given in parentheses. The average C—H distance of the methyl group is 0·95 Å, and the range of the valence angles involving these H atoms is 104 to 114°.

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* See deposition footnote.

where the localization of a negative charge in the nitrimino group has been supported by *ab initio* calculations (Jensen, Klewe & Tjelta, 1977). The latter work clearly points out the importance of resonance forms with negative charge on the amide(imino) N atom. These features might be taken as indicative of a zwitterionic structure for the title compound, where a positive charge is shared between the two amino groups, whereas a negative charge is delocalized in the highly electronegative nitrimino group (Huheey, 1966).

The difference between the two N—O bonds is easily explained by the hydrogen bonding; O(3) is an acceptor for two such bonds, whereas O(2) does not participate in the hydrogen-bonding system. The same effect is observed in several aromatic nitro-substituted molecules (for example, Brown & Towns, 1977; Duesler, Engelmann, Curtin & Paul, 1978). The different degree of hydrogen-bonding participation may also be the reason for the small but significant difference between the C—N(amino) bonds in the guanidine moiety.

Table 3 reveals the bond lengths obtained from some of the subset refinements, and those corrected for rigid-body motion (Schomaker & Trueblood, 1968) from the high-angle refinement. The rigid-body approximation was very good with $(\sum \Delta U_{ij}^2/6n_a - n_p)^{1/2} = 0{\cdot}0007$. Further details of this analysis are given in the deposited material. The average bond-length correction is 0·004 Å, which is approximately the same as the average of the absolute values of the differences between the results from the refinements with $\sin\theta/\lambda$ less than 0·7 Å^{-1} (I) and larger than 0·7 Å^{-1} (III). Comparison of the final results (II) with the high-angle results (III) shows that the remaining differences are associated with atoms having lone-pair electrons in the molecular plane.

A comparison of the bond lengths and angles in the guanidine moieties of several related molecules is shown in Table 4. With the exception of the last compound all of these guanidine moieties have at least one of the formal single bonds shorter than the formal double bond [C—N(2)], the most extreme cases being the azidoformamidinium ion (*c*) and the title compound. The N(2) substituents in these two compounds both have very high electronegativities (Huheey, 1966), which may be the reason for the lengthening of the adjacent C—N bonds. The differences between nitroguanidine and the title compound may seem large, but are not significant due to the high e.s.d.'s of the former. However, comparison of the title compound with the nitrosated analogue, MNNG (*g*), reveals large, highly significant differences. The introduction of an electro-

Table 3. *Bond lengths* (Å) *from various subset refinements*

(I) is based on reflexions with $\sin\theta/\lambda < 0{\cdot}7$ Å^{-1}, (II) corresponds to Table 1, (III) is based on reflexions with $\sin\theta/\lambda > 0{\cdot}7$ Å^{-1}, and (IV) gives bond lengths corrected for rigid-body motion (Schomaker & Trueblood, 1968) from the refinement with $\sin\theta/\lambda > 0{\cdot}7$ Å^{-1}.

	(I)	(II)	(III)	(IV)
C(1)—N(1)	1·328	1·330	1·330	1·337
C(1)—N(2)	1·379	1·377	1·375	1·378
C(1)—N(3)	1·316	1·321	1·322	1·328
C(2)—N(1)	1·453	1·451	1·451	1·453
N(2)—N(5)	1·329	1·325	1·323	1·327
N(5)—O(2)	1·247	1·246	1·245	1·248
N(5)—O(3)	1·266	1·265	1·263	1·269

Table 4. *Comparison of bonding distances* (Å) *and angles* (°) *of the guanidine moieties of several related compounds*

Reference	C—N(1)	C—N(2)	C—N(3)	N(1)CN(2)	N(1)CN(3)	N(2)CN(3)	Substituent on N(2)
(*a*)	1·329	1·344	1·329	117·5	117·5	124·9	$-C(NH_2)_2^+$
(*a*)	1·361	1·329	1·325	111·2	117·5	126·2	$-C(NH_2)NH$
(*b*)	1·333	1·341	1·339	117·5	118·7	123·8	—CN
(*c*)	1·314	1·393	1·302	113·8	123·1	123·0	$-N_2^+$
(*d*)	1·329	1·348	1·334	115·8	118·1	126·0	$-SO_2C_6H_4NH_2$
(*e*)	1·34	1·35	1·34	111	118	129	$-NO_2$
(*f*)	1·330	1·377	1·321	112·2	121·0	126·8	$-NO_2$*
(*g*)	1·403	1·335	1·312	111·9	117·9	130·3	$-NO_2$†
(*h*)	1·406	1·331	1·320	115·6	117·1	127·3	‡
(*i*)	1·42	1·28	1·30	107·7	120·9	131·4	—Cl§

* Methyl group on N(1).
† Methyl and nitroso groups on N(1).
‡ N(2) and N(3) in pyrimidine ring and $-SO_2R$ on N(1)
§ N(1) bonded to N(1)′ through an inversion center.

References: (*a*) Pinkerton & Schwartzenbach (1978); (*b*) Hirshfeld & Hope (1980); (*c*) Henke & Bärnighausen (1972); (*d*) Alléaume, Gulko, Herbstein, Kapon & Marsh (1976); (*e*) Bryden *et al.* (1956); (*f*) this work (e.s.d.'s 0·001 Å and 0·1°); (*g*) Nordenson & Hvoslef (1981); (*h*) Giuseppetti, Tadini, Bettinetti & Giordano (1977); (*i*) Bryden (1958).

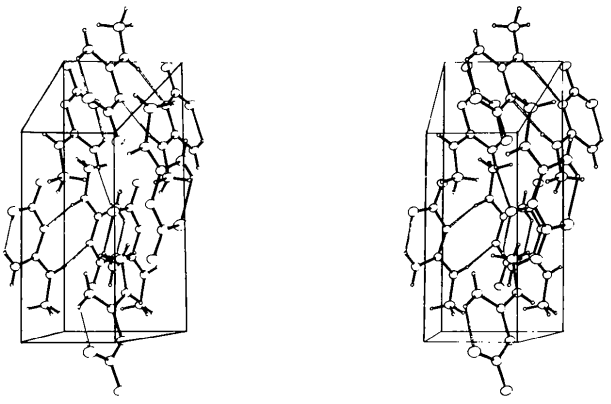

Fig. 2. Stereodrawing (Johnson, 1976) of the crystal structure. The view is approximately along the *c* axis, with the *a* axis pointing to the left and the *b* axis pointing up.

Table 5. *Details of the hydrogen-bonding system*

E.s.d.'s for distances involving H atoms are 0·02 Å, for other distances 0·001 Å, and for angles 1–2°.

D–H···A	D–H	H···A	D···A	D–H···A	H···A–R
N(3)–H(2)···O(3)^{i}	0·86	2·00	2·590	125	102
N(3)–H(1)···O(3)^{ii}	0·86	2·12	2·914	154	104
N(1)–H···N(2)^{iii}	0·86	2·19	3·032	168	120*

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

* Average value.

negative substituent on N(1) would be expected to change the bonding distances in the direction observed, but not to the same extent (see, for example, Talberg, 1979).

Crystal structure

The crystal structure with the hydrogen bonding is shown in Fig. 2 and details of the hydrogen-bonding system are given in Table 5. The crystal consists of hydrogen-bonded sheets parallel to the ($\bar{1}03$) plane, between which van der Waals interactions prevail. The shortest contact between the methyl groups is 4·15 Å. The hydrogen bonds are all of intermediate strength and have normal geometry. O(2) does not participate in the hydrogen bonding. Its shortest O···H interactions are 2·61 and 2·68 Å. N(2) is an acceptor for a hydrogen bond, as in one of the molecules of MNNG.

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