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Carbamoyl(diaminomethylidene)azanium 3-nitro-5-oxo-4,5-dihydro-1*H*-1,2,4-triazol-4-ide

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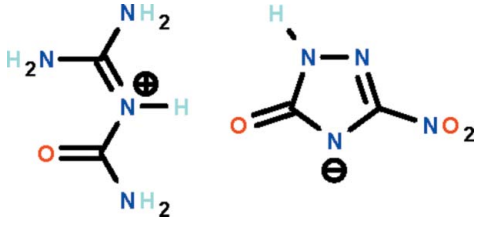
Key indicators: single-crystal X-ray study; T = 293 K; mean σ(N–C) = 0.003 Å; R factor = 0.044; wR factor = 0.116; data-to-parameter ratio = 11.5.

In the anion of the title salt, $C_2H_7N_4O^+\cdot C_2HN_4O_3^-$, the negative charge resides formally on the N^3 atom of the triazole ring. In the crystal, the N^3 and exocyclic O atoms are hydrogen-bond acceptors with respect to the formally double-bond iminium and amido N atoms of the cation. The cation and anion are almost planar (r.m.s. deviations = 0.012 and 0.051 Å, respectively), but they are slightly bent with respect to each other [dihedral angle = 12.6 (1)°]. In the crystal, adjacent anions and cations are linked by extensive N—H···N and N—H···O hydrogen bonds, generating a ribbon running along the *b*-axis direction.

Related literature

For background to applications of similar compounds as propellants and explosives, see: Liu *et al.* (2006); Östmark *et al.* (2002).



Experimental

Crystal data

$C_2H_7N_4O^+\cdot C_2HN_4O_3^-$
M_r = 232.18
Monoclinic, $P2_1/n$
a = 3.7100 (5) Å
b = 13.4195 (19) Å
c = 18.033 (3) Å
β = 94.143 (3)°
V = 895.5 (2) Å^3
Z = 4
Mo $K\alpha$ radiation
μ = 0.15 mm^{-1}
T = 293 K
0.30 × 0.30 × 0.20 mm

Data collection

Bruker SMART APEX diffractometer
5217 measured reflections
2032 independent reflections
1297 reflections with $I > 2\sigma(I)$
R_{int} = 0.035

Refinement

$R[F^2 > 2\sigma(F^2)]$ = 0.044
$wR(F^2)$ = 0.116
S = 1.00
2032 reflections
177 parameters
All H-atom parameters refined
$\Delta\rho_{max}$ = 0.17 e Å^{-3}
$\Delta\rho_{min}$ = −0.23 e Å^{-3}

Table 1
Hydrogen-bond geometry (Å, °).

D—H···A	D—H	H···A	D···A	D—H···A
N1—H1···O4^{i}	0.87 (2)	1.97 (2)	2.819 (2)	166 (2)
N5—H2···N3	0.94 (2)	1.99 (3)	2.926 (3)	173 (2)
N5—H3···O1^{ii}	0.90 (3)	2.13 (3)	3.005 (2)	164 (2)
N6—H4···O1	0.89 (2)	1.96 (2)	2.824 (2)	163 (2)
N8—H5···O1	0.95 (3)	2.15 (3)	2.966 (3)	142 (2)
N8—H6···O3^{iii}	0.87 (2)	2.32 (3)	3.183 (3)	173 (2)
N7—H7···N2^{iii}	0.90 (2)	2.03 (3)	2.913 (2)	166 (2)
N7—H8···O4	0.85 (2)	2.02 (2)	2.645 (2)	129 (2)

Symmetry codes: (i) $-x+\frac{1}{2}, y+\frac{1}{2}, -z+\frac{3}{2}$; (ii) $-x+\frac{1}{2}, y-\frac{1}{2}, -z+\frac{3}{2}$; (iii) $x-\frac{3}{2}, -y+\frac{3}{2}, z-\frac{1}{2}$.

Data collection: *APEX2* (Bruker, 2007); cell refinement: *SAINT* (Bruker, 2007); data reduction: *SAINT*; program(s) used to solve structure: *SHELXS97* (Sheldrick, 2008); program(s) used to refine structure: *SHELXL97* (Sheldrick, 2008); molecular graphics: *X-SEED* (Barbour, 2001); software used to prepare material for publication: *publCIF* (Westrip, 2010).

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Supplementary data and figures for this paper are available from the IUCr electronic archives (Reference: XU5702).

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supporting information

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Carbamoyl(diaminomethylidene)azanium 3-nitro-5-oxo-4,5-dihydro-1*H*-1,2,4-triazol-4-ide

Xin-Ping Huang, Bo-Zhou Wang, Dong-Ping Li and Seik Weng Ng

S1. Comment

We have reported organic compounds that do not possess carbon-bound hydrogen atoms; *N*-guanylurea dinitramide, $NH_2C(NH)NHC(O)NH_2{\cdot}NH(NO_2)_2$ (Liu *et al.*, 2006), exemplifies such a compound that has been evaluated for use as a propellant and an insensitive-munitions explosive (Östmark *et al.*, 2002). The title salt (Scheme I, Fig. 1) features an $(NH_2)_2C(NH)C(O)NH_2$ cation that has been protonated by 3-nitro-1,2,4-triazol-5-one, which is acidic owing to the electron-withdrawing nitro group. The N^3 and exocyclic O atoms are hydrogen bond acceptors with respect to the formally double-bond iminium and amido N atoms of the cation. The cation and anion are planar but they are slightly bent with respect to each other. Adjacent ion-pairs are linked by extensive N···N and N···O hydrogen bonds to generate a ribbon structure (Table 1).

S2. Experimental

3-Nitro-1,2,4-triazol-5-one (26.0 g, 0.2 mol) was suspended in water (150 ml) kept at 303–313 K. Sodium hydroxide (8.2 g, 0.2 mol) dissolved in water (50 ml) was added. Guanylurea hydrochloride (27.8 g, 0.2 mol) dissolved in water (175 ml) was aded. The mixture was warmed to 323–333 K for 1.5 h. This was then cooled to 275–278 K. The solid material was collected and recrystallized from water (yield 35.0 g, 85% yield).

S3. Refinement

Hydrogen atoms were located in a difference Fourier map, and were freely refined.

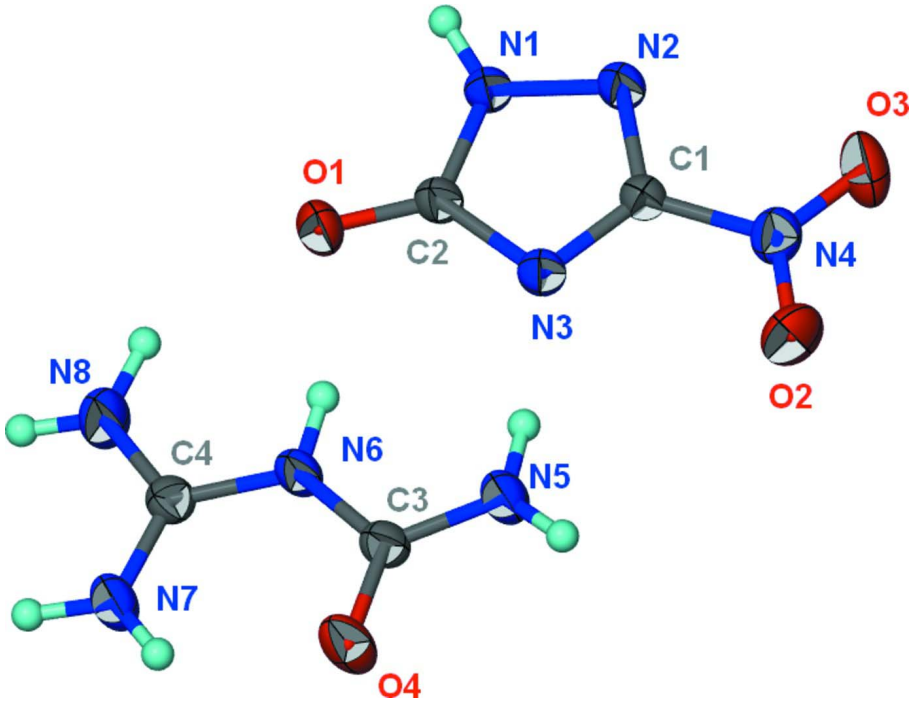

Figure 1

Thermal ellipsoid plot (Barbour, 2001) of $C_2H_7N_4O{\cdot}C_2HN_4O_3$ at the 70% probability level; hydrogen atoms are drawn as spheres of arbitrary radius.

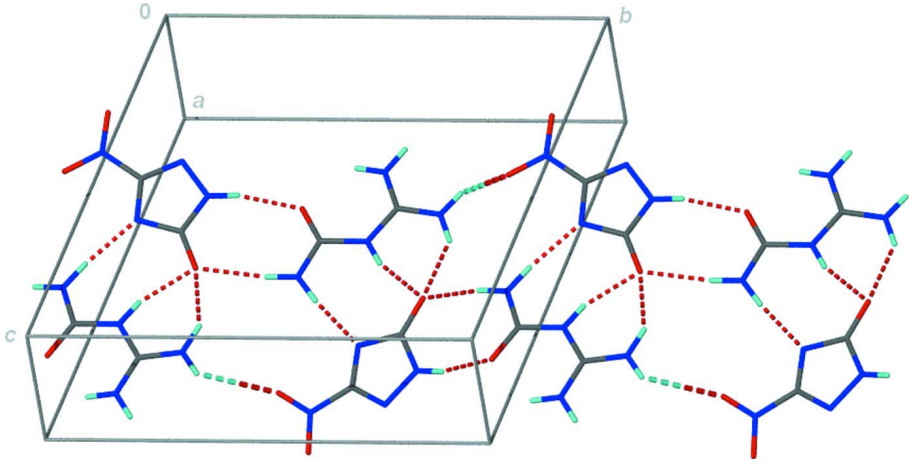

Figure 2

Packing diagram.

Carbamoyl(diaminomethylidene)azanium 3-nitro-5-oxo-4,5-dihydro-1*H*-1,2,4-triazol-4-ide

Crystal data

$C_2H_7N_4O^+{\cdot}C_2HN_4O_3^-$
M_r = 232.18
Monoclinic, $P2_1/n$
Hall symbol: -P 2yn
a = 3.7100 (5) Å
b = 13.4195 (19) Å
c = 18.033 (3) Å
β = 94.143 (3)°
V = 895.5 (2) Å^3
Z = 4

$F(000) = 480$
$D_x = 1.722$ Mg m^{-3}
Mo $K\alpha$ radiation, $\lambda = 0.71073$ Å
Cell parameters from 976 reflections
$\theta = 2.3$–$24.8°$

$\mu = 0.15$ mm^{-1}
$T = 293$ K
Prism, yellow
0.30 × 0.30 × 0.20 mm

Data collection

Bruker SMART APEX diffractometer
Radiation source: fine-focus sealed tube
Graphite monochromator
ω scans
5217 measured reflections
2032 independent reflections

1297 reflections with $I > 2\sigma(I)$
$R_{int} = 0.035$
$\theta_{max} = 27.5°$, $\theta_{min} = 1.9°$
$h = -4 \rightarrow 4$
$k = -14 \rightarrow 17$
$l = -22 \rightarrow 21$

Refinement

Refinement on F^2
Least-squares matrix: full
$R[F^2 > 2\sigma(F^2)] = 0.044$
$wR(F^2) = 0.116$
$S = 1.00$
2032 reflections
177 parameters
0 restraints
Primary atom site location: structure-invariant direct methods

Secondary atom site location: difference Fourier map
Hydrogen site location: inferred from neighbouring sites
All H-atom parameters refined
$w = 1/[\sigma^2(F_o^2) + (0.0564P)^2]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{max} = 0.001$
$\Delta\rho_{max} = 0.17$ e Å^{-3}
$\Delta\rho_{min} = -0.23$ e Å^{-3}

Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters (Å^2)

	x	y	z	U_{iso}*/U_{eq}
O1	0.3898 (4)	0.79686 (10)	0.74934 (8)	0.0387 (4)
O2	0.9393 (5)	0.48678 (10)	0.86971 (9)	0.0526 (5)
O3	1.2361 (4)	0.56912 (12)	0.95758 (9)	0.0502 (5)
O4	0.0042 (5)	0.49420 (10)	0.59852 (8)	0.0429 (4)
N1	0.7380 (5)	0.80376 (12)	0.86167 (9)	0.0309 (4)
N2	0.9228 (5)	0.73874 (11)	0.90892 (8)	0.0301 (4)
N3	0.6699 (5)	0.65784 (11)	0.80734 (9)	0.0288 (4)
N4	1.0257 (5)	0.56440 (12)	0.90216 (9)	0.0343 (4)
N5	0.3102 (5)	0.50638 (14)	0.71153 (9)	0.0356 (5)
N6	0.1708 (5)	0.64808 (12)	0.64482 (9)	0.0304 (4)
N7	−0.1373 (5)	0.66445 (15)	0.52843 (10)	0.0364 (5)
N8	0.0601 (6)	0.80164 (14)	0.59459 (12)	0.0465 (6)
C1	0.8697 (5)	0.65588 (13)	0.87193 (10)	0.0257 (4)
C2	0.5814 (5)	0.75613 (14)	0.80084 (10)	0.0281 (5)
C3	0.1543 (6)	0.54422 (14)	0.64972 (10)	0.0291 (5)
C4	0.0251 (6)	0.70408 (14)	0.58795 (10)	0.0283 (4)
H1	0.703 (6)	0.8651 (17)	0.8753 (12)	0.042 (6)*
H2	0.434 (6)	0.5511 (18)	0.7446 (12)	0.046 (7)*
H3	0.290 (7)	0.440 (2)	0.7190 (14)	0.064 (8)*
H4	0.257 (7)	0.6841 (19)	0.6834 (13)	0.053 (7)*
H5	0.177 (8)	0.8310 (19)	0.6379 (16)	0.076 (9)*

H6	−0.034 (7)	0.8406 (18)	0.5601 (13)	0.052 (7)*
H7	−0.248 (6)	0.7032 (18)	0.4929 (13)	0.051 (7)*
H8	−0.162 (6)	0.6015 (18)	0.5248 (13)	0.046 (7)*

Atomic displacement parameters (Å²)

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
O1	0.0502 (10)	0.0337 (8)	0.0302 (8)	0.0054 (7)	−0.0120 (7)	0.0002 (6)
O2	0.0776 (13)	0.0250 (8)	0.0544 (10)	0.0059 (8)	−0.0017 (9)	−0.0022 (7)
O3	0.0493 (10)	0.0527 (10)	0.0459 (10)	0.0033 (8)	−0.0145 (8)	0.0133 (8)
O4	0.0670 (11)	0.0287 (8)	0.0307 (8)	−0.0121 (7)	−0.0130 (7)	0.0012 (6)
N1	0.0418 (11)	0.0222 (8)	0.0274 (9)	0.0024 (8)	−0.0054 (8)	−0.0039 (7)
N2	0.0362 (11)	0.0268 (8)	0.0262 (9)	0.0003 (7)	−0.0050 (7)	−0.0025 (7)
N3	0.0338 (10)	0.0244 (9)	0.0272 (9)	0.0000 (7)	−0.0036 (7)	−0.0030 (7)
N4	0.0375 (11)	0.0305 (9)	0.0348 (10)	0.0012 (8)	0.0027 (8)	0.0045 (8)
N5	0.0521 (12)	0.0251 (9)	0.0276 (10)	−0.0028 (9)	−0.0099 (9)	0.0016 (8)
N6	0.0434 (11)	0.0234 (8)	0.0231 (9)	−0.0028 (8)	−0.0071 (8)	−0.0013 (7)
N7	0.0484 (12)	0.0306 (10)	0.0283 (10)	−0.0027 (9)	−0.0103 (9)	0.0033 (8)
N8	0.0717 (16)	0.0240 (10)	0.0413 (12)	0.0037 (10)	−0.0129 (11)	0.0019 (9)
C1	0.0271 (11)	0.0260 (10)	0.0238 (10)	0.0004 (8)	0.0004 (8)	−0.0009 (8)
C2	0.0341 (12)	0.0246 (10)	0.0252 (10)	−0.0009 (8)	−0.0014 (9)	−0.0020 (8)
C3	0.0362 (12)	0.0263 (10)	0.0244 (10)	−0.0025 (9)	−0.0002 (9)	−0.0013 (8)
C4	0.0313 (11)	0.0275 (10)	0.0263 (10)	−0.0003 (9)	0.0024 (8)	0.0003 (8)

Geometric parameters (Å, º)

O1—C2	1.253 (2)	N5—H2	0.94 (2)
O2—N4	1.226 (2)	N5—H3	0.90 (3)
O3—N4	1.224 (2)	N6—C4	1.352 (2)
O4—C3	1.240 (2)	N6—C3	1.398 (2)
N1—C2	1.363 (2)	N6—H4	0.89 (2)
N1—N2	1.369 (2)	N7—C4	1.306 (3)
N1—H1	0.87 (2)	N7—H7	0.90 (2)
N2—C1	1.304 (2)	N7—H8	0.85 (2)
N3—C1	1.335 (2)	N8—C4	1.320 (3)
N3—C2	1.362 (2)	N8—H5	0.95 (3)
N4—C1	1.447 (2)	N8—H6	0.87 (2)
N5—C3	1.320 (2)		
C2—N1—N2	111.56 (15)	H7—N7—H8	119 (2)
C2—N1—H1	127.4 (15)	C4—N8—H5	121.4 (16)
N2—N1—H1	120.3 (15)	C4—N8—H6	120.0 (16)
C1—N2—N1	100.07 (14)	H5—N8—H6	118 (2)
C1—N3—C2	102.08 (15)	N2—C1—N3	118.90 (16)
O3—N4—O2	124.25 (18)	N2—C1—N4	119.28 (17)
O3—N4—C1	118.53 (17)	N3—C1—N4	121.82 (16)
O2—N4—C1	117.21 (17)	O1—C2—N3	127.31 (18)
C3—N5—H2	117.0 (14)	O1—C2—N1	125.30 (18)

C3—N5—H3	118.0 (17)	N3—C2—N1	107.39 (16)
H2—N5—H3	125 (2)	O4—C3—N5	124.49 (19)
C4—N6—C3	125.80 (17)	O4—C3—N6	120.77 (18)
C4—N6—H4	113.2 (16)	N5—C3—N6	114.74 (18)
C3—N6—H4	120.6 (16)	N7—C4—N8	121.0 (2)
C4—N7—H7	120.5 (15)	N7—C4—N6	122.19 (18)
C4—N7—H8	120.5 (16)	N8—C4—N6	116.81 (19)
C2—N1—N2—C1	1.2 (2)	C1—N3—C2—O1	−179.1 (2)
N1—N2—C1—N3	−1.0 (2)	C1—N3—C2—N1	0.4 (2)
N1—N2—C1—N4	179.47 (16)	N2—N1—C2—O1	178.41 (19)
C2—N3—C1—N2	0.4 (2)	N2—N1—C2—N3	−1.1 (2)
C2—N3—C1—N4	179.90 (17)	C4—N6—C3—O4	1.9 (3)
O3—N4—C1—N2	−7.9 (3)	C4—N6—C3—N5	−178.53 (19)
O2—N4—C1—N2	172.69 (18)	C3—N6—C4—N7	−3.2 (3)
O3—N4—C1—N3	172.61 (18)	C3—N6—C4—N8	178.0 (2)
O2—N4—C1—N3	−6.8 (3)		

Hydrogen-bond geometry (Å, º)

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
N1—H1···O4^{i}	0.87 (2)	1.97 (2)	2.819 (2)	166 (2)
N5—H2···N3	0.94 (2)	1.99 (3)	2.926 (3)	173 (2)
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