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4,4,5,5-Tetramethyl-2-(4-pyridinio)-imidazoline-1-oxyl-3-oxide chloride

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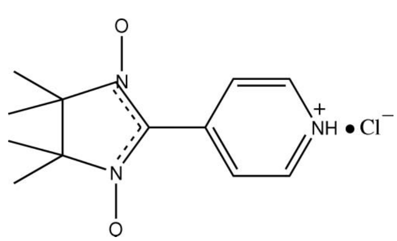
Key indicators: single-crystal X-ray study; T = 291 K; mean σ(C–C) = 0.004 Å; R factor = 0.048; wR factor = 0.140; data-to-parameter ratio = 15.6.

The title compound $C_{12}H_{17}N_3O_2^+\cdot Cl^-$ consists of a discrete $[NITpPyH]^+$ cation [NITpPy = 2-(4′-pyridyl)-4,4,5,5-tetramethylimidazoline-1-oxyl-3-oxide] and a chloride anion. The NITpPy molecule is protonated at the N atom of the pyridyl ring. The anions and cations are connected *via* N—H···Cl hydrogen bonds.

Related literature

For the design and synthesis of molecule-based magnetic materials, see: Bogani *et al.* (2005); Wang *et al.* (2004). For nitronyl nitroxide radicals (NITR), see: Fettouhi *et al.* (2003). For related literature, see: Stroh *et al.* (1999); Hirel *et al.* (2001); Chang *et al.* (2005); Wang *et al.* (2003). For the synthesis of the title compound see: Ullman *et al.* (1970, 1972)



Experimental

Crystal data

$C_{12}H_{17}N_3O_2^+\cdot Cl^-$
M_r = 270.74
Monoclinic, $P2_1/c$
a = 10.863 (14) Å
b = 11.927 (15) Å
c = 11.130 (15) Å
β = 102.81 (2)°
V = 1406 (3) Å^3
Z = 4
Mo $K\alpha$ radiation
μ = 0.27 mm^{-1}
T = 291 (2) K
0.30 × 0.26 × 0.23 mm

Data collection

Bruker SMART CCD area-detector diffractometer
Absorption correction: multi-scan (*SADABS*; Sheldrick, 1996) T_{min} = 0.923, T_{max} = 0.939
7172 measured reflections
2609 independent reflections
2120 reflections with $I > 2\sigma(I)$
R_{int} = 0.037

Refinement

$R[F^2 > 2\sigma(F^2)]$ = 0.048
$wR(F^2)$ = 0.140
S = 1.03
2609 reflections
167 parameters
H-atom parameters constrained
$\Delta\rho_{max}$ = 0.44 e Å^{-3}
$\Delta\rho_{min}$ = −0.22 e Å^{-3}

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

D—H···A	D—H	H···A	D···A	D—H···A
N1—H1D···Cl1^i	0.86	2.17	3.028 (3)	174

Symmetry code: (i) $x+1, y+1, z$.

Data collection: *SMART* (Bruker, 2002); cell refinement: *SAINT* (Bruker, 2002); data reduction: *SAINT*; program(s) used to solve structure: *SHELXS97* (Sheldrick, 2008); program(s) used to refine structure: *SHELXL97* (Sheldrick, 2008); molecular graphics: *SHELXTL* (Sheldrick, 2008); software used to prepare material for publication: *publCIF* (Westrip, 2009).

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Supplementary data and figures for this paper are available from the IUCr electronic archives (Reference: BX2189).

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4,4,5,5-Tetramethyl-2-(4-pyridinio)imidazoline-1-oxyl-3-oxide chloride

Jiu Li Chang, Zhi Yong Gao and Kai Jiang

S1. Comment

The design and synthesis of molecule-based magnetic materials is one of the major subjects of materials science in which the combination of metal ions and organic radicals are used to construct assembled systems (Bogani *et al.*, 2005; Wang *et al.*, 2004). Nitronyl nitroxide radicals (NITR), independently or in combination with metal ions, have been one of the most widely studied systems in molecular magnetism for understanding the radical-radical or metal-radical as well as for synthesizing organic ferromagnets and metal-radical magnetic materials (Fettouhi *et al.*, 2003). However, to our knowledge so far few charge transfer complexes of nitronyl nitroxide radicals used as proton receptor have been reported. In order to better understand the behavior of proton transfer in charge transfer complexes, the synthesis and crystal structure of the title compound have been investigated. The structure of the title compound is shown in Fig. 1. The NITpPy molecule is protonated at N atom of the pyridyl ring by accepting a proton from the acid solution. The transfer of protons result in a intermolecular hydrogen bond between NITpPy and chloride.The anions and cations are connected *via* N—H···Cl hydrogen bonds. The nitronyl nitroxide fragment O—N—C—N—O is almost coplanar, but make a dihedral angle of 8.6 (2)° with the pyridyl ring.

S2. Experimental

NITpPy was synthesized according to a literature procedure (Ullman *et al.*,1970; Ullman *et al.*, 1972). Single crystals of the title compound suitable for X-ray measurements were obtained by recrystallization from acetonitrile solution and HCl 10:1 (*v*/*v*) solution at room temperature.

S3. Refinement

The H atoms were positioned geometrically and refined using the riding-model approximation, with C—H = 0.93 or 0.96 Å and N—H = 0.96 Å and U_{iso}(H) = 1.2U_{eq}(carrier) or U_{iso}(H) = 1.5U_{eq}(methyl carrier).

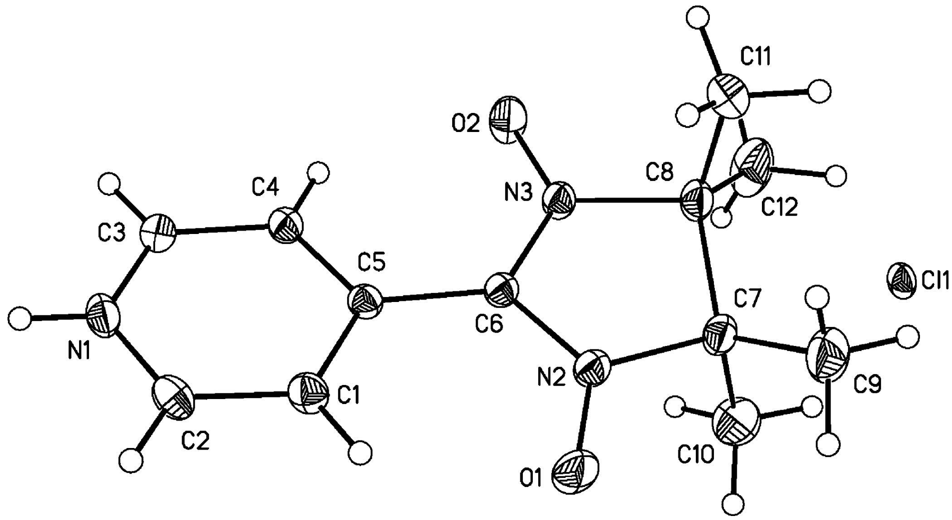

Figure 1

ORTEP drawing of the title compound with atom labeling. The thermal ellipsoids are drawn at 30% probability level.

4,4,5,5-Tetramethyl-2-(4-pyridinio)imidazoline-1-oxyl-3-oxide chloride

Crystal data

$C_{12}H_{17}N_3O_2^{+}\cdot Cl^{-}$
M_r = 270.74
Monoclinic, $P2_1/c$
Hall symbol: -P 2ybc
a = 10.863 (14) Å
b = 11.927 (15) Å
c = 11.130 (15) Å
β = 102.81 (2)°
V = 1406 (3) Å^3
Z = 4
F(000) = 572
D_x = 1.279 Mg m^{-3}
Mo $K\alpha$ radiation, λ = 0.71073 Å
Cell parameters from 3005 reflections
θ = 2.5–27.3°
μ = 0.27 mm^{-1}
T = 291 K
BLOCK, black
0.30 × 0.26 × 0.23 mm

Data collection

Bruker SMART CCD area-detector diffractometer
Radiation source: fine-focus sealed tube
Graphite monochromator
phi and ω scans
Absorption correction: multi-scan (*SADABS*; Sheldrick, 1996)
T_{min} = 0.923, T_{max} = 0.939
7172 measured reflections
2609 independent reflections
2120 reflections with $I > 2\sigma(I)$
R_{int} = 0.037
θ_{max} = 25.5°, θ_{min} = 2.5°
$h = -13 \rightarrow 11$
$k = -13 \rightarrow 14$
$l = -13 \rightarrow 13$

Refinement

Refinement on F^2
Least-squares matrix: full
$R[F^2 > 2\sigma(F^2)]$ = 0.048
$wR(F^2)$ = 0.140
S = 1.03
2609 reflections
167 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
H-atom parameters constrained
$w = 1/[\sigma^2(F_o^2) + (0.0696P)^2 + 0.6783P]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{max}$ = 0.001
$\Delta\rho_{max}$ = 0.44 e Å^{-3}
$\Delta\rho_{min}$ = −0.22 e Å^{-3}

Special details

Geometry. All e.s.d.'s (except the e.s.d. in the dihedral angle between two l.s. planes) are estimated using the full covariance matrix. The cell e.s.d.'s are taken into account individually in the estimation of e.s.d.'s in distances, angles and torsion angles; correlations between e.s.d.'s in cell parameters are only used when they are defined by crystal symmetry. An approximate (isotropic) treatment of cell e.s.d.'s is used for estimating e.s.d.'s involving l.s. planes.

Refinement. Refinement of F^2 against ALL reflections. The weighted R-factor wR and goodness of fit S are based on F^2, conventional R-factors R are based on F, with F set to zero for negative F^2. The threshold expression of $F^2 > \sigma(F^2)$ is used only for calculating R-factors(gt) *etc*. and is not relevant to the choice of reflections for refinement. R-factors based on F^2 are statistically about twice as large as those based on F, and R-factors based on ALL data will be even larger.

Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters (Å²)

	x	y	z	U_{iso}*/U_{eq}
Cl1	0.05896 (6)	0.43910 (4)	0.31130 (6)	0.0548 (2)
O1	0.64998 (19)	1.16127 (15)	0.74894 (19)	0.0700 (6)
O2	0.78794 (19)	0.86362 (13)	0.53807 (17)	0.0633 (5)
N1	0.92752 (17)	1.26424 (15)	0.43535 (17)	0.0452 (5)
H1D	0.9687	1.3101	0.3992	0.054*
N2	0.67191 (18)	1.06169 (14)	0.71219 (17)	0.0425 (4)
N3	0.73993 (17)	0.92068 (14)	0.61433 (16)	0.0395 (4)
C1	0.8082 (2)	1.23103 (17)	0.5864 (2)	0.0437 (5)
H1	0.7700	1.2586	0.6473	0.052*
C2	0.8735 (2)	1.30278 (18)	0.5247 (2)	0.0479 (6)
H2	0.8799	1.3784	0.5454	0.057*
C3	0.9190 (2)	1.15584 (18)	0.4007 (2)	0.0437 (5)
H3	0.9562	1.1321	0.3375	0.052*
C4	0.8550 (2)	1.07904 (17)	0.45867 (19)	0.0391 (5)
H4	0.8484	1.0045	0.4336	0.047*
C5	0.80009 (18)	1.11518 (16)	0.55597 (18)	0.0343 (4)
C6	0.73824 (19)	1.03513 (16)	0.62469 (18)	0.0348 (5)
C7	0.6107 (2)	0.95916 (19)	0.7557 (2)	0.0446 (5)
C8	0.6900 (2)	0.86274 (18)	0.7158 (2)	0.0479 (6)
C9	0.6147 (3)	0.9682 (3)	0.8936 (3)	0.0755 (9)
H9A	0.7009	0.9699	0.9387	0.113*
H9B	0.5727	0.9047	0.9193	0.113*
H9C	0.5729	1.0358	0.9093	0.113*
C10	0.4731 (3)	0.9600 (3)	0.6823 (3)	0.0760 (9)
H10A	0.4336	1.0290	0.6972	0.114*
H10B	0.4285	0.8981	0.7078	0.114*
H10C	0.4713	0.9533	0.5959	0.114*
C11	0.8094 (3)	0.8328 (3)	0.8176 (3)	0.0673 (8)
H11A	0.8630	0.7836	0.7837	0.101*
H11B	0.7839	0.7962	0.8849	0.101*
H11C	0.8546	0.9002	0.8465	0.101*

C12	0.6206 (4)	0.7561 (2)	0.6664 (3)	0.0855 (11)
H12A	0.5572	0.7731	0.5938	0.128*
H12B	0.5814	0.7245	0.7279	0.128*
H12C	0.6795	0.7031	0.6462	0.128*

Atomic displacement parameters (Å^2)

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
Cl1	0.0756 (5)	0.0354 (3)	0.0595 (4)	−0.0077 (2)	0.0278 (3)	0.0030 (2)
O1	0.0900 (14)	0.0420 (9)	0.0963 (15)	0.0035 (9)	0.0597 (12)	−0.0127 (9)
O2	0.0960 (14)	0.0354 (8)	0.0746 (12)	−0.0055 (8)	0.0536 (11)	−0.0104 (8)
N1	0.0455 (11)	0.0403 (10)	0.0500 (11)	−0.0080 (8)	0.0114 (9)	0.0098 (8)
N2	0.0454 (11)	0.0370 (9)	0.0502 (11)	0.0003 (7)	0.0215 (9)	−0.0023 (7)
N3	0.0471 (11)	0.0319 (8)	0.0438 (10)	−0.0033 (7)	0.0197 (8)	−0.0025 (7)
C1	0.0529 (14)	0.0357 (11)	0.0439 (12)	−0.0023 (9)	0.0134 (10)	−0.0043 (9)
C2	0.0575 (15)	0.0315 (10)	0.0526 (14)	−0.0059 (9)	0.0082 (11)	−0.0007 (9)
C3	0.0432 (13)	0.0431 (11)	0.0476 (12)	0.0019 (9)	0.0159 (10)	0.0056 (9)
C4	0.0428 (12)	0.0329 (10)	0.0439 (12)	−0.0007 (8)	0.0142 (10)	0.0002 (8)
C5	0.0331 (11)	0.0316 (9)	0.0378 (11)	−0.0006 (8)	0.0065 (8)	0.0010 (8)
C6	0.0356 (11)	0.0329 (10)	0.0377 (11)	−0.0004 (8)	0.0115 (9)	−0.0010 (8)
C7	0.0433 (13)	0.0459 (12)	0.0495 (13)	−0.0048 (9)	0.0206 (10)	0.0017 (9)
C8	0.0563 (14)	0.0375 (11)	0.0575 (14)	−0.0053 (10)	0.0288 (12)	0.0041 (9)
C9	0.102 (3)	0.0758 (19)	0.0605 (17)	0.0035 (17)	0.0428 (17)	0.0043 (14)
C10	0.0460 (16)	0.080 (2)	0.102 (3)	−0.0054 (14)	0.0163 (16)	0.0114 (17)
C11	0.0678 (18)	0.0642 (16)	0.0743 (18)	0.0131 (13)	0.0250 (15)	0.0262 (14)
C12	0.103 (3)	0.0554 (16)	0.116 (3)	−0.0362 (17)	0.064 (2)	−0.0199 (17)

Geometric parameters (Å, º)

O1—N2	1.295 (3)	C7—C9	1.529 (4)
O2—N3	1.285 (2)	C7—C10	1.536 (4)
N1—C2	1.343 (3)	C7—C8	1.559 (3)
N1—C3	1.346 (3)	C8—C12	1.518 (4)
N1—H1D	0.8600	C8—C11	1.563 (4)
N2—C6	1.371 (3)	C9—H9A	0.9600
N2—C7	1.522 (3)	C9—H9B	0.9600
N3—C6	1.370 (3)	C9—H9C	0.9600
N3—C8	1.523 (3)	C10—H10A	0.9600
C1—C2	1.387 (3)	C10—H10B	0.9600
C1—C5	1.421 (3)	C10—H10C	0.9600
C1—H1	0.9300	C11—H11A	0.9600
C2—H2	0.9300	C11—H11B	0.9600
C3—C4	1.392 (3)	C11—H11C	0.9600
C3—H3	0.9300	C12—H12A	0.9600
C4—C5	1.415 (3)	C12—H12B	0.9600
C4—H4	0.9300	C12—H12C	0.9600
C5—C6	1.475 (3)		

C2—N1—C3	121.81 (19)	C10—C7—C8	112.8 (2)
C2—N1—H1D	119.1	C12—C8—N3	110.0 (2)
C3—N1—H1D	119.1	C12—C8—C7	117.4 (2)
O1—N2—C6	126.78 (18)	N3—C8—C7	100.76 (18)
O1—N2—C7	120.83 (19)	C12—C8—C11	109.7 (3)
C6—N2—C7	112.09 (17)	N3—C8—C11	105.4 (2)
O2—N3—C6	126.73 (17)	C7—C8—C11	112.6 (2)
O2—N3—C8	120.85 (18)	C7—C9—H9A	109.5
C6—N3—C8	112.09 (17)	C7—C9—H9B	109.5
C2—C1—C5	119.6 (2)	H9A—C9—H9B	109.5
C2—C1—H1	120.2	C7—C9—H9C	109.5
C5—C1—H1	120.2	H9A—C9—H9C	109.5
N1—C2—C1	120.7 (2)	H9B—C9—H9C	109.5
N1—C2—H2	119.6	C7—C10—H10A	109.5
C1—C2—H2	119.6	C7—C10—H10B	109.5
N1—C3—C4	120.6 (2)	H10A—C10—H10B	109.5
N1—C3—H3	119.7	C7—C10—H10C	109.5
C4—C3—H3	119.7	H10A—C10—H10C	109.5
C3—C4—C5	119.5 (2)	H10B—C10—H10C	109.5
C3—C4—H4	120.2	C8—C11—H11A	109.5
C5—C4—H4	120.2	C8—C11—H11B	109.5
C4—C5—C1	117.69 (18)	H11A—C11—H11B	109.5
C4—C5—C6	121.17 (19)	C8—C11—H11C	109.5
C1—C5—C6	121.13 (19)	H11A—C11—H11C	109.5
N3—C6—N2	108.00 (17)	H11B—C11—H11C	109.5
N3—C6—C5	125.80 (18)	C8—C12—H12A	109.5
N2—C6—C5	126.18 (19)	C8—C12—H12B	109.5
N2—C7—C9	110.2 (2)	H12A—C12—H12B	109.5
N2—C7—C10	105.5 (2)	C8—C12—H12C	109.5
C9—C7—C10	109.9 (2)	H12A—C12—H12C	109.5
N2—C7—C8	101.19 (18)	H12B—C12—H12C	109.5
C9—C7—C8	116.3 (2)		

C3—N1—C2—C1	1.1 (3)	C6—N2—C7—C9	143.3 (2)
C5—C1—C2—N1	1.0 (3)	O1—N2—C7—C10	76.0 (3)
C2—N1—C3—C4	−1.1 (3)	C6—N2—C7—C10	−98.1 (2)
N1—C3—C4—C5	−0.9 (3)	O1—N2—C7—C8	−166.4 (2)
C3—C4—C5—C1	2.9 (3)	C6—N2—C7—C8	19.6 (2)
C3—C4—C5—C6	−176.24 (19)	O2—N3—C8—C12	−40.2 (3)
C2—C1—C5—C4	−2.9 (3)	C6—N3—C8—C12	146.0 (2)
C2—C1—C5—C6	176.2 (2)	O2—N3—C8—C7	−164.7 (2)
O2—N3—C6—N2	176.6 (2)	C6—N3—C8—C7	21.4 (2)
C8—N3—C6—N2	−10.0 (2)	O2—N3—C8—C11	78.0 (3)
O2—N3—C6—C5	−4.9 (3)	C6—N3—C8—C11	−95.8 (2)
C8—N3—C6—C5	168.50 (19)	N2—C7—C8—C12	−141.9 (2)
O1—N2—C6—N3	179.6 (2)	C9—C7—C8—C12	98.7 (3)
C7—N2—C6—N3	−6.8 (2)	C10—C7—C8—C12	−29.7 (3)
O1—N2—C6—C5	1.1 (4)	N2—C7—C8—N3	−22.6 (2)

C7—N2—C6—C5	174.72 (19)	C9—C7—C8—N3	−142.0 (2)
C4—C5—C6—N3	8.4 (3)	C10—C7—C8—N3	89.7 (2)
C1—C5—C6—N3	−170.7 (2)	N2—C7—C8—C11	89.3 (2)
C4—C5—C6—N2	−173.4 (2)	C9—C7—C8—C11	−30.1 (3)
C1—C5—C6—N2	7.5 (3)	C10—C7—C8—C11	−158.4 (2)
O1—N2—C7—C9	−42.7 (3)		

Hydrogen-bond geometry (Å, º)

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
N1—H1*D*···Cl1^{i}	0.86	2.17	3.028 (3)	174

Symmetry code: (i) *x*+1, *y*+1, *z*.