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[Oxalylbis(azanediyl)]bis{[amino(2-pyridyl)methylene]ammonium}

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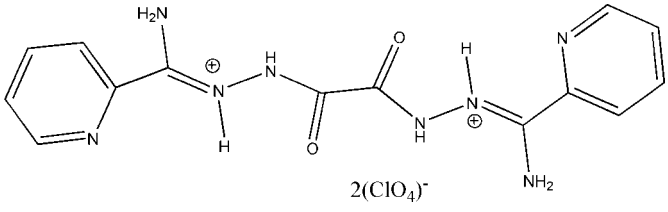
Key indicators: single-crystal X-ray study; T = 273 K; mean σ(C–C) = 0.004 Å; R factor = 0.045; wR factor = 0.128; data-to-parameter ratio = 11.8.

The title compound, $C_{14}H_{16}N_8O_2^{2+}\cdot 2ClO_4^-$, was prepared by reaction of bis[amino(2-pyridyl)methylene]oxalohydrazide with perchloric acid. The molecular symmetry is C_i and thus the asymmetric unit comprises one half-molecule. The dihedral angle between the aromatic ring and the plane of the oxamide group is 70.8 (3)°. The perchlorate anions and the cations are connected by intermolecular N—H···O hydrogen bonds.

Related literature

For background to the design and synthesis of polynuclear molecule-based magnetic materials, see: Niel *et al.* (2008); Zhao *et al.* (2004); Xu *et al.* (2001, 2003).



Experimental

Crystal data

$C_{14}H_{16}N_8O_2^{2+}\cdot 2ClO_4^-$
M_r = 527.25
Monoclinic, $P2_1/n$
a = 5.0751 (11) Å
b = 13.725 (3) Å
c = 15.162 (3) Å
β = 98.605 (3)°
V = 1044.2 (4) Å^3
Z = 2
Mo $K\alpha$ radiation
μ = 0.39 mm^{-1}
T = 273 K
0.31 × 0.25 × 0.22 mm

Data collection

Bruker SMART CCD area-detector diffractometer
Absorption correction: multi-scan (*SADABS*; Bruker, 2000)
T_{min} = 0.882, T_{max} = 0.914
5016 measured reflections
1824 independent reflections
1455 reflections with $I > 2\sigma(I)$
R_{int} = 0.078

Refinement

$R[F^2 > 2\sigma(F^2)]$ = 0.045
$wR(F^2)$ = 0.128
S = 1.09
1824 reflections
154 parameters
H-atom parameters constrained
$\Delta\rho_{max}$ = 0.33 e Å^{-3}
$\Delta\rho_{min}$ = −0.47 e Å^{-3}

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

D—H···A	D—H	H···A	D···A	D—H···A
N1—H1A···O4^{i}	0.86	2.51	2.975 (3)	115
N1—H1A···O5^{i}	0.86	2.58	3.404 (4)	162
N2—H2A···O3^{ii}	0.86	2.21	2.970 (3)	147
N4—H4A···O1^{iii}	0.86	2.23	2.974 (3)	145
N4—H4B···O2^{iii}	0.86	2.05	2.820 (3)	148

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

Data collection: *SMART* (Bruker, 2000); cell refinement: *SAINT* (Bruker, 2000); 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: *SHELXTL*.

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Supplementary data and figures for this paper are available from the IUCr electronic archives (Reference: KP2209).

References

Bruker (2000). *SADABS*, *SMART* and *SAINT*. Bruker AXS Inc., Madison, Wisconsin, USA.
Niel, V., Milway, V. A., Dawe, L. N. & Thompson, L. K. (2008). *Inorg. Chem.* **47**, 176–189.
Sheldrick, G. M. (2008). *Acta Cryst.* A**64**, 112–122.
Xu, Z. Q., Thompson, L. K., Black, D. A. & Miller, D. O. (2001). *Dalton Trans.* pp. 2042–2048.
Xu, Z. Q., Thompson, L. K., Milway, V. A., Zhao, L. & Miller, D. O. (2003). *Inorg. Chem.* **42**, 2950–2959.
Zhao, L., Niel, V., Thompson, L. K. & Xu, Z. Q. (2004). *Dalton Trans.* pp. 1446–1455.

supporting information

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[Oxalylbis(azanediyl)]bis{[amino(2-pyridyl)methylene]ammonium}

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S1. Comment

In recent years, researchers showed considerable interest in design and synthesis of polynuclear molecule-based magnetic materials, which were prepared by reactions of special organic molecules with transitional metals. (Niel *et al.*, 2008; Xu *et al.*, 2001; Xu *et al.*, 2003); Zhao *et al.*, 2004). Here we report a new compound, $[(C_{14}H_{16}N_8O_2)(ClO_4)_2]$.

The asymmetric unit of the title compound comprises a half of the molecule (Fig. 1). In the structure of title compound, the dihedral angle between the aromatic ring and the plane of oxamide group is 70.8 °. Perchlorate anions and cations are connected by intermolecular N—H···O hydrogen bonds (Fig. 2, Table 1).

S2. Experimental

All solvents and chemicals were of analytical grade and were used without further purification. Ligand was prepared by similar procedure reported in the literature (Zhao *et al.*, 2004). For the synthesis of title compoud, a solution of ligand (0.1 mmol), $HClO_4$(0.1 mmol) in 20 ml methanol was refluxed for 1 h, and then cooled to room temperature and filtered. Single crystals suitable for X-ray analysis were grown from the methanol solution by slow evaporation at room temperature in air. Anal. Calcd. for $C_{14}H_{16}N_8O_{10}Cl_2$: C, 31.89; H, 3.06; N, 21.25. Found: C, 32.15; H, 3.18; N, 21.20.

S3. Refinement

All hydrogen atoms were geomemetrically positioned (C—H 0.93–0.97 Å, N–H 0.86 Å) and refined as riding, with U_{iso}(H)=1.2 U_{eq} of the parent atom.

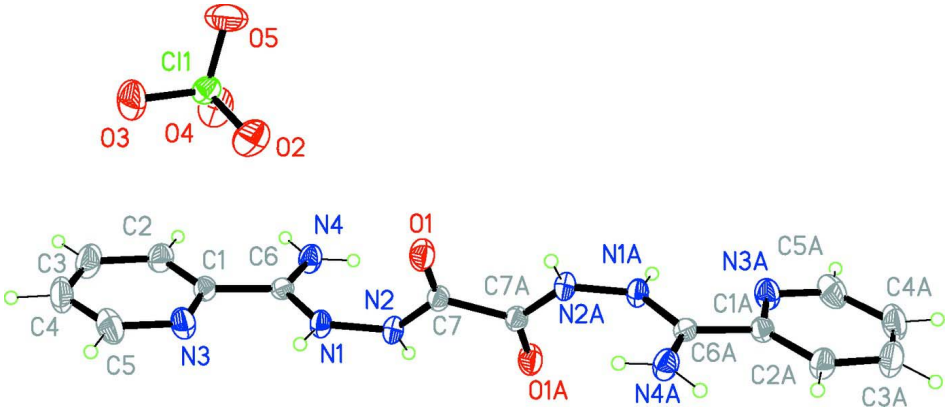

Figure 1

Molecular structure of the title compound, showing the 30% probability displacement ellipsoids and the atom-numbering [symmetry code: 1 - *x*,1 - *y*,1 - *z*].

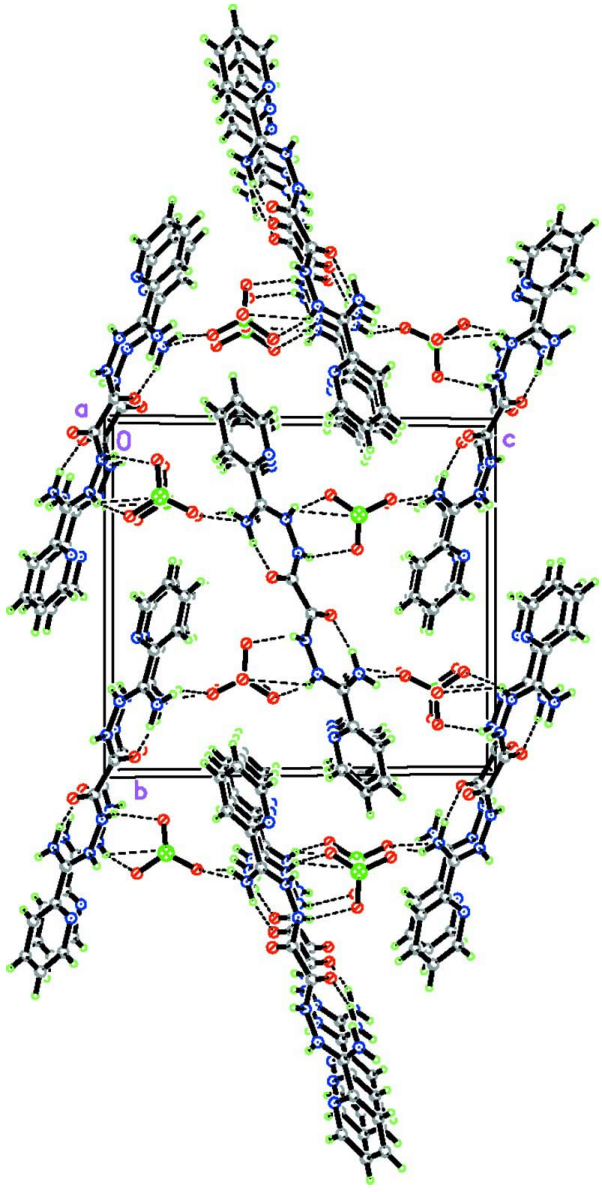

Figure 2

The crystal packing of the title compound generated by intermolecular hydrogen bonds.

[Oxalylbis(azanediyl)]bis{[amino(2-pyridyl)methylene]ammonium}

Crystal data

$C_{14}H_{16}N_8O_2^{2+}\cdot 2ClO_4^-$
$M_r = 527.25$
Monoclinic, $P2_1/n$
Hall symbol: -P 2yn
$a = 5.0751\ (11)$ Å
$b = 13.725\ (3)$ Å
$c = 15.162\ (3)$ Å
$\beta = 98.605\ (3)°$
$V = 1044.2\ (4)$ Å^3
$Z = 2$
$F(000) = 540$
$D_x = 1.677$ Mg m^{-3}
Mo $K\alpha$ radiation, $\lambda = 0.71073$ Å
Cell parameters from 2046 reflections
$\theta = 2.7–26.2°$
$\mu = 0.39$ mm^{-1}
$T = 273$ K
Block, colourless
0.31 × 0.25 × 0.22 mm

Data collection

Bruker SMART CCD area-detector diffractometer
Radiation source: fine-focus sealed tube
Graphite monochromator
φ and ω scans
Absorption correction: multi-scan (*SADABS*; Bruker, 2000)
T_{min} = 0.882, T_{max} = 0.914
5016 measured reflections
1824 independent reflections
1455 reflections with $I > 2\sigma(I)$
R_{int} = 0.078
θ_{max} = 25.0°, θ_{min} = 2.0°
h = −6→5
k = −13→16
l = −15→18

Refinement

Refinement on F^2
Least-squares matrix: full
$R[F^2 > 2\sigma(F^2)]$ = 0.045
$wR(F^2)$ = 0.128
S = 1.09
1824 reflections
154 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
$w = 1/[\sigma^2(F_o^2) + (0.076P)^2]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{max}$ = 0.001
$\Delta\rho_{max}$ = 0.33 e Å^{-3}
$\Delta\rho_{min}$ = −0.47 e Å^{-3}

Special details

Experimental. The structure was solved by direct methods (Bruker, 2000) and successive difference Fourier syntheses.
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}
C1	0.7001 (5)	0.85329 (16)	0.62706 (14)	0.0356 (5)
C2	0.8640 (5)	0.90729 (18)	0.68880 (16)	0.0487 (6)
H2	1.0229	0.8819	0.7183	0.058*
C3	0.7835 (7)	1.0013 (2)	0.70546 (19)	0.0654 (8)
H3	0.8887	1.0402	0.7469	0.078*
C4	0.5517 (8)	1.03584 (19)	0.6612 (2)	0.0679 (9)
H4	0.4953	1.0987	0.6719	0.081*
C5	0.4000 (6)	0.97677 (19)	0.6001 (2)	0.0604 (8)
H5	0.2409	1.0013	0.5698	0.073*
C6	0.7662 (4)	0.75221 (14)	0.60311 (13)	0.0326 (5)
C7	0.4737 (4)	0.54717 (15)	0.52444 (14)	0.0352 (5)
Cl1	0.54991 (12)	0.77141 (5)	0.85280 (3)	0.0450 (3)
N1	0.5942 (4)	0.71127 (13)	0.53978 (12)	0.0370 (5)
H1A	0.4530	0.7425	0.5175	0.044*
N2	0.6387 (4)	0.61898 (12)	0.50903 (11)	0.0380 (5)

H2A	0.7700	0.6079	0.4805	0.046*
N3	0.4704 (4)	0.88577 (14)	0.58226 (14)	0.0486 (5)
N4	0.9765 (4)	0.70721 (15)	0.64025 (12)	0.0445 (5)
H4A	1.0082	0.6489	0.6240	0.053*
H4B	1.0852	0.7355	0.6813	0.053*
O1	0.3033 (4)	0.55207 (11)	0.57270 (12)	0.0506 (5)
O2	0.4275 (4)	0.72826 (16)	0.77218 (13)	0.0710 (6)
O3	0.4490 (5)	0.86811 (15)	0.85821 (15)	0.0793 (7)
O4	0.8287 (4)	0.77292 (16)	0.85160 (15)	0.0730 (7)
O5	0.4956 (6)	0.71688 (18)	0.92652 (16)	0.0973 (9)

Atomic displacement parameters (Å²)

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
C1	0.0392 (13)	0.0310 (11)	0.0376 (11)	−0.0040 (9)	0.0085 (10)	−0.0016 (9)
C2	0.0529 (16)	0.0435 (14)	0.0490 (14)	−0.0095 (11)	0.0050 (11)	−0.0077 (11)
C3	0.085 (2)	0.0450 (16)	0.0675 (18)	−0.0155 (16)	0.0163 (17)	−0.0204 (14)
C4	0.096 (3)	0.0315 (15)	0.083 (2)	−0.0006 (15)	0.0361 (19)	−0.0080 (13)
C5	0.0628 (19)	0.0409 (15)	0.080 (2)	0.0110 (13)	0.0182 (15)	0.0084 (13)
C6	0.0362 (13)	0.0330 (11)	0.0287 (12)	−0.0032 (9)	0.0049 (9)	−0.0007 (9)
C7	0.0380 (13)	0.0308 (12)	0.0348 (12)	0.0014 (9)	−0.0012 (9)	−0.0029 (8)
Cl1	0.0409 (4)	0.0542 (4)	0.0375 (4)	0.0076 (3)	−0.0019 (3)	−0.0028 (2)
N1	0.0371 (11)	0.0292 (10)	0.0422 (10)	0.0005 (7)	−0.0027 (8)	−0.0062 (7)
N2	0.0419 (11)	0.0300 (10)	0.0419 (10)	−0.0024 (8)	0.0058 (8)	−0.0090 (8)
N3	0.0509 (13)	0.0354 (11)	0.0576 (12)	0.0061 (9)	0.0020 (10)	−0.0010 (9)
N4	0.0430 (12)	0.0399 (12)	0.0466 (12)	0.0047 (9)	−0.0069 (9)	−0.0093 (8)
O1	0.0580 (12)	0.0374 (9)	0.0614 (11)	−0.0018 (8)	0.0246 (9)	−0.0084 (8)
O2	0.0610 (14)	0.0874 (16)	0.0572 (12)	−0.0057 (10)	−0.0150 (9)	−0.0154 (10)
O3	0.0885 (16)	0.0567 (14)	0.0966 (15)	0.0244 (12)	0.0262 (13)	−0.0043 (11)
O4	0.0374 (12)	0.0868 (16)	0.0894 (16)	0.0060 (10)	−0.0079 (10)	−0.0147 (11)
O5	0.130 (2)	0.106 (2)	0.0648 (14)	0.0451 (16)	0.0439 (15)	0.0363 (13)

Geometric parameters (Å, º)

C1—N3	1.335 (3)	C7—O1	1.215 (3)
C1—C2	1.373 (3)	C7—N2	1.336 (3)
C1—C6	1.485 (3)	C7—C7^{i}	1.535 (4)
C2—C3	1.387 (4)	Cl1—O5	1.406 (2)
C2—H2	0.9300	Cl1—O2	1.415 (2)
C3—C4	1.351 (5)	Cl1—O4	1.418 (2)
C3—H3	0.9300	Cl1—O3	1.429 (2)
C4—C5	1.377 (5)	N1—N2	1.380 (2)
C4—H4	0.9300	N1—H1A	0.8600
C5—N3	1.338 (3)	N2—H2A	0.8600
C5—H5	0.9300	N4—H4A	0.8600
C6—N4	1.287 (3)	N4—H4B	0.8600
C6—N1	1.322 (3)		

N3—C1—C2	124.1 (2)	O1—C7—C7^{i}	121.9 (2)
N3—C1—C6	113.56 (19)	N2—C7—C7^{i}	112.3 (2)
C2—C1—C6	122.3 (2)	O5—Cl1—O2	110.53 (18)
C1—C2—C3	117.5 (3)	O5—Cl1—O4	109.43 (16)
C1—C2—H2	121.3	O2—Cl1—O4	107.75 (14)
C3—C2—H2	121.3	O5—Cl1—O3	109.51 (14)
C4—C3—C2	119.6 (3)	O2—Cl1—O3	108.86 (13)
C4—C3—H3	120.2	O4—Cl1—O3	110.74 (13)
C2—C3—H3	120.2	C6—N1—N2	120.78 (19)
C3—C4—C5	119.0 (3)	C6—N1—H1A	119.6
C3—C4—H4	120.5	N2—N1—H1A	119.6
C5—C4—H4	120.5	C7—N2—N1	118.65 (18)
N3—C5—C4	123.1 (3)	C7—N2—H2A	120.7
N3—C5—H5	118.4	N1—N2—H2A	120.7
C4—C5—H5	118.4	C1—N3—C5	116.6 (2)
N4—C6—N1	121.8 (2)	C6—N4—H4A	120.0
N4—C6—C1	123.0 (2)	C6—N4—H4B	120.0
N1—C6—C1	115.2 (2)	H4A—N4—H4B	120.0
O1—C7—N2	125.83 (19)		

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

Hydrogen-bond geometry (Å, º)

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
N1—H1*A*···O4^{ii}	0.86	2.51	2.975 (3)	115
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N4—H4*A*···O1^{iv}	0.86	2.23	2.974 (3)	145
N4—H4*B*···O2^{iv}	0.86	2.05	2.820 (3)	148

Symmetry codes: (ii) *x*−1/2, −*y*+3/2, *z*−1/2; (iii) *x*+1/2, −*y*+3/2, *z*−1/2; (iv) *x*+1, *y*, *z*.