metal-organic compounds

catena-Poly[[bis[3-(1H-imidazol-1-yl)-1-phenylpropan-1-one-κN^3]nickel(II)]-μ-oxalato-$\kappa^4 O^1,O^2$:$O^{1'},O^{2'}$]

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In the title compound, $[Ni(C_2O_4)(C_{12}H_{12}N_2O)_2]_n$, the Ni^{II} atom, lying on a twofold rotation axis, is coordinated by two N atoms from two monodentate 3-(1H-imidazol-1-yl)-1-phenyl-propan-1-one (L) ligands and four O atoms from two oxalate anions in a distorted octahedral geometry. The oxalate anion has a twofold rotation axis through the mid-point of the C—C bond and acts as a bridging ligand, linking the Ni^{II} atoms into a polymeric chain along [010]. Weak intermolecular C—H···O hydrogen bonds connect the chains, resulting in a three-dimensional supramolecular structure. >

Related literature

For background to the construction of metal-organic frame-works using a mixed-ligand strategy, see: Du et al. (2005); Tao et al. (2000); Ye et al. (2005).

Experimental

Crystal data

$[Ni(C_2O_4)(C_{12}H_{12}N_2O)_2]$	$V = 2374.3$ (3) Å^3
$M_r = 547.20$	$Z = 4$
Monoclinic, $C2/c$	Mo $K\alpha$ radiation
$a = 15.3065$ (11) Å	$\mu = 0.87$ mm^{-1}
$b = 5.6605$ (4) Å	$T = 296$ K
$c = 27.536$ (2) Å	$0.28 \times 0.22 \times 0.20$ mm
$\beta = 95.613$ (1)°	

Data collection

Bruker APEXII CCD diffractometer	5794 measured reflections
Absorption correction: multi-scan (SADABS; Sheldrick, 1996)	2105 independent reflections
	1676 reflections with $I > 2\sigma(I)$
$T_{min} = 0.793$, $T_{max} = 0.845$	$R_{int} = 0.043$

Refinement

$R[F^2 > 2\sigma(F^2)] = 0.045$	1 restraint
$wR(F^2) = 0.096$	H-atom parameters constrained
$S = 1.02$	$\Delta\rho_{max} = 0.63$ e Å^{-3}
2105 reflections	$\Delta\rho_{min} = -0.45$ e Å^{-3}
168 parameters	

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

D—H···A	D—H	H···A	D···A	D—H···A
C3—H3···O1^i	0.93	2.46	3.290 (4)	149
C4—H4B···O2^i	0.97	2.58	3.467 (5)	152
C10—H10···O2^{ii}	0.93	2.42	3.318 (5)	162

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

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: XP in SHELXTL (Sheldrick, 2008) and DIAMOND (Brandenburg & Berndt, 1999); software used to prepare material for publication: SHELXTL.

References

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supporting information

catena-Poly[[bis[3-(1*H*-imidazol-1-yl)-1-phenylpropan-1-one-*κN*³]nickel(II)]-*μ*-oxalato-*κ*⁴*O*¹,*O*²:*O*¹′,*O*²′]

Jian-Hua Guo

S1. Comment

Currently, the rational construction of new structurally defined metal-organic frameworks using a mixed-ligand strategy seems to be a marvelous success (Du *et al.*, 2005; Tao *et al.*, 2000; Ye *et al.*, 2005). In our recent research, we have initiated a synthetic approach employing multicarboxylates and 3-(1*H*-imidazol-1-yl)-1-phenylpropan-1-one (*L*) upon reactions with different metal ions to construct new functional frameworks. To explore this series, we synthesized the title compound, a new Ni(II) complex based on the *L* ligand.

In the title complex (Fig. 1), the Ni^{II} atom, lying on a twofold rotation axis, is six-coordinated in a distorted octahedral geometry by four O atoms from two oxalate anions in the equatorial plane and two N atoms from two monodentate *L* ligands occupying the axial positions, with the N1—Ni1—N1^i angle of 178.56 (17)° [symmetry code: (i) 1-*x*, *y*, 3/2-*z*]. As depicted in Fig. 2, the oxalate dianions as bridging ligands joint the Ni^{II} atoms into a one-dimensional polymeric [Ni(C$_2$O$_4$)]$_n$ chain along [0 1 0]. The imidazole and benzene rings in the *L* ligands are not coplanar, the dihedral angel between the imidazole and benzene rings being 68.7 (2)°. Analysis of the crystal packing indicates that weak intermolecular C—H⋯O hydrogen bonds (Table 1) connect the chains, producing a three-dimensional supramolecular structure.

S2. Experimental

Ni(CH$_3$CO$_2$)$_2$.4H$_2$O (24.9 mg, 0.1 mmol), 3-(1*H*-imidazol-1-yl)-1-phenylpropan-1-one (22.2 mg, 0.1 mmol) and oxalic acid were mixed in a CH$_3$CN/H$_2$O solution (20 ml, v/v = 1:1) with vigorous stirring for *ca* 30 min. The resulting solution was filtered and left to stand at room temperature. Green block crystals of the title compound suitable for X-ray analysis were obtained in 65% yield by slow evaporation of the solvent over a period of one week. Analysis, calculated for C$_{26}$H$_{24}$N$_4$NiO$_6$: C 57.07, H 4.42, N 10.24%; found: C 57.13, H 4.47, N 10.32%.

S3. Refinement

Although all H atoms were visible in difference Fourier maps, they were finally placed in geometrically calculated positions and refined as riding atoms, with C—H = 0.93 (CH) and 0.97 (CH$_2$) Å and with U_{iso}(H) = 1.2U_{eq}(C).
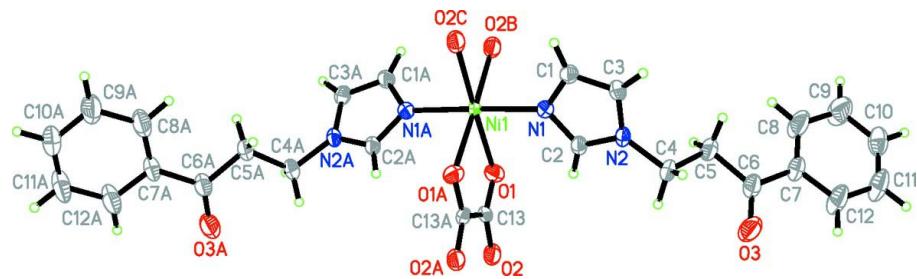
Figure 1

The asymmetric unit of the title compound, showing the 30% probability ellipsoids. [Symmetry codes: (A) 1-x, y, 3/2-z; (B) x, -1+y, z; (C) 1-x, -1+y, 3/2-z.]
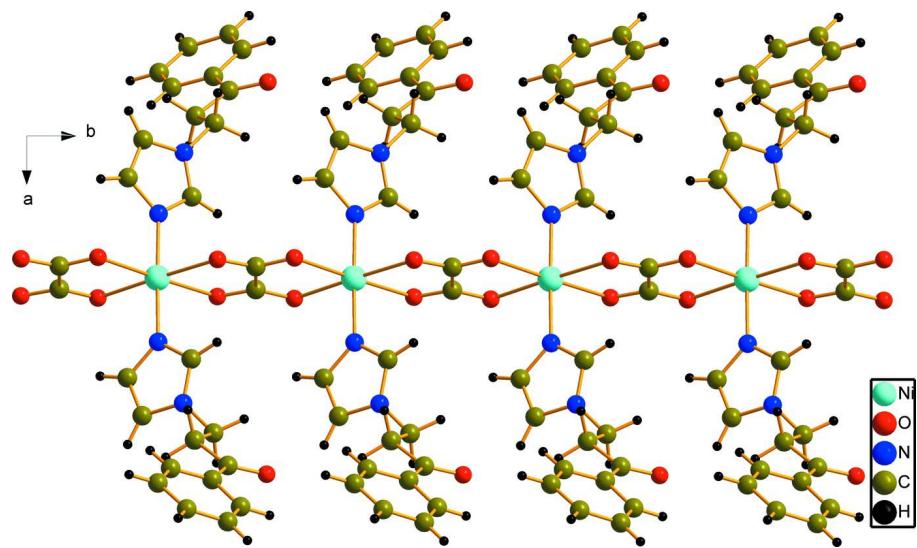
Figure 2

The one-dimensional structure of the title compound.

catena-Poly[[bis[3-(1*H*-imidazol-1-yl)-1-phenylpropan-1-one- *κN³*]nickel(II)]-*µ*-oxalato- *κ⁴O¹,O²:O¹′,O²′*]

Crystal data

[Ni(C₂O₄)(C₁₂H₁₂N₂O)₂]
$M_r = 547.20$
Monoclinic, *C*2/*c*
Hall symbol: -C 2yc
$a = 15.3065\,(11)$ Å
$b = 5.6605\,(4)$ Å
$c = 27.536\,(2)$ Å
$\beta = 95.613\,(1)°$
$V = 2374.3\,(3)$ Å³
$Z = 4$

$F(000) = 1136$
$D_x = 1.531$ Mg m⁻³
Mo *Kα* radiation, $\lambda = 0.71073$ Å
Cell parameters from 1107 reflections
$\theta = 2.9–21.3°$
$\mu = 0.87$ mm⁻¹
$T = 296$ K
Block, colorless
$0.28 \times 0.22 \times 0.20$ mm

Data collection

Bruker APEXII CCD
   diffractometer
Radiation source: fine-focus sealed tube
Graphite monochromator
φ and *ω* scans

Absorption correction: multi-scan
   (*SADABS*; Sheldrick, 1996)
$T_{min} = 0.793$, $T_{max} = 0.845$
5794 measured reflections
2105 independent reflections

1676 reflections with $I > 2\sigma(I)$	$h = -16 \rightarrow 18$
$R_{int} = 0.043$	$k = -6 \rightarrow 5$
$\theta_{max} = 25.0°$, $\theta_{min} = 2.7°$	$l = -32 \rightarrow 32$

Refinement

Refinement on F^2	Secondary atom site location: difference Fourier map
Least-squares matrix: full	
$R[F^2 > 2\sigma(F^2)] = 0.045$	Hydrogen site location: inferred from neighbouring sites
$wR(F^2) = 0.096$	
$S = 1.02$	H-atom parameters constrained
2105 reflections	$w = 1/[\sigma^2(F_o^2) + (0.0315P)^2 + 4.5895P]$
168 parameters	where $P = (F_o^2 + 2F_c^2)/3$
1 restraint	$(\Delta/\sigma)_{max} < 0.001$
Primary atom site location: structure-invariant direct methods	$\Delta\rho_{max} = 0.63$ e Å^{-3}
	$\Delta\rho_{min} = -0.45$ e Å^{-3}

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

	x	*y*	*z*	U_{iso}*/U_{eq}
Ni1	0.5000	−0.13748 (10)	0.7500	0.02887 (19)
O1	0.46495 (16)	0.1678 (4)	0.79304 (9)	0.0464 (6)
O2	0.46148 (17)	0.5597 (5)	0.79161 (9)	0.0528 (7)
O3	0.8709 (3)	0.4212 (6)	0.93819 (12)	0.0912 (12)
N1	0.62123 (18)	−0.1330 (5)	0.78683 (10)	0.0388 (7)
N2	0.73701 (18)	−0.0047 (5)	0.83297 (10)	0.0400 (7)
C1	0.6893 (2)	−0.2891 (6)	0.78450 (13)	0.0418 (9)
H1	0.6865	−0.4274	0.7662	0.050*
C2	0.6534 (2)	0.0357 (7)	0.81664 (12)	0.0421 (9)
H2	0.6216	0.1666	0.8252	0.050*
C3	0.7610 (2)	−0.2127 (7)	0.81268 (13)	0.0433 (9)
H3	0.8155	−0.2863	0.8173	0.052*
C4	0.7933 (2)	0.1504 (7)	0.86521 (13)	0.0497 (10)
H4A	0.7668	0.3059	0.8657	0.060*
H4B	0.8499	0.1668	0.8525	0.060*
C5	0.8066 (3)	0.0541 (8)	0.91673 (13)	0.0591 (12)
H5A	0.7498	0.0192	0.9279	0.071*
H5B	0.8394	−0.0925	0.9166	0.071*
C6	0.8547 (3)	0.2235 (9)	0.95153 (15)	0.0573 (11)
C7	0.8814 (3)	0.1443 (9)	1.00249 (14)	0.0572 (11)
C8	0.8604 (4)	−0.0672 (11)	1.02023 (18)	0.099 (2)
H8	0.8268	−0.1721	1.0002	0.119*
C9	0.8881 (5)	−0.1321 (11)	1.06808 (19)	0.115 (2)
H9	0.8725	−0.2788	1.0797	0.138*
C10	0.9371 (3)	0.0151 (11)	1.09733 (17)	0.0782 (15)
H10	0.9546	−0.0283	1.1294	0.094*
C11	0.9609 (4)	0.2238 (12)	1.08058 (18)	0.0918 (18)
H11	0.9964	0.3247	1.1006	0.110*
C12	0.9326 (4)	0.2895 (10)	1.03341 (16)	0.0855 (17)
H12	0.9488	0.4367	1.0222	0.103*
C13	0.4784 (2)	0.3633 (6)	0.77433 (12)	0.0357 (8)

Atomic displacement parameters (Å²)

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
Ni1	0.0301 (3)	0.0254 (3)	0.0291 (3)	0.000	−0.0074 (2)	0.000
O1	0.0468 (15)	0.0402 (14)	0.0522 (15)	0.0059 (12)	0.0044 (12)	0.0031 (11)
O2	0.0515 (16)	0.0561 (18)	0.0480 (16)	0.0131 (13)	−0.0091 (13)	−0.0136 (13)
O3	0.125 (3)	0.075 (3)	0.066 (2)	−0.038 (2)	−0.027 (2)	0.0008 (18)
N1	0.0409 (17)	0.0349 (16)	0.0391 (16)	0.0020 (14)	−0.0034 (13)	−0.0032 (14)
N2	0.0357 (17)	0.0447 (18)	0.0372 (16)	0.0003 (14)	−0.0086 (13)	−0.0031 (15)
C1	0.042 (2)	0.040 (2)	0.042 (2)	0.0066 (17)	0.0002 (17)	−0.0078 (17)
C2	0.039 (2)	0.043 (2)	0.041 (2)	0.0053 (17)	−0.0081 (17)	−0.0050 (18)
C3	0.033 (2)	0.048 (2)	0.048 (2)	0.0055 (17)	−0.0039 (17)	0.0025 (18)
C4	0.045 (2)	0.055 (2)	0.046 (2)	−0.008 (2)	−0.0118 (18)	−0.006 (2)
C5	0.065 (3)	0.060 (3)	0.048 (2)	−0.011 (2)	−0.017 (2)	−0.002 (2)
C6	0.056 (3)	0.067 (3)	0.046 (2)	−0.008 (2)	−0.007 (2)	−0.005 (2)
C7	0.053 (2)	0.073 (3)	0.042 (2)	−0.010 (2)	−0.0081 (19)	−0.010 (2)
C8	0.133 (5)	0.095 (4)	0.060 (3)	−0.046 (4)	−0.035 (3)	0.004 (3)
C9	0.169 (6)	0.106 (5)	0.062 (3)	−0.048 (5)	−0.038 (4)	0.019 (3)
C10	0.077 (3)	0.109 (4)	0.046 (3)	0.002 (3)	−0.010 (3)	−0.008 (3)
C11	0.084 (4)	0.134 (5)	0.053 (3)	−0.036 (4)	−0.014 (3)	−0.015 (3)
C12	0.093 (4)	0.110 (4)	0.050 (3)	−0.042 (3)	−0.010 (3)	−0.009 (3)
C13	0.0311 (18)	0.0335 (19)	0.0402 (19)	0.0031 (16)	−0.0089 (15)	0.0039 (17)

Geometric parameters (Å, º)

Ni1—N1	2.026 (3)	C4—H4A	0.9700
Ni1—N1^i	2.026 (3)	C4—H4B	0.9700
Ni1—O2^{ii}	2.175 (3)	C5—C6	1.497 (5)
Ni1—O2^{iii}	2.175 (3)	C5—H5A	0.9700
Ni1—O1	2.191 (3)	C5—H5B	0.9700
Ni1—O1^i	2.191 (3)	C6—C7	1.492 (6)
O1—C13	1.247 (4)	C7—C8	1.344 (7)
O2—C13	1.247 (4)	C7—C12	1.372 (6)
O3—C6	1.211 (5)	C8—C9	1.393 (7)
N1—C2	1.322 (4)	C8—H8	0.9300
N1—C1	1.372 (4)	C9—C10	1.337 (7)
N2—C2	1.334 (4)	C9—H9	0.9300
N2—C3	1.369 (4)	C10—C11	1.332 (7)
N2—C4	1.467 (4)	C10—H10	0.9300
C1—C3	1.352 (5)	C11—C12	1.380 (6)
C1—H1	0.9300	C11—H11	0.9300
C2—H2	0.9300	C12—H12	0.9300
C3—H3	0.9300	C13—C13^i	1.550 (7)
C4—C5	1.515 (5)		
N1—Ni1—N1^i	178.56 (17)	C5—C4—H4A	109.3
N1—Ni1—O2^{ii}	89.51 (10)	N2—C4—H4B	109.3
N1^i—Ni1—O2^{ii}	91.63 (11)	C5—C4—H4B	109.3

N1—Ni1—O2ⁱⁱⁱ	91.63 (11)	H4A—C4—H4B	108.0
N1ⁱ—Ni1—O2ⁱⁱⁱ	89.51 (11)	C6—C5—C4	112.4 (4)
O2ⁱⁱ—Ni1—O2ⁱⁱⁱ	75.98 (14)	C6—C5—H5A	109.1
N1—Ni1—O1	88.86 (10)	C4—C5—H5A	109.1
N1ⁱ—Ni1—O1	90.01 (11)	C6—C5—H5B	109.1
O2ⁱⁱ—Ni1—O1	178.36 (9)	C4—C5—H5B	109.1
O2ⁱⁱⁱ—Ni1—O1	104.07 (9)	H5A—C5—H5B	107.8
N1—Ni1—O1ⁱ	90.01 (11)	O3—C6—C7	121.2 (4)
N1ⁱ—Ni1—O1ⁱ	88.86 (10)	O3—C6—C5	120.0 (4)
O2ⁱⁱ—Ni1—O1ⁱ	104.07 (9)	C7—C6—C5	118.8 (4)
O2ⁱⁱⁱ—Ni1—O1ⁱ	178.36 (9)	C8—C7—C12	116.8 (4)
O1—Ni1—O1ⁱ	75.92 (13)	C8—C7—C6	123.8 (4)
C13—O1—Ni1	114.7 (2)	C12—C7—C6	119.4 (5)
C13—O2—Ni1ⁱᵛ	115.1 (2)	C7—C8—C9	121.1 (5)
C2—N1—C1	104.8 (3)	C7—C8—H8	119.4
C2—N1—Ni1	125.9 (2)	C9—C8—H8	119.4
C1—N1—Ni1	129.1 (2)	C10—C9—C8	120.4 (6)
C2—N2—C3	107.3 (3)	C10—C9—H9	119.8
C2—N2—C4	126.1 (3)	C8—C9—H9	119.8
C3—N2—C4	126.6 (3)	C11—C10—C9	120.0 (5)
C3—C1—N1	110.2 (3)	C11—C10—H10	120.0
C3—C1—H1	124.9	C9—C10—H10	120.0
N1—C1—H1	124.9	C10—C11—C12	119.6 (5)
N1—C2—N2	111.8 (3)	C10—C11—H11	120.2
N1—C2—H2	124.1	C12—C11—H11	120.2
N2—C2—H2	124.1	C7—C12—C11	122.0 (5)
C1—C3—N2	105.9 (3)	C7—C12—H12	119.0
C1—C3—H3	127.0	C11—C12—H12	119.0
N2—C3—H3	127.0	O2—C13—O1	125.8 (3)
N2—C4—C5	111.6 (3)	O2—C13—C13ⁱ	116.9 (2)
N2—C4—H4A	109.3	O1—C13—C13ⁱ	117.3 (2)
N1—Ni1—O1—C13	91.5 (2)	C2—N2—C4—C5	105.2 (4)
N1ⁱ—Ni1—O1—C13	−87.6 (2)	C3—N2—C4—C5	−77.3 (5)
O2ⁱⁱⁱ—Ni1—O1—C13	−177.1 (2)	N2—C4—C5—C6	−173.1 (3)
O1ⁱ—Ni1—O1—C13	1.18 (18)	C4—C5—C6—O3	7.0 (7)
O2ⁱⁱ—Ni1—N1—C2	170.1 (3)	C4—C5—C6—C7	−173.5 (4)
O2ⁱⁱⁱ—Ni1—N1—C2	−114.0 (3)	O3—C6—C7—C8	175.5 (5)
O1—Ni1—N1—C2	−9.9 (3)	C5—C6—C7—C8	−4.0 (7)
O1ⁱ—Ni1—N1—C2	66.0 (3)	O3—C6—C7—C12	−6.5 (7)
O2ⁱⁱ—Ni1—N1—C1	−5.1 (3)	C5—C6—C7—C12	173.9 (5)
O2ⁱⁱⁱ—Ni1—N1—C1	70.8 (3)	C12—C7—C8—C9	1.3 (9)
O1—Ni1—N1—C1	174.9 (3)	C6—C7—C8—C9	179.3 (6)
O1ⁱ—Ni1—N1—C1	−109.2 (3)	C7—C8—C9—C10	−0.6 (11)
C2—N1—C1—C3	−0.1 (4)	C8—C9—C10—C11	−1.0 (10)
Ni1—N1—C1—C3	175.9 (2)	C9—C10—C11—C12	1.8 (9)
C1—N1—C2—N2	0.1 (4)	C8—C7—C12—C11	−0.5 (9)
Ni1—N1—C2—N2	−176.0 (2)	C6—C7—C12—C11	−178.6 (5)

C3—N2—C2—N1	−0.1 (4)	C10—C11—C12—C7	−1.1 (9)
C4—N2—C2—N1	177.8 (3)	Ni1^{iv}—O2—C13—O1	177.3 (3)
N1—C1—C3—N2	0.0 (4)	Ni1^{iv}—O2—C13—C13^{i}	−1.6 (4)
C2—N2—C3—C1	0.1 (4)	Ni1—O1—C13—O2	178.1 (3)
C4—N2—C3—C1	−177.8 (3)	Ni1—O1—C13—C13^{i}	−3.0 (4)

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

Hydrogen-bond geometry (Å, º)

D—H⋯A	D—H	H⋯A	D⋯A	D—H⋯A
C3—H3⋯O1^{v}	0.93	2.46	3.290 (4)	149
C4—H4B⋯O2^{v}	0.97	2.58	3.467 (5)	152
C10—H10⋯O2^{vi}	0.93	2.42	3.318 (5)	162

Symmetry codes: (v) x+1/2, y−1/2, z; (vi) −x+3/2, −y+1/2, −z+2.