organic compounds

N,N'-Bis(2-acetylphenyl)ethanediamide

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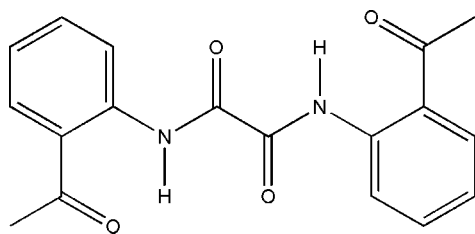Correspondence e-mail: jjasinski@keene.edu

Two new polymorphs of N,N'-bis(2-acetylphenyl)ethanediamide, $C_{18}H_{16}N_2O_4$, are reported, one crystallizing in the space group $P2_1/n$, in the present paper, and one in $P\bar{1}$ [Brewer, Jasinski, Butcher & Scheidt, (2007). *Acta Cryst.* E**63**, o4889]. In both structures, the 2-acetyl group is twisted slightly away from the plane of the benzene ring. In the present paper, this polymorph crystallizes with one-half of the molecule in the asymmetric unit, as the molecule lies on an inversion centre located at the centroid of the ethanamide system. The molecule adopts a *trans* ethanediamide (oxanilide) configuration, whereby the amide H atoms are bifurcated intramolecular hydrogen-bonding donors to oxamide and acetyl O atoms. The crystal packing is stabilized by intermolecular C—H···Cg packing interactions.

Related literature

For the $P\bar{1}$ polymorph, see: Brewer et al. (2007). For the synthesis of the title compound, see: Black *et al.* (1982); Black & Moss, 1987). For related structures, see: Liu *et al.* (2007); Jimenez *et al.* (2007); Lloret *et al.* (1996); Martinez-Martinez *et al.* (1993, 1998); Sun *et al.* (2006). For related literature see: Coucouvanis *et al.* (1990).

Experimental

Crystal data

$C_{18}H_{16}N_2O_4$	$V = 762.10$ (9) $Å^3$
$M_r = 324.33$	$Z = 2$
Monoclinic, $P2_1/n$	Mo $K\alpha$ radiation
$a = 6.9062$ (5) Å	$\mu = 0.10$ mm^{-1}
$b = 11.8955$ (8) Å	$T = 130$ K
$c = 9.6702$ (7) Å	$0.5 \times 0.5 \times 0.4$ mm
$\beta = 106.402$ (1)°	

Data collection

Bruker SMART CCD area-detector diffractometer	4600 measured reflections
	1762 independent reflections
Absorption correction: multi-scan (*SADABS*; Sheldrick, 1996)	1606 reflections with $I > 2\sigma(I)$
$T_{min} = 0.702$, $T_{max} = 0.962$	$R_{int} = 0.018$

Refinement

$R[F^2 > 2\sigma(F^2)] = 0.039$	105 parameters
$wR(F^2) = 0.111$	H-atom parameters constrained
$S = 1.12$	$\Delta\rho_{max} = 0.32$ e Å^{-3}
1762 reflections	$\Delta\rho_{min} = -0.28$ e Å^{-3}

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

$Cg1$ is the centroid of the C1–C6 benzene ring.

$D—H···A$	$D—H$	$H···A$	$D···A$	$D—H···A$
N1—H1A···O2	0.88	1.91	2.6315 (12)	138
N1—H1A···O1[i]	0.88	2.22	2.6615 (12)	111
C9—H9B···O1[ii]	0.98	2.55	3.4525 (14)	153
C9—H9A···Cg1[iii]	0.98	2.98	3.497 (1)	113
C9—H9C···Cg1[iii]	0.98	2.98	3.497 (1)	105

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

Data collection: *SMART* (Bruker, 2000); cell refinement: *SAINT* (Bruker, 2006); data reduction: *SAINT*; program(s) used to solve structure: *SHELXS97* (Sheldrick, 1990); program(s) used to refine structure: *SHELXL97* (Sheldrick, 1997); molecular graphics: *SHELXTL* (Bruker, 2000); software used to prepare material for publication: *SHELXTL*.

References

Black, D., Bos Vanderzalm, C. H. & Wong, L. C. H. (1982). *Aust. J. Chem.* **35**, 2435-2443.
Black, D. & Moss, G. I. (1987). *Aust. J. Chem.* **40**, 129-42.
Brewer, G., Jasinski, J. P., Butcher, R. J. & Scheidt, W. R. (2007). *Acta Cryst.* E**63**, o4889.
Bruker (2000). *SMART* and *SHELXTL*. Bruker AXS Inc., Madison, Wisconsin, USA.
Bruker (2006). *SAINT*. Bruker AXS Inc., Madison, Wisconsin, USA.
Coucouvanis, D., Christodoulou, D. & Kanatzidis, M. G. (1990). *Inorg. Chem.* **29**, 191–201.
Jimenez, C. A., Belmar, J. B., Delgado, F. S., Julve, M. & Ruiz-Perez, C. (2007). *CrystEngComm*, **9**, 746–752.
Liu, C. M., Zhang, J. Y., Zhang, D. Q., Gao, S. & Zhu, D. B. (2007). *CrystEngComm*, **9**, 799–805.
Lloret, F., Sanz, J. L., Cervera, B., Ruiz, R., Bois, C., Faus, J. & Julve, M. (1996). *J. Chem. Soc. Dalton Trans.* pp. 1359–1366.

organic compounds

Martinez-Martinez, F. J., Ariza-Castolo, A., Tlahuext, H., Tlahuextl, M. & Contreras, R. (1993). *J. Chem. Soc. Perkin Trans. 2*, pp. 1481–1485.

Martinez-Martinez, F. J., Padilla-Martinez, I. I., Brito, M. A., Geniz, E. D., Rojas, R. C., Saavedra, J. B. R., Höpfl, H., Tlahuextl, M. & Contreras, R. (1998). *J. Chem. Soc. Perkin Trans. 2*, pp. 401–406.

Sheldrick, G. M. (1990). *Acta Cryst.* A**46**, 467–473.

Sheldrick, G. M. (1996). *SADABS*. University of Göttingen, Germany.

Sheldrick, G. M. (1997). *SHELXL97*. University of Göttingen, Germany.

Sun, F., Li, Y.-T., Wu, Z.-Y., Song, Y.-L. & Jiang, M. (2006). *Acta Cryst.* C**62**, m584–m586.

supplementary materials

N,N'-Bis(2-acetylphenyl)ethanediamide

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Comment

Bis oxamides are precursors to both mono- and dinuclear metal complexes that can bind in both *cis* and *trans* conformations of the ligands (Liu *et al.* 2007; Jimenez *et al.* 2007; Lloret *et al.* 1996; Sun *et al.* 2006). In addition the reaction of bis oxamides, which contain two aldehyde or ketone groups, with bifunctional amines (with or without a templating metal) can afford macrocycles, which exhibit a *cis* geometry (Coucouvanis *et al.* 1990). The factors that control the outcome of reaction to form acylic (*cis* and *trans*) or macrocyclic (*cis*) ligands in part depend on the factors that determine the conformation of the parent oxamide. In order to form *cis* complexes, either acylic or macrocyclic, the parent *trans* oxamide must undergo a change in conformation to *cis*. The structures of the parent *trans* oxamides and an understanding of the factors that stabilize them are important to the further development of these versatile synthetic precursors. The title oxamide is symmetric about the C—C bond of the oxamide unit, RNH-(O)C—C(O)-NHR. In both polymorphs the amide proton is hydrogen bound to both the oxamide and acetyl oxygen atoms as has been observed in the structure of related oxamides (Martinez-Martinez *et al.* 1993, 1998). In view of the importance of these bis oximide derivatives, the present paper reports the crystal structure of one of the two polymorphs of the title compound, $C_{18}H_{16}N_2O_4$, which crystalizes in the spacegroup P 2_1/n, (Fig.1). The second polymorph crystallizes in the spacegroup P −1 as outlined in the following paper [Brewer *et al.* (2007). Acta Cryst E63, xxxx].

In both polymorphs the 2-acetyl group is twisted slightly away from the plane of the phenyl ring as evidenced by their respective torsion angles [C1–C2–C8–C9 (170.26 (9)°) in this polymorph; C1A–C2A–C8A–C9A (175.20 (12)°) and C1B–C2B–C8B–C9B (179.63 (13)°) in the second polymorph]. In the present paper this polymorph crystallizes with one-half of the molecule in the asymmetric unit as the molecule lies about an inversion centre located at the centroid of the ethanediamide system. This molecule adopts a *trans* ethanediamide (oxanilide) conformation, whereby, the amide hydrogen atoms are bifurcated intramolecular hydrogen bonding donors to oxamide and acetyl oxygen atoms [N1—H1A···O1[i]; i = −x, −y + 1, −z + 2]. In the present paper crystal packing is stabilized by intermolecular C9—H9A···*Cg*1 packing interactions (Fig. 2). [*Cg*1 = center of gravity of a phenyl ring, C1–C6]. In spite of crystallizing in different space groups [(I) in P 2_1/n, (II) in P −1] both compounds are nearly isostructural.

Experimental

The title molecule was synthesized by the condensation of oxalyl chloride and 2-aminoacetophenone as described previously (Black *et al.*, 1982; Black & Moss, 1987). The resulting solid was recrystallized from *N,N*-dimethylformamide (DMF) and gave two polymorphs, orange-brown irregular bocks [P 2_1/n, (I)] and colorless needles [P −1. (II)] which were analyzed by *x*-ray diffraction.

supplementary materials

Refinement

The amide hydrogen atom (H2A) was located in a difference Fourier map and along with all other H atoms were placed in their calculated positions and then refined using the riding model with N—H = 0.88 Å; C—H = 0.95 to 0.98 Å, and $U_{iso}(H) = 1.18–1.50U_{eq}(C,N)$.

Figures
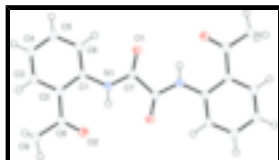

Fig. 1. Molecular structure of $C_{18}H_{16}N_2O_4$, (I), showing atom labeling and 50% probability displacement ellipsoids. Labelled atoms are related to unlabelled atoms by the symmetry operation $-x, -y + 1, -z + 2$.
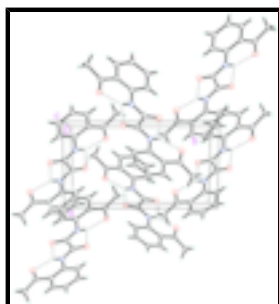

Fig. 2. Packing diagram of (I), viewed down the *c* axis. Dashed lines indicate C–H···O intermolecular and N–H···O intramolecular hydrogen bonds.

N,N'-bis(2-acetylphenyl) ethanediamide

Crystal data

$C_{18}H_{16}N_2O_4$	$F_{000} = 340$
$M_r = 324.33$	$D_x = 1.413$ Mg m^{-3}
Monoclinic, $P2_1/n$	Mo $K\alpha$ radiation $\lambda = 0.71073$ Å
Hall symbol: -P 2yn	Cell parameters from 3118 reflections
$a = 6.9062$ (5) Å	$\theta = 2.8–28.3°$
$b = 11.8955$ (8) Å	$\mu = 0.10$ mm^{-1}
$c = 9.6702$ (7) Å	$T = 130$ K
$\beta = 106.402$ (1)°	Block, orange–brown
$V = 762.10$ (9) Å^3	$0.5 \times 0.5 \times 0.4$ mm
$Z = 2$	

Data collection

Bruker SMART CCD area-detector diffractometer	1762 independent reflections
Radiation source: fine-focus sealed tube	1606 reflections with $I > 2\sigma(I)$
Monochromator: graphite	$R_{int} = 0.018$
$T = 130$ K	$\theta_{max} = 28.3°$

φ and ω scans

Absorption correction: multi-scan (SADABS; Sheldrick, 1996)

$T_{min} = 0.702$, $T_{max} = 0.962$

4600 measured reflections

$\theta_{min} = 2.8°$

$h = -9 \rightarrow 9$

$k = -11 \rightarrow 15$

$l = -9 \rightarrow 12$

Refinement

Refinement on F^2

Least-squares matrix: full

$R[F^2 > 2\sigma(F^2)] = 0.039$

$wR(F^2) = 0.111$

$S = 1.12$

1762 reflections

105 parameters

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

$w = 1/[\sigma^2(F_o^2) + (0.0624P)^2 + 0.1553P]$
where $P = (F_o^2 + 2F_c^2)/3$

$(\Delta/\sigma)_{max} = <0.001$

$\Delta\rho_{max} = 0.32$ e Å^{-3}

$\Delta\rho_{min} = -0.27$ e Å^{-3}

Extinction correction: none

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 $F^2 > 2sigma(F^2)$ 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 (Å^2)

	x	y	z	$U_{iso}*/U_{eq}$
N1	0.19444 (13)	0.55957 (7)	0.93637 (9)	0.0193 (2)*
H1A	0.1919	0.6224	0.9838	0.023*
O1	0.03494 (11)	0.38702 (6)	0.89371 (8)	0.0240 (2)
C1	0.33539 (15)	0.55559 (9)	0.85690 (10)	0.0184 (2)
O2	0.33333 (13)	0.76212 (7)	1.01843 (9)	0.0304 (2)
C2	0.46463 (15)	0.64932 (9)	0.86234 (11)	0.0192 (2)
C3	0.60129 (15)	0.64574 (9)	0.77928 (11)	0.0216 (2)
H3A	0.6869	0.7085	0.7805	0.026*
C4	0.61485 (16)	0.55348 (10)	0.69564 (11)	0.0233 (3)
H4A	0.7073	0.5533	0.6392	0.028*
C5	0.49177 (16)	0.46099 (9)	0.69504 (12)	0.0228 (3)
H5A	0.5028	0.3966	0.6396	0.027*
C6	0.35294 (15)	0.46151 (9)	0.77436 (11)	0.0206 (2)

H6A	0.2694	0.3978	0.7727	0.025*
C7	0.06217 (15)	0.47977 (9)	0.94969 (10)	0.0187 (2)
C8	0.46247 (16)	0.74933 (9)	0.95502 (11)	0.0213 (2)
C9	0.62608 (17)	0.83580 (10)	0.97243 (12)	0.0265 (3)
H9A	0.6074	0.8951	1.0378	0.040*
H9B	0.6197	0.8686	0.8783	0.040*
H9C	0.7579	0.8001	1.0125	0.040*

Atomic displacement parameters (Å^2)

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
O1	0.0246 (4)	0.0221 (4)	0.0275 (4)	−0.0041 (3)	0.0111 (3)	−0.0050 (3)
C1	0.0165 (5)	0.0210 (5)	0.0171 (5)	0.0011 (4)	0.0037 (4)	0.0020 (4)
O2	0.0336 (5)	0.0231 (4)	0.0407 (5)	−0.0042 (3)	0.0207 (4)	−0.0067 (3)
C2	0.0180 (5)	0.0203 (5)	0.0183 (5)	0.0007 (4)	0.0037 (4)	0.0023 (4)
C3	0.0184 (5)	0.0237 (5)	0.0222 (5)	−0.0003 (4)	0.0049 (4)	0.0042 (4)
C4	0.0204 (5)	0.0296 (6)	0.0213 (5)	0.0035 (4)	0.0083 (4)	0.0034 (4)
C5	0.0225 (5)	0.0249 (5)	0.0209 (5)	0.0032 (4)	0.0061 (4)	−0.0014 (4)
C6	0.0190 (5)	0.0218 (5)	0.0205 (5)	−0.0007 (4)	0.0047 (4)	0.0003 (4)
C7	0.0165 (5)	0.0213 (5)	0.0179 (5)	0.0006 (4)	0.0042 (4)	0.0003 (4)
C8	0.0222 (5)	0.0194 (5)	0.0215 (5)	0.0001 (4)	0.0047 (4)	0.0029 (4)
C9	0.0267 (6)	0.0248 (5)	0.0274 (6)	−0.0057 (4)	0.0066 (4)	−0.0024 (4)

Geometric parameters (Å, °)

N1—C7	1.3486 (14)	C4—C5	1.3892 (16)
N1—C1	1.4010 (13)	C4—H4A	0.9500
N1—H1A	0.8800	C5—C6	1.3867 (15)
O1—C7	1.2200 (13)	C5—H5A	0.9500
C1—C6	1.3996 (14)	C6—H6A	0.9500
C1—C2	1.4199 (14)	C7—C7^i	1.545 (2)
O2—C8	1.2261 (13)	C8—C9	1.5019 (15)
C2—C3	1.4021 (14)	C9—H9A	0.9800
C2—C8	1.4922 (14)	C9—H9B	0.9800
C3—C4	1.3821 (15)	C9—H9C	0.9800
C3—H3A	0.9500		
C7—N1—C1	128.52 (9)	C4—C5—H5A	119.6
C7—N1—H1A	115.7	C5—C6—C1	120.25 (10)
C1—N1—H1A	115.7	C5—C6—H6A	119.9
C6—C1—N1	121.70 (9)	C1—C6—H6A	119.9
C6—C1—C2	119.62 (10)	O1—C7—N1	128.17 (9)
N1—C1—C2	118.68 (9)	O1—C7—C7^i	121.18 (11)
C3—C2—C1	118.21 (10)	N1—C7—C7^i	110.66 (11)
C3—C2—C8	119.40 (9)	O2—C8—C2	122.10 (10)
C1—C2—C8	122.38 (9)	O2—C8—C9	119.31 (10)
C4—C3—C2	121.81 (10)	C2—C8—C9	118.59 (10)
C4—C3—H3A	119.1	C8—C9—H9A	109.5
C2—C3—H3A	119.1	C8—C9—H9B	109.5

C3—C4—C5	119.26 (10)	H9A—C9—H9B	109.5
C3—C4—H4A	120.4	C8—C9—H9C	109.5
C5—C4—H4A	120.4	H9A—C9—H9C	109.5
C6—C5—C4	120.79 (10)	H9B—C9—H9C	109.5
C6—C5—H5A	119.6		
C7—N1—C1—C6	1.17 (16)	C4—C5—C6—C1	−0.21 (16)
C7—N1—C1—C2	−178.07 (9)	N1—C1—C6—C5	178.94 (9)
C6—C1—C2—C3	2.47 (15)	C2—C1—C6—C5	−1.83 (16)
N1—C1—C2—C3	−178.27 (8)	C1—N1—C7—O1	−1.00 (18)
C6—C1—C2—C8	−176.24 (9)	C1—N1—C7—C7ⁱ	179.18 (10)
N1—C1—C2—C8	3.02 (15)	C3—C2—C8—O2	172.52 (10)
C1—C2—C3—C4	−1.15 (15)	C1—C2—C8—O2	−8.79 (16)
C8—C2—C3—C4	177.60 (9)	C3—C2—C8—C9	−8.44 (14)
C2—C3—C4—C5	−0.85 (16)	C1—C2—C8—C9	170.26 (9)
C3—C4—C5—C6	1.56 (16)		

Symmetry codes: (i) −*x*, −*y*+1, −*z*+2.

Hydrogen-bond geometry (Å, °)

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
N1—H1A···O2	0.88	1.91	2.6315 (12)	138
N1—H1A···O1ⁱ	0.88	2.22	2.6615 (12)	111
C9—H9B···O1ⁱⁱ	0.98	2.55	3.4525 (14)	153
C9—H9A···Cg1ⁱⁱⁱ	0.98	2.98	3.497 (1)	113
C9—H9C···Cg1ⁱⁱⁱ	0.98	2.98	3.497 (1)	105

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

Fig. 1

Fig. 2