organic compounds

2-(4-Methylanilino)acetohydrazide

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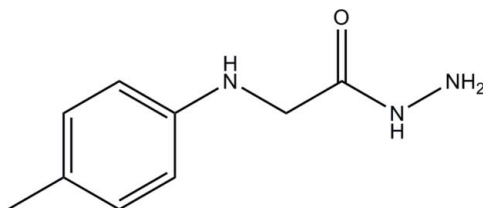Correspondence e-mail: hkfun@usm.my

In the title molecule, $C_9H_{13}N_3O$, the non-hydrogen atoms of the hydrazide group are essentially planar [maximum deviation = 0.028 (1) Å for one of the N atoms]. The mean plane of this group forms a dihedral angle of 83.34 (5)° with the plane of the benzene ring. In the crystal structure, molecules are linked by intermolecular N—H···O, N—H···N and weak C—H···N hydrogen bonds into a two-dimensional network parallel to the ab plane. Additional stabilization is provided by a weak C—H···π interaction.

Related literature

For the biological activity of hydrazide derivatives, see: Ozdemir *et al.* (2009); Khattab (2005). For synthetic applications, see: Isloor *et al.* (2009); Holla & Udupa (1992). For a related structure, see: Zhang & Shi (2009). For the stability of the temperature controller used for the data collection, see: Cosier & Glazer (1986).

Experimental

Crystal data

$C_9H_{13}N_3O$	c = 15.4756 (4) Å
M_r = 179.22	α = 87.002 (2)°
Triclinic, $P\bar{1}$	β = 84.282 (2)°
a = 5.1481 (1) Å	γ = 82.849 (2)°
b = 5.9262 (2) Å	V = 465.76 (2) Å³

Z = 2	T = 100 K
Mo $K\alpha$ radiation	0.52 × 0.15 × 0.07 mm
μ = 0.09 mm^{-1}	

Data collection

Bruker SMART APEXII CCD area-detector diffractometer	11697 measured reflections
	2703 independent reflections
Absorption correction: multi-scan ($SADABS$; Bruker, 2005)	2301 reflections with $I > 2\sigma(I)$
T_{min} = 0.956, T_{max} = 0.994	R_{int} = 0.026

Refinement

$R[F^2 > 2\sigma(F^2)]$ = 0.039	H atoms treated by a mixture of independent and constrained refinement
$wR(F^2)$ = 0.110	
S = 1.03	
2703 reflections	$\Delta\rho_{max}$ = 0.34 e Å^{-3}
170 parameters	$\Delta\rho_{min}$ = −0.20 e Å^{-3}

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

D—H···A	D—H	H···A	D···A	D—H···A
N1—H1N1···O1^i	0.875 (17)	2.162 (17)	3.0271 (11)	170.0 (13)
N2—H1N2···N3^{ii}	0.887 (17)	2.287 (16)	3.0302 (12)	141.3 (13)
N3—H1N3···O1^{iii}	0.901 (14)	2.252 (13)	3.0614 (11)	149.4 (13)
N3—H2N3···O1^{iv}	0.914 (14)	2.281 (15)	3.0889 (12)	147.1 (12)
C7—H7B···N3^v	0.991 (15)	2.545 (14)	3.4341 (14)	149.2 (11)
C9—H9B···Cg^{iv}	0.96 (2)	2.94 (2)	3.7469 (16)	142.3 (14)

Symmetry codes: (i) $x, y + 1, z$; (ii) $-x, -y + 1, -z + 1$; (iii) $-x + 1, -y, -z + 1$; (iv) $x - 1, y, z$; (v) $-x + 1, -y + 1, -z + 1$. Cg is the centroid of the C1–C6 benzene ring.

Data collection: *APEX2* (Bruker, 2005); cell refinement: *SAINT* (Bruker, 2005); data reduction: *SAINT*; program(s) used to solve structure: *SHELXTL* (Sheldrick, 2008); program(s) used to refine structure: *SHELXTL*; molecular graphics: *SHELXTL*; software used to prepare material for publication: *SHELXTL* and *PLATON* (Spek, 2009).

Supplementary data and figures for this paper are available from the IUCr electronic archives (Reference: LH2884).

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supporting information

2-(4-Methylanilino)acetohydrazide

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S1. Comment

In organic chemistry, hydrazides are a class of organic compounds sharing a common functional group characterized by a nitrogen to nitrogen covalent bond with 4 substituents with at least one of them being an acyl group. They are also starting materials for many heterocycles including 1,2,4-triazoles (Isloor *et al.*, 2009; Holla & Udupa, 1992). Many hydrazide derivatives have showed significant biological activities (Ozdemir *et al.*, 2009; Khattab, 2005). In view of the biological and synthetic importance of hydrazides, we hereby report the crystal structure of the title compound (I).

The bond lengths and angles of the title compound (I), (Fig. 1) are comparable to its related structure (Zhang & Shi, 2009). A maximum deviation of 0.028 (1) Å for atom N2 from the mean plane of the hydrazide group form by atoms O1, N2, N3, C7 and C8 indicates that it is essentially coplanar. The mean plane of the hydrazide makes dihedral angle of 83.34 (5)° with C1–C6 benzene ring. In the crystal structure, the molecules are linked by intermolecular N1—H1N1···O1[i], N2—H1N2···N3[ii], N3—H1N3···O1[iii], N3—H2N3···O1[iv] and C7—H7B···N3[v] (see Table 1 for symmetry codes) hydrogen bonds into two-dimensional network parallel to *ab* plane (Fig. 2, Table 1). The crystal structure is also stabilized by a C—H···π interaction (Table 1).

S2. Experimental

Ethyl [(4-methylphenyl)amino]acetate (19.3 g, 0.1 mol) and hydrazine hydrate (99%, 0.2 mol) in ethanol (200 ml) was heated on a water-bath for 6 h. Excess of ethanol was removed by distillation. On cooling, colourless block-shaped single crystals of 2-[(4-methylphenyl)amino]acetohydrazide begin to separate (Holla & Udupa, 1992). It was collected by filtration and recrystallized from ethanol. Yield: 13.2 g, 73.7%, *M.p.* 423–426 K.

S3. Refinement

All hydrogen atoms were located from the difference Fourier map and refined freely, with N—H = 0.876 (15)–0.912 (15) Å; C–H = 0.96 (2)–1.04 (2) Å.
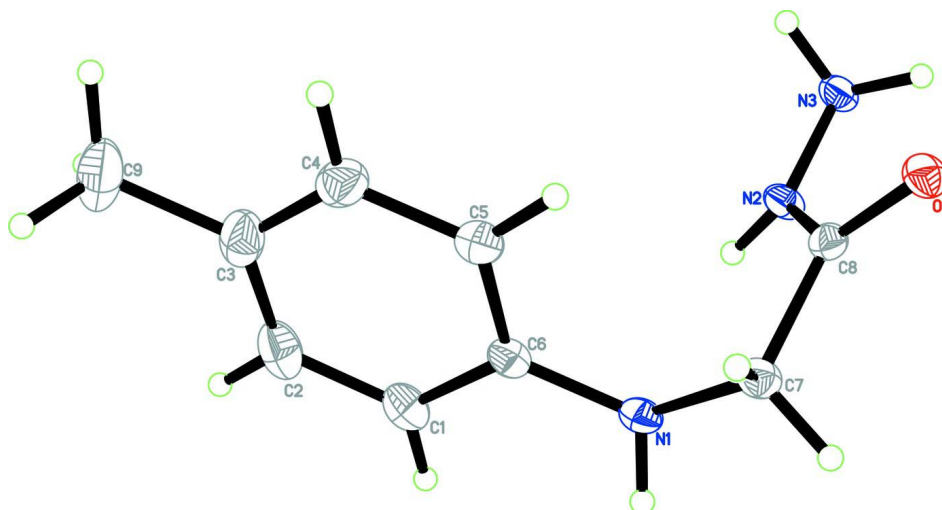
Figure 1

The molecular structure of the title compound with atom labels and 50% probability ellipsoids for non-H atoms.
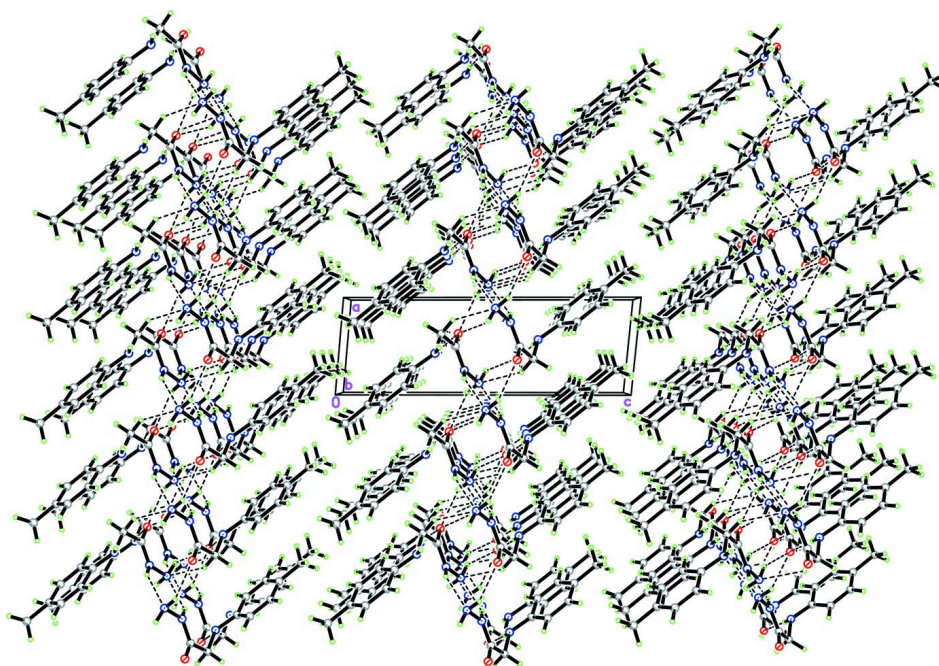
Figure 2

Part of the crystal structure of (I), viewed along the *b* axis, showing the two-dimensional network parallel to *ab* plane. Intermolecular hydrogen bonds are shown in as dashed lines.

2-(4-Methylanilino)acetohydrazide

Crystal data

$C_9H_{13}N_3O$	$b = 5.9262 (2)$ Å
$M_r = 179.22$	$c = 15.4756 (4)$ Å
Triclinic, $P\overline{1}$	$\alpha = 87.002 (2)°$
Hall symbol: -P 1	$\beta = 84.282 (2)°$
$a = 5.1481 (1)$ Å	$\gamma = 82.849 (2)°$

$V = 465.76$ (2) Å^3

$Z = 2$

$F(000) = 192$

$D_\text{x} = 1.278$ Mg m^{-3}

Mo $K\alpha$ radiation, $\lambda = 0.71073$ Å

Cell parameters from 6230 reflections

$\theta = 2.7–31.2°$

$\mu = 0.09$ mm^{-1}

$T = 100$ K

Block, colourless

$0.52 \times 0.15 \times 0.07$ mm

Data collection

Bruker SMART APEXII CCD area-detector diffractometer

Radiation source: fine-focus sealed tube

Graphite monochromator

φ and ω scans

Absorption correction: multi-scan (*SADABS*; Bruker, 2005)

$T_\text{min} = 0.956$, $T_\text{max} = 0.994$

11697 measured reflections

2703 independent reflections

2301 reflections with $I > 2\sigma(I)$

$R_\text{int} = 0.026$

$\theta_\text{max} = 30.0°$, $\theta_\text{min} = 2.7°$

$h = -7 \rightarrow 7$

$k = -8 \rightarrow 8$

$l = -21 \rightarrow 21$

Refinement

Refinement on F^2

Least-squares matrix: full

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

$wR(F^2) = 0.110$

$S = 1.03$

2703 reflections

170 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 atoms treated by a mixture of independent and constrained refinement

$w = 1/[\sigma^2(F_\text{o}^2) + (0.0559P)^2 + 0.1344P]$ where $P = (F_\text{o}^2 + 2F_\text{c}^2)/3$

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

$\Delta\rho_\text{max} = 0.34$ e Å^{-3}

$\Delta\rho_\text{min} = -0.20$ e Å^{-3}

Special details

Experimental. The crystal was placed in the cold stream of an Oxford Cyrosystems Cobra open-flow nitrogen cryostat (Cosier & Glazer, 1986) operating at 100.0 (1) K.

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 (Å^2)

	x	y	z	U_iso*/U_eq
O1	0.63005 (14)	0.15409 (11)	0.39934 (5)	0.01915 (17)
N1	0.43188 (17)	0.74133 (14)	0.33320 (6)	0.01783 (18)
N2	0.26039 (16)	0.39523 (13)	0.43209 (5)	0.01650 (18)
N3	0.12716 (17)	0.23345 (14)	0.48445 (6)	0.01765 (18)
C1	0.1124 (2)	0.95874 (17)	0.24809 (7)	0.0231 (2)
C2	−0.0573 (2)	0.9751 (2)	0.18340 (8)	0.0282 (2)
C3	−0.0748 (2)	0.7920 (2)	0.13190 (7)	0.0280 (2)
C4	0.0857 (2)	0.5915 (2)	0.14806 (7)	0.0257 (2)

C5	0.2577 (2)	0.57064 (17)	0.21309 (7)	0.0208 (2)
C6	0.27314 (19)	0.75502 (16)	0.26432 (6)	0.01770 (19)
C7	0.62171 (19)	0.54434 (16)	0.34553 (7)	0.01795 (19)
C8	0.50373 (18)	0.34635 (15)	0.39460 (6)	0.01517 (18)
C9	−0.2624 (3)	0.8132 (3)	0.06212 (9)	0.0405 (3)
H1A	0.116 (3)	1.091 (3)	0.2848 (10)	0.031 (4)*
H2A	−0.167 (3)	1.117 (3)	0.1751 (11)	0.042 (4)*
H4A	0.081 (3)	0.457 (3)	0.1131 (10)	0.032 (4)*
H5A	0.363 (3)	0.424 (3)	0.2235 (10)	0.031 (4)*
H7A	0.713 (3)	0.484 (2)	0.2916 (9)	0.024 (3)*
H7B	0.757 (3)	0.592 (2)	0.3797 (9)	0.026 (3)*
H9A	−0.199 (4)	0.895 (4)	0.0098 (14)	0.070 (6)*
H9B	−0.435 (4)	0.878 (3)	0.0840 (13)	0.064 (6)*
H9C	−0.290 (4)	0.655 (4)	0.0400 (14)	0.072 (6)*
H1N1	0.485 (3)	0.869 (3)	0.3461 (9)	0.029 (4)*
H1N2	0.185 (3)	0.538 (3)	0.4343 (9)	0.028 (3)*
H1N3	0.250 (3)	0.125 (2)	0.5030 (9)	0.023 (3)*
H2N3	0.031 (3)	0.167 (2)	0.4485 (9)	0.028 (4)*

Atomic displacement parameters (Å^2)

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
O1	0.0173 (3)	0.0139 (3)	0.0257 (4)	0.0010 (2)	−0.0024 (3)	−0.0012 (3)
N1	0.0208 (4)	0.0112 (3)	0.0219 (4)	−0.0032 (3)	−0.0028 (3)	−0.0005 (3)
N2	0.0155 (4)	0.0110 (3)	0.0223 (4)	−0.0013 (3)	0.0001 (3)	0.0014 (3)
N3	0.0159 (4)	0.0135 (4)	0.0233 (4)	−0.0029 (3)	−0.0015 (3)	0.0035 (3)
C1	0.0247 (5)	0.0183 (5)	0.0246 (5)	0.0002 (4)	0.0004 (4)	0.0021 (4)
C2	0.0246 (5)	0.0294 (6)	0.0277 (6)	0.0031 (4)	−0.0008 (4)	0.0081 (4)
C3	0.0224 (5)	0.0410 (6)	0.0209 (5)	−0.0083 (4)	−0.0023 (4)	0.0072 (4)
C4	0.0274 (5)	0.0303 (5)	0.0210 (5)	−0.0109 (4)	−0.0013 (4)	−0.0011 (4)
C5	0.0225 (5)	0.0183 (4)	0.0216 (5)	−0.0041 (3)	−0.0006 (4)	−0.0007 (3)
C6	0.0178 (4)	0.0160 (4)	0.0189 (4)	−0.0033 (3)	0.0006 (3)	0.0015 (3)
C7	0.0156 (4)	0.0158 (4)	0.0222 (5)	−0.0027 (3)	−0.0004 (3)	0.0010 (3)
C8	0.0156 (4)	0.0140 (4)	0.0165 (4)	−0.0020 (3)	−0.0032 (3)	−0.0019 (3)
C9	0.0291 (7)	0.0679 (10)	0.0258 (6)	−0.0130 (6)	−0.0079 (5)	0.0127 (6)

Geometric parameters (Å, º)

O1—C8	1.2421 (11)	C2—H2A	0.962 (17)
N1—C6	1.3995 (13)	C3—C4	1.3867 (17)
N1—C7	1.4437 (12)	C3—C9	1.5094 (17)
N1—H1N1	0.876 (15)	C4—C5	1.3961 (15)
N2—C8	1.3313 (12)	C4—H4A	0.988 (15)
N2—N3	1.4205 (11)	C5—C6	1.3978 (14)
N2—H1N2	0.886 (15)	C5—H5A	0.979 (15)
N3—H1N3	0.902 (14)	C7—C8	1.5213 (13)
N3—H2N3	0.912 (15)	C7—H7A	0.978 (14)
C1—C2	1.3850 (16)	C7—H7B	0.989 (14)

C1—C6	1.4022 (13)	C9—H9A	0.97 (2)
C1—H1A	0.993 (15)	C9—H9B	0.96 (2)
C2—C3	1.3963 (18)	C9—H9C	1.04 (2)
C6—N1—C7	120.37 (8)	C4—C5—C6	120.25 (10)
C6—N1—H1N1	115.9 (10)	C4—C5—H5A	119.3 (9)
C7—N1—H1N1	113.7 (10)	C6—C5—H5A	120.4 (9)
C8—N2—N3	122.60 (8)	C5—C6—N1	122.90 (9)
C8—N2—H1N2	120.9 (10)	C5—C6—C1	118.06 (10)
N3—N2—H1N2	115.5 (10)	N1—C6—C1	118.97 (9)
N2—N3—H1N3	107.5 (9)	N1—C7—C8	113.36 (8)
N2—N3—H2N3	106.4 (9)	N1—C7—H7A	114.3 (8)
H1N3—N3—H2N3	107.4 (12)	C8—C7—H7A	106.5 (8)
C2—C1—C6	120.65 (10)	N1—C7—H7B	107.1 (8)
C2—C1—H1A	119.7 (9)	C8—C7—H7B	108.4 (8)
C6—C1—H1A	119.6 (9)	H7A—C7—H7B	107.0 (12)
C1—C2—C3	121.82 (10)	O1—C8—N2	123.22 (9)
C1—C2—H2A	118.2 (10)	O1—C8—C7	121.37 (9)
C3—C2—H2A	120.0 (10)	N2—C8—C7	115.40 (8)
C4—C3—C2	117.19 (10)	C3—C9—H9A	113.1 (13)
C4—C3—C9	121.88 (12)	C3—C9—H9B	111.3 (12)
C2—C3—C9	120.92 (12)	H9A—C9—H9B	111.5 (17)
C3—C4—C5	122.03 (10)	C3—C9—H9C	112.3 (12)
C3—C4—H4A	120.2 (9)	H9A—C9—H9C	103.7 (17)
C5—C4—H4A	117.8 (9)	H9B—C9—H9C	104.4 (17)
C6—C1—C2—C3	−0.26 (17)	C7—N1—C6—C1	−172.79 (9)
C1—C2—C3—C4	−0.06 (17)	C2—C1—C6—C5	0.35 (15)
C1—C2—C3—C9	179.61 (11)	C2—C1—C6—N1	−176.67 (9)
C2—C3—C4—C5	0.28 (16)	C6—N1—C7—C8	−83.08 (11)
C9—C3—C4—C5	−179.38 (11)	N3—N2—C8—O1	2.92 (14)
C3—C4—C5—C6	−0.19 (16)	N3—N2—C8—C7	−176.34 (8)
C4—C5—C6—N1	176.77 (9)	N1—C7—C8—O1	169.09 (8)
C4—C5—C6—C1	−0.13 (15)	N1—C7—C8—N2	−11.63 (12)
C7—N1—C6—C5	10.34 (14)		

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Symmetry codes: (i) x, $y+1$, z; (ii) $−x$, $−y+1$, $−z+1$; (iii) $−x+1$, $−y$, $−z+1$; (iv) $x−1$, y, z; (v) $−x+1$, $−y+1$, $−z+1$.