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Key indicators

Single-crystal X-ray study
 $T = 294$ K
Mean $\sigma(\text{C}-\text{C}) = 0.004$ Å
Disorder in main residue
 R factor = 0.049
 wR factor = 0.157
Data-to-parameter ratio = 12.1For details of how these key indicators were
automatically derived from the article, see
<http://journals.iucr.org/e>.2-Amino-5,6,7,8-tetrahydro-4-*p*-tolyl-naphthalene-1,3-dicarbonitrile

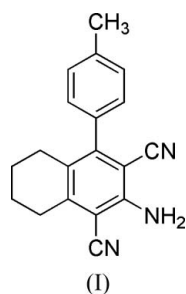
The title compound, $\text{C}_{19}\text{H}_{17}\text{N}_3$, was synthesized by the reaction of 4-methylbenzaldehyde and cyclohexane with malononitrile under solvent-free conditions at 293 K. Conformational disorder is observed in the cyclohexene ring and both the major and minor components adopt half-chair conformations. N—H...N hydrogen bonds generate a centrosymmetric $R_2^2(12)$ dimer.

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Comment

1,3-Dicyanoanilines are typical acceptor–donor–acceptor (A–D–A) systems (Dumur *et al.*, 2004). However, the investigation of the optical properties of this system has rarely been documented (Sepiol & Milart, 1985; Griffiths *et al.*, 1977). 1,3-Dicyanoanilines are prepared from arylidenemalonodinitriles and 1-arylethylidenemalonodinitriles in the presence of piperidine (Sepiol & Milart, 1985); and other methods have also been reported (Victory *et al.*, 1991, 1993; Cui *et al.*, 2005).



Solvent-free reactions have attracted great attention in recent years (Tanaka & Toda, 2000) and have proved to have

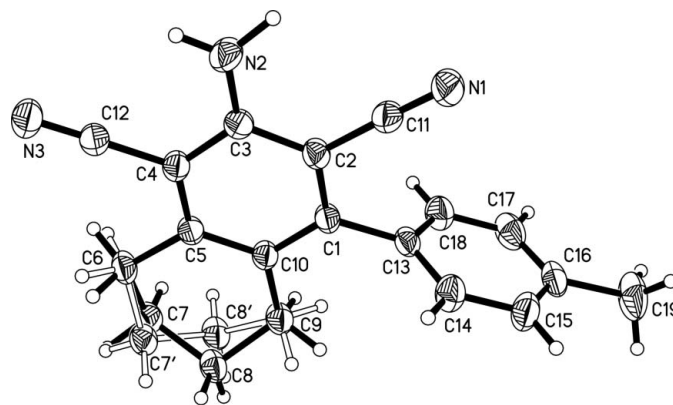


Figure 1
The molecular structure of (I), showing 40% probability displacement ellipsoids and the atom-numbering scheme. Both disorder components are shown.

many advantages: reduced pollution, low costs, and simplicity in processing and handling. Solvent-free reactions have performed well recently (Liu *et al.*, 2001; Kaupp *et al.*, 2003; Goud *et al.*, 1995; Annunziata *et al.*, 1997).

We report here the crystal structure of the title compound, (I), which has been synthesized by the solvent-free reaction of 4-methylbenzaldehyde and cyclohexane with malononitrile at 293 K.

In the title molecule, (I), conformational disorder is observed in the cyclohexene ring and both the major and minor components adopt half-chair conformations (Fig. 1). The dihedral angle between the two benzene rings is 85.74 (9)°. In the crystal structure, N—H···N hydrogen bonds (Table 1) generate a centrosymmetric $R_2^2(12)$ dimer (Fig. 2). A short N1···N1(1/2-x, 1/2-y, -z) contact of 2.944 (4) Å is also observed.

Experimental

Compound (I) was prepared by the reaction of 4-methylbenzaldehyde (2 mmol) and cyclohexane (2 mmol) with malononitrile (4 mmol) under solvent-free conditions using sodium hydroxide (0.20 g) as catalyst at 293 K. Single crystals of (I) suitable for X-ray diffraction were obtained by slow evaporation of an ethanol solution.

Crystal data

$C_{19}H_{17}N_3$	$Z = 8$
$M_r = 287.36$	$D_x = 1.227 \text{ Mg m}^{-3}$
Monoclinic, $C2/c$	Mo $K\alpha$ radiation
$a = 22.837 (6) \text{ \AA}$	$\mu = 0.07 \text{ mm}^{-1}$
$b = 7.906 (2) \text{ \AA}$	$T = 294 (2) \text{ K}$
$c = 17.920 (4) \text{ \AA}$	Plate, colourless
$\beta = 105.955 (5)^\circ$	$0.24 \times 0.20 \times 0.08 \text{ mm}$
$V = 3110.7 (14) \text{ \AA}^3$	

Data collection

Bruker SMART CCD area-detector diffractometer	7651 measured reflections
φ and ω scans	2752 independent reflections
Absorption correction: multi-scan (SADABS; Sheldrick, 1996)	1327 reflections with $I > 2\sigma(I)$
$T_{\min} = 0.972$, $T_{\max} = 0.994$	$R_{\text{int}} = 0.047$
	$\theta_{\text{max}} = 25.0^\circ$

Refinement

Refinement on F^2	$w = 1/[\sigma^2(F_o^2) + (0.0708P)^2 + 0.4798P]$
$R[F^2 > 2\sigma(F^2)] = 0.049$	where $P = (F_o^2 + 2F_c^2)/3$
$wR(F^2) = 0.157$	$(\Delta/\sigma)_{\text{max}} = 0.001$
$S = 1.00$	$\Delta\rho_{\text{max}} = 0.19 \text{ e \AA}^{-3}$
2752 reflections	$\Delta\rho_{\text{min}} = -0.16 \text{ e \AA}^{-3}$
227 parameters	
H atoms treated by a mixture of independent and constrained refinement	

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

D—H···A	D—H	H···A	D···A	D—H···A
N2—H2B···N1 ⁱ	0.98 (4)	2.32 (4)	3.288 (4)	171 (3)

Symmetry code: (i) $-x + \frac{1}{2}, -y + \frac{1}{2}, -z$.

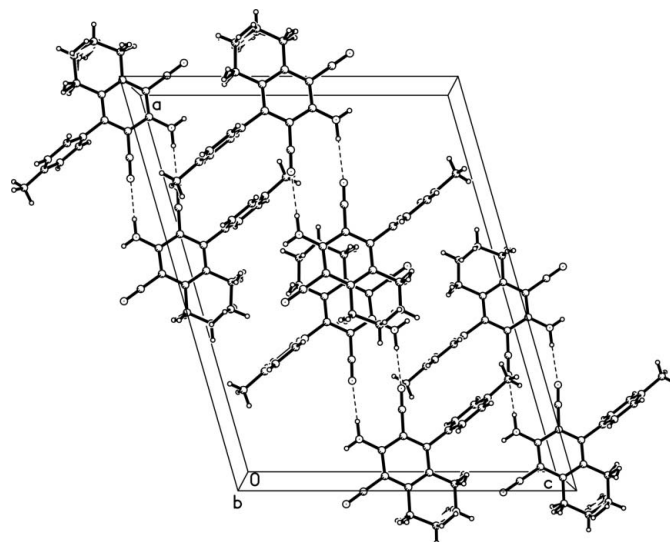


Figure 2

A projection of the crystal packing of (I) along the b axis, showing N—H···N hydrogen-bonded (dashed lines) dimers. Both disorder components are shown.

Atoms C7 and C8 are disordered over two sites, with occupation factors of 0.487 (10) and 0.513 (10). The C—C bond lengths involving the disordered atoms were restrained to 1.54 (1) Å. In addition, the C6—C8, C6—C8', C7—C9 and C7'—C9 distances were restrained to 2.50 (1) Å. The amino H atoms were located in a difference map and refined freely. C-bound H atoms were placed in calculated positions, with C—H = 0.93–0.97 Å, and refined using a riding model, with $U_{\text{iso}}(\text{H}) = 1.2\text{--}1.5U_{\text{eq}}(\text{C})$.

Data collection: SMART (Bruker, 1998); cell refinement: SAINT (Bruker, 1999); data reduction: SAINT; program(s) used to solve structure: SHELXS97 (Sheldrick, 1997); program(s) used to refine structure: SHELXL97 (Sheldrick, 1997); molecular graphics: SHELXTL (Bruker, 1999); software used to prepare material for publication: SHELXTL.

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