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2-Amino-4-(2-chlorophenyl)-7,7-dimethyl-1-(4-methylphenyl)-5-oxo-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

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Key indicators

Single-crystal X-ray study
T = 173 K
Mean σ(C–C) = 0.003 Å
R factor = 0.029
wR factor = 0.076
Data-to-parameter ratio = 12.5

For details of how these key indicators were automatically derived from the article, see http://journals.iucr.org/e.

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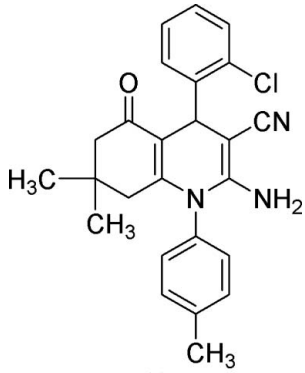
The title compound, $C_{25}H_{24}ClN_3O$, was synthesized by the reaction of 2-chlorophenylmethylidenemalononitrile and 3-(4-tolylamino)-5,5-dimethylcyclohex-2-enone in the presence of triethylbenzylammonium chloride in an aqueous medium. All bond lengths and angles are normal. In the crystal structure, intermolecular N—H···O hydrogen bonds link the molecules into chains extending in the [101] direction.

Comment

Quinolines and their derivatives exhibit diverse pharmacological activities, such as cholinesterase (Isomae *et al.*, 2002), antiplasmoidal (Beagley *et al.*, 2003), cytotoxic (Ma *et al.*, 2004), functional (Denton *et al.*, 2005) and antibacterial (Fokialakis *et al.*, 2002) activities. To avoid the inherent disadvantages of many organic solvents, such as toxicity and non-stability, we are working under a new environmentally friendly procedure for the synthesis of the above-mentioned compounds. Specifically, we have focused our attention on the use of water as a reaction medium following Breslow & Rideout (1980), who rediscovered the use of water as a solvent in organic chemistry. We report here the crystal structure of the title compound, (I).



(I)

In (I), all bond lengths and angles (Table 1) are normal. The N1/C1–C5 ring is not planar, adopting a boat conformation (Fig. 1). Atoms C3 and N1 deviate from the basal plane defined by atoms C1/C2/C4/C5 by 0.200 (2) and 0.073 (1) Å, respectively. Similar distortions were observed in 7,7-dimethyl-2-(4-bromophenyl)-4-phenyl-5-oxo-1,4,5,6,7,8-hexahydroquinoline (Shi *et al.*, 2002) and 3,3,6,6-tetramethyl-9-(4-chlorophenyl)-10-(4-methylphenyl)-1,2,3,4,5,6,7,8,9,10-decahydroacridine-1,8-dione (Wang *et al.*, 2003). The C4/C5/C7–C10 ring adopts a half-chair conformation; atom C9 deviates from the mean plane of the remaining atoms by 0.630 (2) Å. A

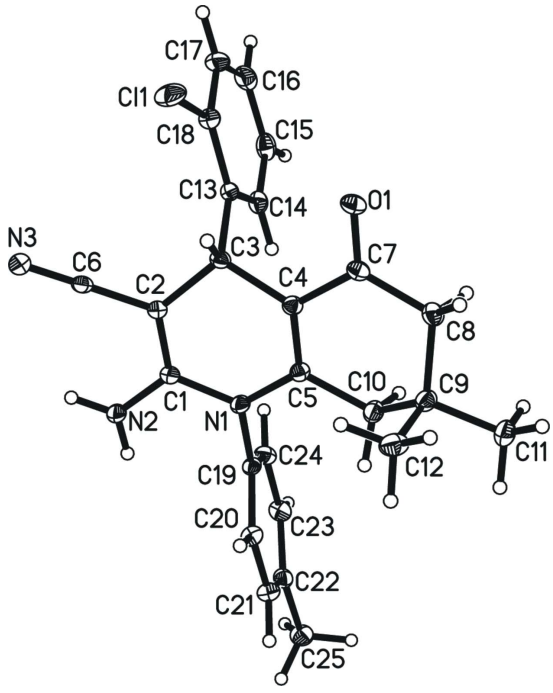


Figure 1
A view of (I), showing the atom-numbering scheme and with displacement ellipsoids drawn at the 50% probability level.

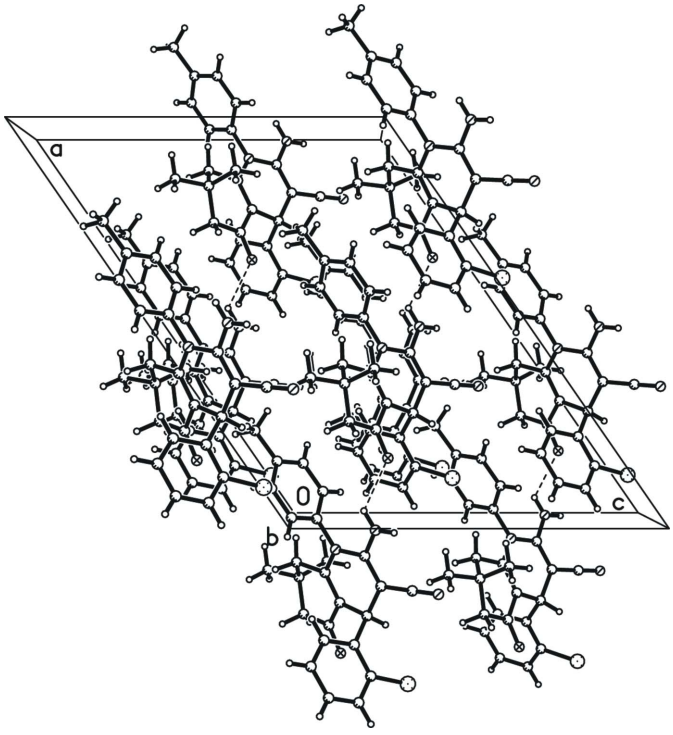


Figure 2
The molecular packing, viewed down the *b* axis, showing the intermolecular hydrogen bonds as dashed lines.

similar conformation has been found in the structure of 7,7-dimethyl-2-amino-3-cyano-4-(3,4-methylenedioxylphenyl)-5-oxo- 5,6,7,8-tetrahydro-4*H*-benzo[*b*]pyran (Wang *et al.*, 2002). The benzene rings make a dihedral angle of 9.0 (1)°.

In the crystal structure, intermolecular N—H···O hydrogen bonds (Table 2) link the molecules into chains extending in the [101] direction (Fig. 2).

Experimental

The title compound, (I), was prepared by the reaction of 2-chlorophenylmethylidenemalononitrile (2 mmol) and 3-(4-tolylamino)-5,5-dimethylcyclohex-2-enone (2 mmol) in the presence of triethylbenzylammonium chloride (0.1 g) in water at 363 K for 8 h (yield 98%; m.p. 525–527 K). Crystals of (I) suitable for X-ray diffraction were obtained by slow evaporation of a dimethylformamide solution. Elemental analysis calculated: C 71.85, H 5.79, N 10.05%; found: C 71.73, H 6.01, N 9.98%. ^{1}H NMR (DMSO-d_6, δ, p.p.m.): 0.73 (*s*, 3H, CH_3), 0.89 (*s*, 3H, CH_3), 1.73 (*d*, J = 16.0 Hz, 1H, CH), 1.90 (*d*, J = 16.0 Hz, 1H, CH), 2.19 (*d*, J = 16.8 Hz, 2H, CH_2), 4.47 (*s*, 1H, CH), 5.33 (*s*, 2H, NH_2), 7.04–7.48 (*m*, 8H, ArH).

Crystal data

$C_{25}H_{24}ClN_3O$
M_r = 417.92
Monoclinic, *Cc*
a = 19.541 (2) Å
b = 9.0199 (8) Å
c = 14.8424 (16) Å
β = 124.628 (2)°
V = 2152.7 (4) Å^3
Z = 4
D_x = 1.290 Mg m^{-3}
Mo $K\alpha$ radiation
Cell parameters from 4284 reflections
θ = 3.3–25.3°
μ = 0.20 mm^{-1}
T = 173 (2) K
Block, yellow
0.59 × 0.50 × 0.30 mm

Data collection

Rigaku Mercury diffractometer
ω scans
Absorption correction: multi-scan (Jacobson, 1998)
T_{min} = 0.892, T_{max} = 0.943
10014 measured reflections
3434 independent reflections
3389 reflections with $I > 2\sigma(I)$
R_{int} = 0.016
θ_{max} = 25.3°
$h = -23 \rightarrow 21$
$k = -10 \rightarrow 10$
$l = -15 \rightarrow 17$

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)]$ = 0.029
$wR(F^2)$ = 0.076
S = 1.05
3434 reflections
275 parameters
H-atom parameters constrained
$w = 1/[\sigma^2(F_o^2) + (0.0474P)^2 + 0.7163P]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{max} < 0.001$
$\Delta\rho_{max}$ = 0.20 e Å^{-3}
$\Delta\rho_{min}$ = −0.22 e Å^{-3}
Absolute structure: Flack (1983), with 1478 Friedel pairs
Flack parameter: −0.04 (5)

Table 1
Selected geometric parameters (Å, °).

N1—C1	1.392 (2)	C2—C3	1.524 (2)
N1—C5	1.396 (2)	C3—C4	1.514 (2)
C1—C2	1.360 (2)	C4—C5	1.355 (2)
C1—N1—C5	119.95 (14)	C4—C3—C2	108.62 (14)
C2—C1—N1	120.82 (15)	C5—C4—C3	123.57 (15)
C1—C2—C3	123.42 (15)	C4—C5—N1	121.15 (15)
C5—N1—C1—C2	6.5 (2)	C2—C3—C4—C5	15.2 (2)
N1—C1—C2—C3	6.6 (3)	C3—C4—C5—N1	−4.5 (3)
C1—C2—C3—C4	−16.2 (2)		

Table 2
Hydrogen-bond geometry (Å, °).

D—H···A	D—H	H···A	D···A	D—H···A
N2—H2B···O1^{i}	0.88	2.36	2.937 (2)	123

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

The H atoms were positioned geometrically and refined as riding, with C—H = 0.95–1.00 Å and N—H = 0.88 Å, and with U_{iso}(H) = 1.2–1.5U_{eq} of the parent atom.

Data collection: *CrystalClear* (Rigaku, 1999); cell refinement: *CrystalClear*; data reduction: *CrystalStructure* (Rigaku/MSC, 2003); program(s) used to solve structure: *SHELXS97* (Sheldrick, 1997*a*); program(s) used to refine structure: *SHELXL97* (Sheldrick, 1997*a*); molecular graphics: *SHELXTL* (Sheldrick, 1997*b*); software used to prepare material for publication: *SHELXL97*.

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