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2-Amino-3-cyano-4-(4-methoxyphenyl)-1,4,5,6-tetrahydrobenzo[*h*]chromene

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Key indicators

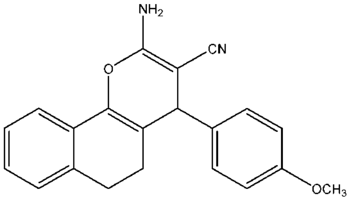
Single-crystal X-ray study
T = 296 K
Mean σ(C–C) = 0.002 Å
R factor = 0.037
wR factor = 0.093
Data-to-parameter ratio = 13.3

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

The title compound, $C_{21}H_{18}N_2O_2$, has been synthesized by the reaction of 2-(4-methoxyphenylmethylidene)-3,4-dihydronaphthalen-1(2*H*)-one and malononitrile in refluxing ethyl alcohol catalyzed by KF–Al_2O_3. The pyran ring adopts a half-chair conformation, while the fused, partially saturated six-membered ring is in a distorted boat form.

Comment

2-Aminochromenes are an important class of compounds, found in many naturally occurring products, and employed as cosmetics and pigments and utilized as potential biodegradable agrochemicals (Morinaka & Takahashi, 1977; Witte *et al.*, 1986; Hafez *et al.*, 1987). We report here the X-ray crystal structure of the title compound, (I).



(I)

The pyran ring adopts a half-chair conformation. Atoms C1, C2, C4, C5 and O1 are coplanar, while atom C3 deviates from this plane by 0.149 (2) Å. The bond distance C1═C2 [1.350 (2) Å] is significantly longer than C4═C5 [1.324 (2) Å], and there is a corresponding shortening of C1—N1 [1.342 (2) Å] relative to the normal Csp^2—N bond (1.426 Å; Lorente *et al.*, 1995). The six-membered ring C4–C6/C11–C13 adopts a distorted boat conformation. Atoms C4, C5, C6 and C13 are coplanar, while atoms C11 and C12 deviate from the plane by 0.260 (2) and 0.711 (2) Å, respectively. A similar conformation was observed for the partially saturated six-membered ring in 2-amino-4-phenyl-5,6-dihydrobenzo[*h*]-quinazoline (Wang *et al.*, 2003). The molecules are linked by N—H···O and N—H···N hydrogen bonds (Table 2), forming a polymer in the crystal structure (Figs. 2 and 3).

Experimental

The utility of fluoride salts as potential bases in variety of synthetic reactions has been recognized in recent years. In particular, alumina coated with potassium fluoride (KF–alumina), resulting in higher selectivity, milder reaction conditions and easier work-up, has been reported as a useful catalyst for many reactions (Clark, 1980). The title compound, (I), was prepared by the reaction of 2-(4-methoxyphenylmethylidene)-3,4-dihydronaphthalen-1(2*H*)-one and malononitrile in refluxing ethyl alcohol catalyzed by KF–Al_2O_3. Single

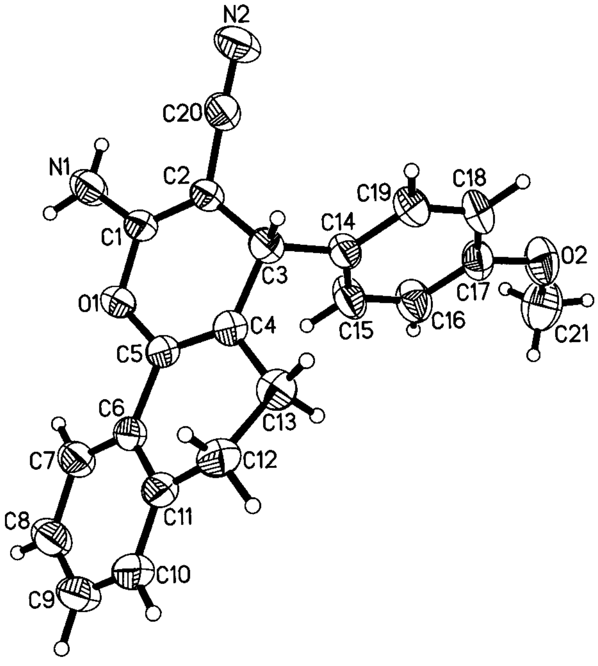


Figure 1
The molecular structure of (I), showing 50% probability displacement ellipsoids and the atom-numbering scheme.

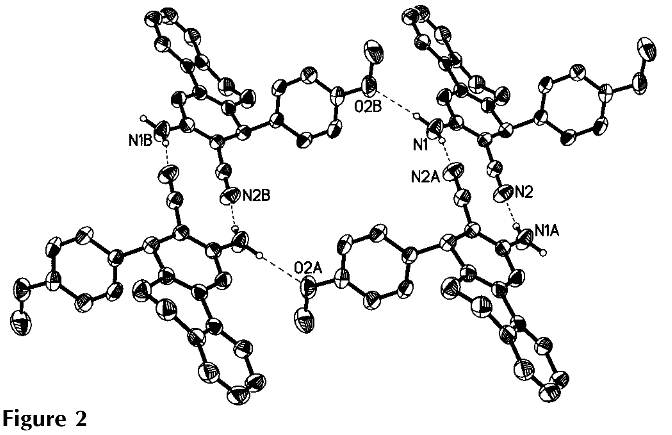


Figure 2
The intermolecular hydrogen bonds for (I). [Symmetry codes: *A* $-x$, $1-y$, $1-z$; *B* x, y, $z-1$, corresponding to (ii) and (i), respectively, in Table 2.]

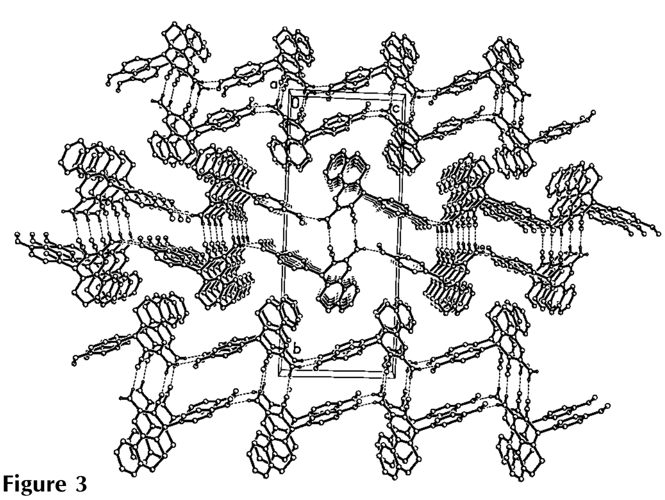


Figure 3
The molecular packing diagram of (I), viewed along **a**.

crystals of (I) suitable for X-ray diffraction were obtained from an ethanol solution by slow evaporation (m.p. 456–457 K).

Crystal data

$C_{21}H_{18}N_2O_2$
$M_r = 330.37$
Monoclinic, $P2_1/c$
$a = 6.478$ (1) Å
$b = 25.557$ (5) Å
$c = 10.549$ (2) Å
$\beta = 96.75$ (2)°
$V = 1734.4$ (5) Å^3
$Z = 4$

$D_x = 1.265$ Mg m^{-3}
Mo $K\alpha$ radiation
Cell parameters from 31 reflections
$\theta = 3.1$–$18.5°$
$\mu = 0.08$ mm^{-1}
$T = 296$ (2) K
Block, yellow
0.58 × 0.44 × 0.40 mm

Data collection

Siemens *P*4 diffractometer
ω scans
Absorption correction: none
3660 measured reflections
3147 independent reflections
1977 reflections with $I > 2\sigma(I)$
$R_{\rm int} = 0.010$

$\theta_{\max} = 25.3°$
$h = 0 \rightarrow 7$
$k = 0 \rightarrow 30$
$l = -12 \rightarrow 12$
3 standard reflections every 97 reflections intensity decay: 3.0%

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)] = 0.037$
$wR(F^2) = 0.093$
$S = 0.89$
3147 reflections
236 parameters
H atoms treated by a mixture of independent and constrained refinement

$w = 1/[\sigma^2(F_o^2) + (0.052P)^2]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{\max} < 0.001$
$\Delta\rho_{\max} = 0.15$ e Å^{-3}
$\Delta\rho_{\min} = -0.11$ e Å^{-3}
Extinction correction: *SHELXTL*
Extinction coefficient: 0.0165 (14)

Table 1
Selected geometric parameters (Å, °).

O1—C1	1.3560 (17)	C2—C3	1.514 (2)
O1—C5	1.3985 (17)	C3—C4	1.501 (2)
O2—C17	1.3691 (19)	C4—C5	1.324 (2)
O2—C21	1.413 (2)	C4—C13	1.510 (2)
N1—C1	1.342 (2)	C11—C12	1.501 (2)
N2—C20	1.1467 (19)	C12—C13	1.517 (2)
C1—C2	1.350 (2)		
C1—O1—C5	118.04 (12)	C5—C4—C3	122.54 (14)
N2—C20—C2	176.91 (17)	C4—C5—O1	123.56 (14)
C2—C1—O1	122.40 (14)	C11—C12—C13	112.26 (16)
C1—C2—C3	123.15 (13)	C4—C13—C12	112.18 (14)
C4—C3—C2	109.26 (12)		
C5—O1—C1—C2	4.8 (2)	C1—O1—C5—C4	−4.4 (2)
O1—C1—C2—C3	3.1 (2)	C4—C5—C6—C11	15.3 (2)
C1—C2—C3—C4	−10.1 (2)	C6—C11—C12—C13	−37.6 (2)
C2—C3—C4—C5	10.4 (2)	C5—C4—C13—C12	−27.5 (2)
C3—C4—C5—O1	−4.0 (2)	C11—C12—C13—C4	46.7 (2)
C13—C4—C5—C6	−3.9 (2)		

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

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
N1—H1N*A*···O2$^{\rm i}$	0.871 (9)	2.189 (10)	3.046 (2)	168.1 (15)
N1—H1N*B*···N2$^{\rm ii}$	0.870 (9)	2.151 (10)	3.016 (2)	173.0 (16)

Symmetry codes: (i) x, y, $z-1$; (ii) $-x$, $1-y$, $1-z$.

Atoms H1N*A* and H1N*B* were refined isotropically with the N—H bond length restrained to 0.86 Å. Other H atoms were positioned geometrically and refined as riding [C—H = 0.93–0.98 Å and $U_{\rm iso}$(H) = 1.2$U_{\rm eq}$(C)].

Data collection: *XSCANS* (Siemens, 1994); cell refinement: *XSCANS*; data reduction: *SHELXTL* (Sheldrick, 1997); program(s) used to solve structure: *SHELXTL*; program(s) used to refine structure: *SHELXTL*; molecular graphics: *SHELXTL*; software used to prepare material for publication: *SHELXTL*.

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