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Diethyl 2-(acridin-9-ylmethylene)malonate

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Key indicators

Single-crystal X-ray study
T = 173 K
Mean σ(C–C) = 0.002 Å
R factor = 0.036
wR factor = 0.086
Data-to-parameter ratio = 15.1

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

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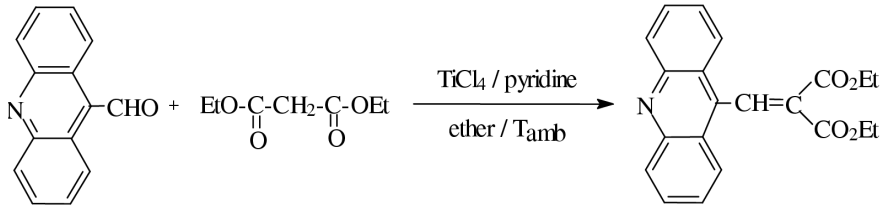
The title compound, $C_{21}H_{19}NO_4$, (II), is an acridine derivative with an angle of 68.30 (2)° between the aromatic ring system and the ethylene moiety. It is remarkable that (II) is isostructural with diethyl 2-(anthracen-9-ylmethylene)-malonate, which bears a C atom at the position where in (II) an N atom is found.

Comment

Derivatives of acridine are very important building blocks in synthetic chemistry. They have interesting biological and photophysical properties (Galy *et al.*, 1980). We have recently described the preparation of diethyl 2-(anthracen-9-yl-methylene)malonate (Elazami *et al.*, 1999). Such compounds are prepared by the condensation reaction of 9-aldehyde acridine (I) and ethyl malonate (see scheme below). We present here the structure of diethyl 2-(acridin-9-ylmethyl-ene)malonate, (II), which was synthesized according to the method of Lehnert (1972). (II) displays a high stability at room temperature and its structure has been determined by IR, mass, UV and NMR (^{1}H, ^{13}C) spectroscopy. Since NMR and mass spectroscopy did not provide sufficient information about the conformation of the reaction product, we have carried out the X-ray structure analysis.



(I) (II)

The title compound contains an acridine ring system to which an ethylene bond is attached at position 9. Due to steric hindrance this ethylene bond cannot be coplanar with the aromatic ring system; the angle between the acridine ring system and the ethylene moiety is 68.30 (2)°. Whereas one of the ester groups (O17, C17, O18, C19, C20) is nearly planar (r.m.s.d. = 0.232 Å) and coplanar with the ethylene bond (Table 1), the other one (C21, O21, O22, C23, C24) is less planar (r.m.s.d. = 0.318 Å) and it is tilted by 77.34 (5)° out of the plane of the ethylene bond. The aromatic ring systems of two adjacent molecules are partly stacked with an interplanar distance of approximately 3.5 Å.

It is remarkable that compound (II) is isostructural with diethyl 2-(anthracen-9-ylmethylene)malonate (Elazami *et al.*, 1999). A least-squares fit of all non-H atoms gives an r.m.s.

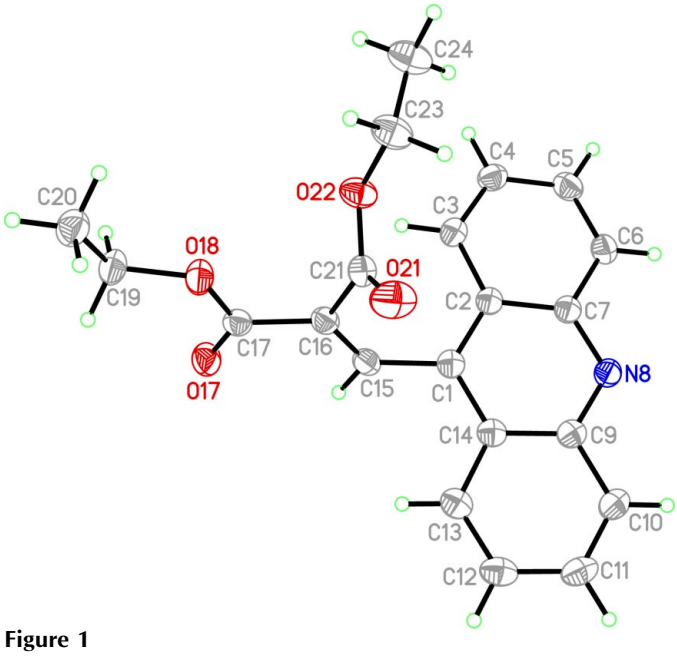


Figure 1
A perspective view of (II) with the atom-numbering scheme. Displacement ellipsoids are at the 50% probability level and H atoms are drawn as small spheres of arbitrary radii.

deviation of 0.082 Å. There are no significant differences between both structures apart from the geometry around the N atom: the C—N bonds (Table 1) are of course shorter than the respective C—C bonds [1.395 (3) and 1.397 (3) Å] and the C—N—C angle is smaller than the corresponding C—C—C angle [122.4 (2)°].

Experimental

Equimolar quantities of ethyl malonate and pyridine, each 0.5 g (6.3 mmol), and 9-aldehyde acridine (1 g, 0.5 mmol) in 30 ml diethyl ether were added to a 100 ml three-necked flask fitted with a reflux condenser. The mixture was stirred at room temperature. Then 3 ml titanium tetrachloride ($TiCl_4$) dissolved in 20 ml diethyl ether was added. At the end of the addition reaction, the mixture was stirred and refluxed for 8 h. The solution was filtered and extracted with diethyl ether (50 ml). The solvent was removed using a rotary evaporator. The residue was recrystallized from ethyl ether and hexane (2:3 ratio) leading to yellow crystals of the title compound.

Crystal data

$C_{21}H_{19}NO_4$
M_r = 349.37
Monoclinic, $P2_1/n$
a = 9.8535 (7) Å
b = 14.888 (1) Å
c = 12.5317 (8) Å
β = 108.805 (5)°
V = 1740.3 (2) Å^3
Z = 4
D_x = 1.333 Mg m^{-3}
Mo $K\alpha$ radiation
Cell parameters from 509 reflections
θ = 1–20°
μ = 0.09 mm^{-1}
T = 173 (2) K
Block, yellow
0.68 × 0.62 × 0.28 mm

Data collection

Siemens CCD three-circle diffractometer
ω scans
Absorption correction: empirical (*SADABS*; Sheldrick, 1996)
T_{min} = 0.940, T_{max} = 0.975
25 802 measured reflections
3553 independent reflections
3240 reflections with $I > 2\sigma(I)$
R_{int} = 0.025
θ_{max} = 27.1°
$h = -12 \rightarrow 12$
$k = -17 \rightarrow 18$
$l = -15 \rightarrow 15$
434 standard reflections
frequency: 1200 min
intensity decay: none

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)]$ = 0.036
$wR(F^2)$ = 0.086
S = 1.05
3553 reflections
236 parameters
H-atom parameters constrained
$w = 1/[\sigma^2(F_o^2) + (0.0336P)^2 + 0.6851P]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{max} < 0.001$
$\Delta\rho_{max}$ = 0.21 e Å^{-3}
$\Delta\rho_{min}$ = −0.17 e Å^{-3}
Extinction correction: *SHELXL*97
Extinction coefficient: 0.0117 (11)

Table 1
Selected geometric parameters (Å, °).

C7—N8	1.3451 (15)	N8—C9	1.3471 (15)
C7—N8—C9	117.74 (10)		
C1—C15—C16—C17	175.44 (10)	C17—O18—C19—C20	166.89 (10)
C1—C15—C16—C21	−6.31 (18)	C15—C16—C21—O21	−72.84 (17)
C15—C16—C17—O17	−0.21 (17)	C15—C16—C21—O22	107.05 (13)
C15—C16—C17—O18	179.12 (10)	C16—C21—O22—C23	178.21 (10)
C16—C17—O18—C19	173.01 (9)	C21—O22—C23—C24	158.67 (11)

All H atoms were located by difference Fourier synthesis and were refined with fixed individual displacement parameters [U_{iso}(H) = 1.2U_{eq}(C) or 1.5U_{eq}(C_{methyl})] using a riding model with aromatic C—H = 0.95 Å, methyl C—H = 0.98 Å or methylene C—H = 0.99 Å.

Data collection: *SMART* (Siemens, 1995); cell refinement: *SMART*; data reduction: *SAINT* (Siemens, 1995); program(s) used to solve structure: *SHELXS*97 (Sheldrick, 1990); program(s) used to refine structure: *SHELXL*97 (Sheldrick, 1997); molecular graphics: XP in *SHELXTL-Plus* (Siemens, 1991).

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