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Diethyl 1,1′-ethane-1,2-diylbis(2-methyl-5-oxo-4,5-dihydro-1*H*-pyrrole-3-carboxylate)

Zhen-Feng Zhang,[a]* Si-Qian Wang,[b] Jian-Ping Li[a] and Gui-Rong Qu[a]

[a]College of Chemistry and Environmental Science, Henan Normal University, Xinxiang 453007, People's Republic of China, and [b]Institute of Chemistry, Chinese Academy of Sciences, Beijing 100080, People's Republic of China
Correspondence e-mail: zzf5188@sohu.com

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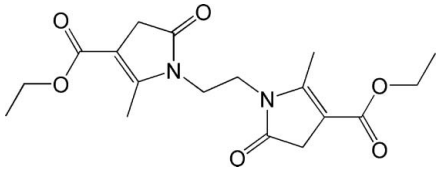
Key indicators: single-crystal X-ray study; T = 291 K; mean σ(C–C) = 0.003 Å; R factor = 0.052; wR factor = 0.139; data-to-parameter ratio = 14.3.

In the title compound, $C_{18}H_{24}N_2O_6$, the two pyrrolinone rings are planar and adopt *anti* conformations. The molecule lies on an inversion centre. The molecules are linked into a three-dimensional framework structure by three independent C—H···O hydrogen bonds.

Related literature

For related literature, see: Bernstein *et al.* (1995); Zhang *et al.* (2004, 2007).



Experimental

Crystal data

$C_{18}H_{24}N_2O_6$
M_r = 364.39
Monoclinic, $P2_1/n$
a = 4.5403 (10) Å
b = 12.418 (3) Å
c = 16.439 (4) Å
β = 91.812 (3)°
V = 926.4 (4) Å^3
Z = 2
Mo $K\alpha$ radiation
μ = 0.10 mm^{-1}
T = 291 (2) K
0.30 × 0.15 × 0.12 mm

Data collection

Bruker SMART CCD area-detector diffractometer
Absorption correction: multi-scan (*SADABS*; Sheldrick, 1997*a*)
T_{min} = 0.971, T_{max} = 0.988
6085 measured reflections
1716 independent reflections
1164 reflections with $I > 2\sigma(I)$
R_{int} = 0.032

Refinement

$R[F^2 > 2\sigma(F^2)]$ = 0.052
$wR(F^2)$ = 0.139
S = 1.04
1716 reflections
120 parameters
78 restraints
H-atom parameters constrained
$\Delta\rho_{max}$ = 0.17 e Å^{-3}
$\Delta\rho_{min}$ = −0.14 e Å^{-3}

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

D—H···A	D—H	H···A	D···A	D—H···A
C7—H7A···O2	0.96	2.31	2.966 (4)	125
C8—H8B···O1^{i}	0.97	2.71	3.429 (3)	131
C7—H7C···O1^{ii}	0.96	2.55	3.417 (3)	150
C7—H7A···O1^{iii}	0.96	2.71	3.337 (3)	123

Symmetry codes: (i) $x-1, y, z$; (ii) $-x+\frac{3}{2}, y-\frac{1}{2}, -z+\frac{3}{2}$; (iii) $-x+\frac{5}{2}, y-\frac{1}{2}, -z+\frac{3}{2}$.

Data collection: *SMART* (Bruker, 1997); cell refinement: *SAINT* (Bruker, 1997); data reduction: *SAINT*; 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: *SHELXTL*.

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Supplementary data and figures for this paper are available from the IUCr electronic archives (Reference: PV2017).

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supplementary materials

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Diethyl 1,1'-ethane-1,2-diylbis(2-methyl-5-oxo-4,5-dihydro-1*H*-pyrrole-3-carboxylate)

Z.-F. Zhang, S.-Q. Wang, J.-P. Li and G.-R. Qu

Comment

The reaction of ethylenediamine with ethyl acetoacetate yields dienamine,(I) (scheme 1), cyclization of which with glyoxal and thereafter rearrangement yields bicyclic pyrrolinone (II). We have reported the structure of the dienamine,(I) (Zhang *et al.*, 2004). We have now prepared several novel ethylenedi(2-pyrrolin-5-one) derivatives (Zhang *et al.*, 2007), but unfortunately, only the title compound, (II) has provided crystals suitable for single-crystal structure determination. We report here the molecular and supramolecular structure of the title compound, (II).

The structural unit in the title compound (Fig. 1) adopts a low-energy anti conformation with torsion angle N1—C4—C4^{i}—N1^{i} [symmetry code: (i) $1-x,1-y,1-z$] 180.0 (2)°. The anti-conformation is thought to result from a van der Waals repulsion effect between the two pyrrolinone rings. The pyrrolinone rings are perfectly planar, with a mean deviation of 0.006 Å. The dihedral angle between the planes of the two rings is 0 °, indicating that the two rings are parallel to one another. Within the pyrrolinone rings, there is a clear distinction between single and double bonds. The lengths for C2═C3 and C1═O1 bonds [1.349 (3) and 1.211 (3) Å, respectively] are shorter than the corresponding bonds found in the precursor (I) [1.370 (4) and 1.222 (4) Å, respectively; Zhang *et al.*, 2004]. Conversely, the N1—C3 and C1—C2 bonds are longer [1.411 (3) and 1.459 (3) Å *versus* 1.342 (4) and 1.439 (4) Å, respectively]. It is also interestingly found that within pyrrolinone rings, the distance of 1.385 Å for N1—C9 is shorter than that of 1.411Å for N1—C3 bond, indicating that as compared with C3═C2, the C9═O3 bond is in π-π conjugation with the N1 atom. The sum of the three angles around each of the N1, C3 and C2 atoms is 359.98 (2)°, implying that the N6, C3 and C2 atoms take on sp^2 hybridization.

In the molecule (II), there are two intramolecular C—H···O hydrogen bonds (Table 1), which, though not strong, contribute to the relatively stable co-planarity for the structural unit O2/C1/C2/C3/C7. This presumably sets the stage in turn for the interactions within the crystal lattice.

The molecules of compound (II) are linked by six independent C—H···O hydrogen bonds (Table 1) into a three-dimensional framework structure, whose formation is rather easily analysed in terms of two simple substructures, one of which is one-dimensional and the other is two-dimensional.

In the one-dimensional substructure, the methylene C8 atom in the pyrrolinone ring acts as a hydrogen-bond donor, *via* H8B, to the acyl atoms, thus forming the centrosymmetric $R^2_2(24)$ dimer (Bernstein *et al.*, 1995) centred at (0,1/2,1/2); details of hydrogen-bonding geometry has been given in Table 1. Propagation by translation of these two hydrogen bonds generates a C(4) C(4) (Bernstein *et al.*, 1995) hydrogen-bonded chains (column) along the *a* axis.

The column centred at (0,1/2,1/2) associates further *via* two independent bifurcated donor hydrogen bonds and two bifurcated acceptor hydrogen bonds with nearby four columns centred at (0,0,0), (0,1,0),(0,0,1) and (0,1,1), respectively. These lateral interactions reinforce structures by adding hydrogen bonds and extending them forming a two-dimensional substructure. The methyl atom C7 acts as a hydrogen-bond donor, *via* atoms H7A and H7C, to O atoms forming a two-dimensional

hydrogen-bonded substructure (details in Table 1). The combination of the one- and two-dimensional substructures suffices to generate the three-dimensional framework structure (Fig. 2).

Experimental

Into a three-necked round-bottomed flask equipped with a mechanical stirrer were introduced dienamine, (I), (28.4 g, 0.1 mol), ethanol (95%, 50 ml) and glyoxal (18 g, 40%). The mixture was then heated at about 333 K with stirring for 30 min under an inert atmosphere. Natural cooling of the reaction mixture overnight gave a crystalline product (yield: 18%). which was recrystallized from ethyl acetate. Single crystals of (II) were obtained by cooling of the hot solution of the above product in ethyl acetate very slowly over one week. ^{1}HNMR($CDCl_3$, 400 MHz): δ 4.17 (q, J = 7.2 Hz, 4H, $2CH_2$), δ 3.66 (s, 4H, $2CH_2$), δ 3.23 (q, J = 2.4 Hz, 4H, $2CH_2$), δ 2.40 (t, J = 2.4 Hz, 6H, $2CH_3$), δ 1.27 (t, J = 7.2 Hz, 6H, $2CH_3$).

Refinement

H atoms were placed in idealized positions and were allowed to ride on the respective parent atoms with C—H = 0.98 Å and N—H = 0.86 Å, and with $U_{iso}(H) = xU_{eq}$(carrier atom), where x = 1.2 for C—H and N—H.

Figures

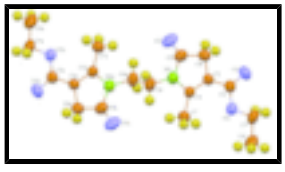


Fig. 1. A view of the molecule of (II); displacement ellipsoids are drawn at the 50% probability level.

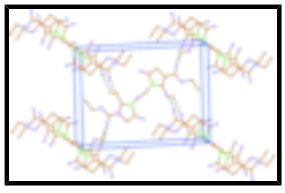


Fig. 2. Part of the crystal structure of (II), showing the formation of two-dimensional substructure; H atoms not involved in the motifs have been omitted for clarity.

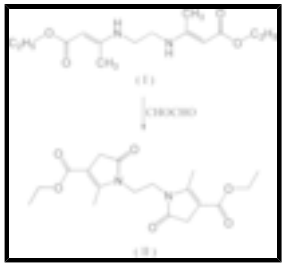


Fig. 3. The formation of the title compound.

Diethyl 1,1'-ethane-1,2-diylbis(2-methyl-5-oxo-4,5-dihydro-1*H*-pyrrole-3-carboxylate)

Crystal data

$C_{18}H_{24}N_2O_6$	F_{000} = 388
M_r = 364.39	D_x = 1.306 Mg m^{-3}
Monoclinic, $P2_1/n$	Mo $K\alpha$ radiation λ = 0.71073 Å

Hall symbol: -P 2yn	Cell parameters from 1143 reflections
a = 4.5403 (10) Å	θ = 2.5–20.9º
b = 12.418 (3) Å	μ = 0.10 mm^{-1}
c = 16.439 (4) Å	T = 291 (2) K
β = 91.812 (3)º	Block, colourless
V = 926.4 (4) $Å^3$	0.30 × 0.15 × 0.12 mm
Z = 2	

Data collection

Bruker SMART CCD area-detector diffractometer	1716 independent reflections
Radiation source: fine-focus sealed tube	1164 reflections with $I > 2\sigma(I)$
Monochromator: graphite	R_{int} = 0.032
T = 291(2) K	θ_{max} = 25.5º
φ and ω scans	θ_{min} = 2.5º
Absorption correction: multi-scan (SADABS; Sheldrick, 1997a)	$h = -5 \rightarrow 5$
T_{min} = 0.971, T_{max} = 0.988	$k = -15 \rightarrow 14$
6085 measured reflections	$l = -19 \rightarrow 19$

Refinement

Refinement on F^2	Secondary atom site location: difference Fourier map
Least-squares matrix: full	Hydrogen site location: inferred from neighbouring sites
$R[F^2 > 2\sigma(F^2)]$ = 0.052	H-atom parameters constrained
$wR(F^2)$ = 0.139	$w = 1/[\sigma^2(F_o^2) + (0.0615P)^2 + 0.2115P]$ where $P = (F_o^2 + 2F_c^2)/3$
S = 1.04	$(\Delta/\sigma)_{max} < 0.001$
1716 reflections	$\Delta\rho_{max}$ = 0.17 e $Å^{-3}$
120 parameters	$\Delta\rho_{min}$ = −0.14 e $Å^{-3}$
78 restraints	Extinction correction: none
Primary atom site location: structure-invariant direct methods	

Special details

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 $F^2 > 2sigma(F^2)$ 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	1.1869 (4)	0.81344 (14)	0.73896 (11)	0.0720 (6)
O2	1.1711 (4)	0.64213 (13)	0.78091 (9)	0.0626 (5)
O3	0.3884 (4)	0.74182 (16)	0.49802 (11)	0.0807 (6)
N1	0.5743 (4)	0.60518 (15)	0.57889 (11)	0.0524 (5)
C1	1.0922 (5)	0.72278 (18)	0.73102 (13)	0.0509 (6)
C2	0.8829 (5)	0.69234 (17)	0.66586 (12)	0.0472 (5)
C3	0.7712 (4)	0.59462 (17)	0.64654 (12)	0.0451 (5)
C4	0.4149 (5)	0.5203 (2)	0.53604 (14)	0.0590 (6)
H4A	0.2245	0.5473	0.5169	0.071*
H4B	0.3813	0.4611	0.5731	0.071*
C5	1.3733 (6)	0.6677 (2)	0.84821 (14)	0.0687 (7)
H5A	1.2941	0.7254	0.8808	0.082*
H5B	1.5615	0.6908	0.8280	0.082*
C6	1.4108 (8)	0.5687 (3)	0.89787 (18)	0.1009 (11)
H6A	1.2285	0.5517	0.9230	0.151*
H6B	1.5615	0.5803	0.9392	0.151*
H6C	1.4668	0.5101	0.8636	0.151*
C7	0.8173 (6)	0.48586 (17)	0.68252 (15)	0.0613 (7)
H7A	0.9800	0.4881	0.7212	0.092*
H7B	0.8593	0.4354	0.6402	0.092*
H7C	0.6425	0.4639	0.7094	0.092*
C8	0.7540 (5)	0.77494 (18)	0.60915 (14)	0.0566 (6)
H8A	0.9063	0.8096	0.5784	0.068*
H8B	0.6477	0.8294	0.6387	0.068*
C9	0.5496 (5)	0.7116 (2)	0.55439 (14)	0.0575 (6)

Atomic displacement parameters (Å^2)

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
O1	0.0810 (13)	0.0532 (10)	0.0810 (12)	−0.0135 (9)	−0.0110 (10)	−0.0072 (9)
O2	0.0747 (12)	0.0589 (10)	0.0531 (9)	−0.0057 (8)	−0.0155 (8)	−0.0015 (8)
O3	0.0725 (13)	0.1035 (15)	0.0649 (11)	0.0129 (10)	−0.0145 (10)	0.0175 (10)
N1	0.0479 (11)	0.0587 (12)	0.0502 (10)	−0.0034 (9)	−0.0018 (8)	−0.0048 (9)
C1	0.0487 (13)	0.0517 (14)	0.0526 (13)	0.0012 (11)	0.0036 (10)	−0.0060 (11)
C2	0.0488 (13)	0.0452 (12)	0.0476 (11)	−0.0003 (10)	0.0020 (10)	−0.0016 (10)
C3	0.0407 (12)	0.0518 (13)	0.0427 (11)	−0.0015 (10)	0.0026 (9)	−0.0025 (10)
C4	0.0465 (13)	0.0757 (17)	0.0549 (13)	−0.0113 (12)	0.0001 (10)	−0.0131 (12)
C5	0.0692 (17)	0.085 (2)	0.0506 (14)	−0.0030 (14)	−0.0114 (12)	−0.0105 (13)
C6	0.138 (3)	0.090 (2)	0.0721 (19)	0.016 (2)	−0.0422 (19)	−0.0024 (17)
C7	0.0724 (16)	0.0497 (14)	0.0615 (14)	−0.0048 (12)	−0.0037 (12)	0.0025 (11)
C8	0.0581 (15)	0.0505 (13)	0.0610 (14)	0.0040 (11)	0.0015 (12)	0.0022 (11)
C9	0.0514 (14)	0.0681 (16)	0.0532 (14)	0.0061 (12)	0.0037 (11)	0.0049 (12)

Geometric parameters (Å, °)

O1—C1	1.211 (3)	C4—H4B	0.9700
O2—C1	1.336 (3)	C5—C6	1.482 (4)
O2—C5	1.450 (3)	C5—H5A	0.9700
O3—C9	1.221 (3)	C5—H5B	0.9700
N1—C9	1.385 (3)	C6—H6A	0.9600
N1—C3	1.411 (3)	C6—H6B	0.9600
N1—C4	1.449 (3)	C6—H6C	0.9600
C1—C2	1.459 (3)	C7—H7A	0.9600
C2—C3	1.349 (3)	C7—H7B	0.9600
C2—C8	1.493 (3)	C7—H7C	0.9600
C3—C7	1.487 (3)	C8—C9	1.496 (3)
C4—C4^{i}	1.521 (4)	C8—H8A	0.9700
C4—H4A	0.9700	C8—H8B	0.9700
C1—O2—C5	117.06 (18)	C6—C5—H5B	110.3
C9—N1—C3	111.19 (18)	H5A—C5—H5B	108.5
C9—N1—C4	121.21 (19)	C5—C6—H6A	109.5
C3—N1—C4	127.58 (19)	C5—C6—H6B	109.5
O1—C1—O2	122.9 (2)	H6A—C6—H6B	109.5
O1—C1—C2	122.7 (2)	C5—C6—H6C	109.5
O2—C1—C2	114.35 (19)	H6A—C6—H6C	109.5
C3—C2—C1	129.6 (2)	H6B—C6—H6C	109.5
C3—C2—C8	109.52 (19)	C3—C7—H7A	109.5
C1—C2—C8	120.90 (19)	C3—C7—H7B	109.5
C2—C3—N1	109.02 (18)	H7A—C7—H7B	109.5
C2—C3—C7	132.5 (2)	C3—C7—H7C	109.5
N1—C3—C7	118.43 (18)	H7A—C7—H7C	109.5
N1—C4—C4^{i}	111.2 (2)	H7B—C7—H7C	109.5
N1—C4—H4A	109.4	C2—C8—C9	103.77 (19)
C4^{i}—C4—H4A	109.4	C2—C8—H8A	111.0
N1—C4—H4B	109.4	C9—C8—H8A	111.0
C4^{i}—C4—H4B	109.4	C2—C8—H8B	111.0
H4A—C4—H4B	108.0	C9—C8—H8B	111.0
O2—C5—C6	107.3 (2)	H8A—C8—H8B	109.0
O2—C5—H5A	110.3	O3—C9—N1	123.7 (2)
C6—C5—H5A	110.3	O3—C9—C8	129.8 (2)
O2—C5—H5B	110.3	N1—C9—C8	106.47 (19)

Symmetry codes: (i) −*x*+1, −*y*+1, −*z*+1.

Hydrogen-bond geometry (Å, °)

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
C7—H7A···O2	0.96	2.31	2.966 (4)	125
C8—H8B···O1^{ii}	0.97	2.71	3.429 (3)	131
C7—H7C···O1^{iii}	0.96	2.55	3.417 (3)	150
C7—H7A···O1^{iv}	0.96	2.71	3.337 (3)	123

Symmetry codes: (ii) $x-1$, y, z; (iii) $-x+3/2$, $y-1/2$, $-z+3/2$; (iv) $-x+5/2$, $y-1/2$, $-z+3/2$.

Fig. 1

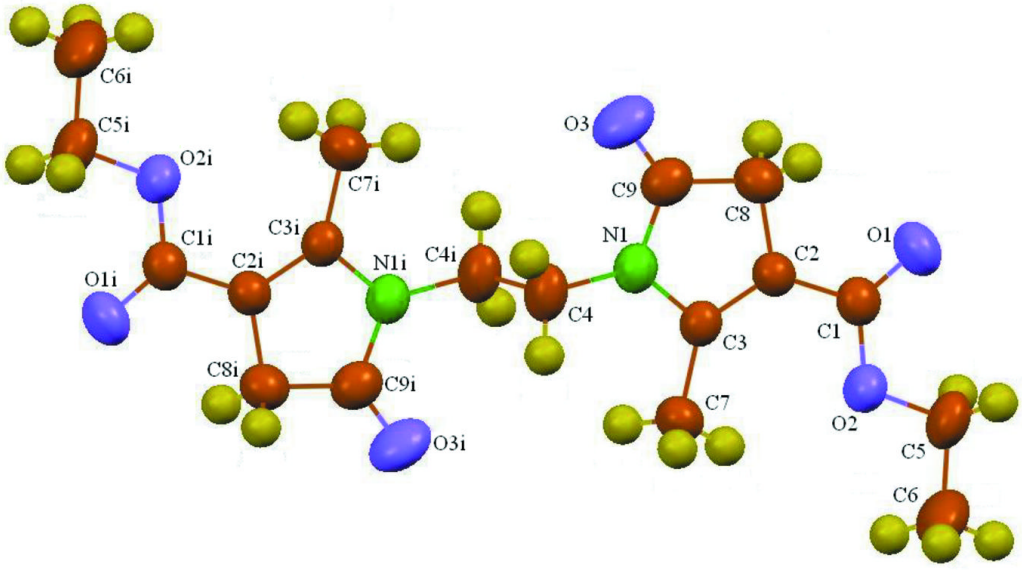


Fig. 2

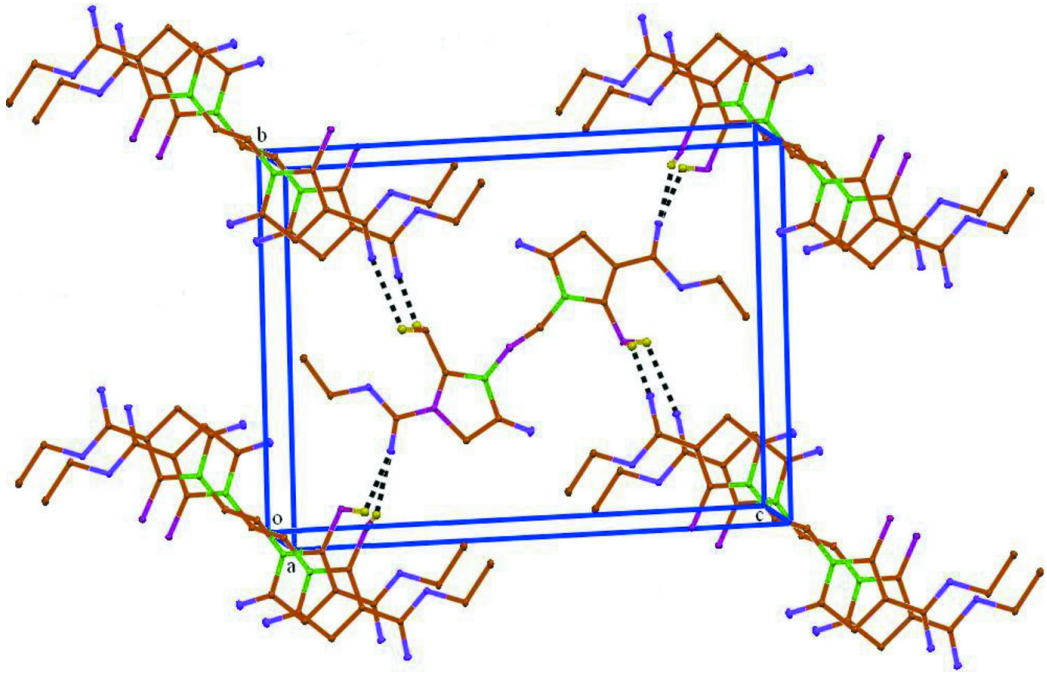
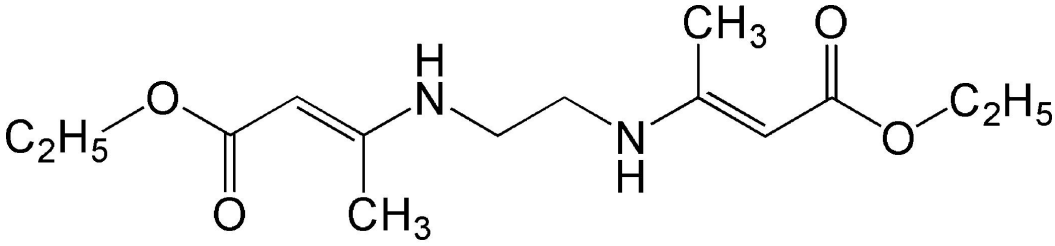
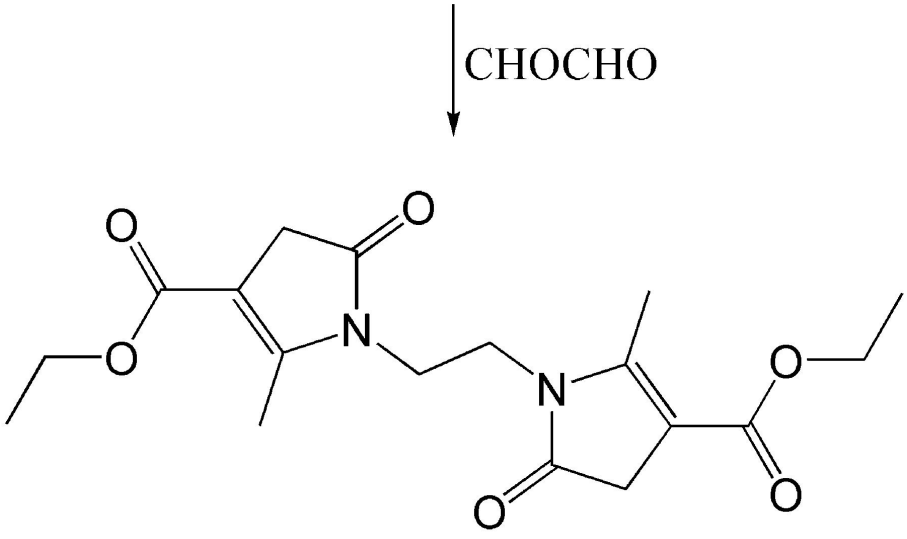


Fig. 3



(I)



(II)