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## Structure Reports

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## 5-(3,4-Dimethoxybenzylidene)-1,3-dimethyl-1,3-diazinane-2,4,6-trione

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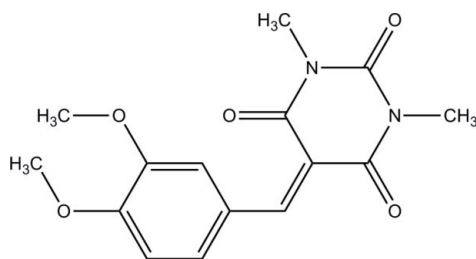
Received 21 November 2011; accepted 8 December 2011

Key indicators: single-crystal X-ray study;  $T = 100$  K; mean  $\sigma(\text{C}-\text{C}) = 0.002$  Å;  $R$  factor = 0.042;  $wR$  factor = 0.113; data-to-parameter ratio = 15.9.

In the title compound,  $\text{C}_{15}\text{H}_{16}\text{N}_2\text{O}_5$ , the dihedral angle between 1,3-diazinane and benzene rings is only  $4.27$  ( $1$ )°. The essentially planar molecular structure is characterized by a short intramolecular  $\text{C}-\text{H}\cdots\text{O}$  separation and by an exceptionally large bond angle of  $138.25$  ( $14$ )° at the bridging methine C atom. The methoxy groups deviate somewhat from the plane of the benzene ring, with  $\text{C}-\text{C}-\text{O}-\text{C}$  torsion angles of  $-15.6$  ( $1$ ) and  $9.17$  ( $6$ )°. In the crystal, molecules form centrosymmetric dimers *via* donor–acceptor  $\pi-\pi$  interactions, with a centroid–centroid distance of  $3.401$  ( $1$ ) Å.

## Related literature

For the biological activity of 1,3-diazinane derivatives, see: Negwar (2001); Tanaka *et al.* (1986, 1988). For the use of pyridine-type ligands in catalysis models, see: Roodt *et al.* (2011); van der Westhuizen *et al.* (2010). For related structures, see: Panchatcharam *et al.* (2009); Rezende *et al.* (2005). For the synthesis, see: Prajapati *et al.* (2006). For standard bond lengths, see: Allen *et al.* (1987).



## Experimental

## Crystal data

$\text{C}_{15}\text{H}_{16}\text{N}_2\text{O}_5$   
 $M_r = 304.3$   
 Triclinic,  $P\bar{1}$   
 $a = 7.3086$  ( $2$ ) Å

$b = 8.4033$  ( $3$ ) Å  
 $c = 11.8705$  ( $5$ ) Å  
 $\alpha = 82.5685$  ( $18$ )°  
 $\beta = 77.6686$  ( $17$ )°

$\gamma = 71.1469$  ( $15$ )°  
 $V = 672.58$  ( $4$ ) Å<sup>3</sup>  
 $Z = 2$   
 Mo  $K\alpha$  radiation

$\mu = 0.11$  mm<sup>-1</sup>  
 $T = 100$  K  
 $0.15 \times 0.12 \times 0.06$  mm

## Data collection

Bruker APEXII CCD diffractometer  
 Absorption correction: multi-scan (SADABS; Bruker, 2008)  
 $T_{\min} = 0.984$ ,  $T_{\max} = 0.994$

12172 measured reflections  
 3233 independent reflections  
 2478 reflections with  $I > 2\sigma(I)$   
 $R_{\text{int}} = 0.032$

## Refinement

$R[F^2 > 2\sigma(F^2)] = 0.042$   
 $wR(F^2) = 0.113$   
 $S = 1.05$   
 3233 reflections

203 parameters  
 H-atom parameters constrained  
 $\Delta\rho_{\text{max}} = 0.32$  e Å<sup>-3</sup>  
 $\Delta\rho_{\text{min}} = -0.30$  e Å<sup>-3</sup>

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

$D-H\cdots A$	$D-H$	$H\cdots A$	$D\cdots A$	$D-H\cdots A$
$\text{C7}-\text{H7}\cdots\text{O3}$	0.93	2.08	2.871 (2)	142

Data collection: APEX2 (Bruker, 2008); cell refinement: SAINT-Plus (Bruker, 2008); data reduction: SAINT-Plus; program(s) used to solve structure: SHELXS97 (Sheldrick, 2008); program(s) used to refine structure: SHELXL97 (Sheldrick, 2008); molecular graphics: DIAMOND (Brandenburg & Putz, 2005); software used to prepare material for publication: WinGX (Farrugia, 1999).

The University of the Free State and Sasol Ltd are gratefully acknowledged, for financial support, and Johannes van Tonder for the NMR data and help with the synthesis of the title compound. Special thanks are due to Professor Andreas Roodt.

Supplementary data and figures for this paper are available from the IUCr electronic archives (Reference: LD2039).

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**supplementary materials**

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## 5-(3,4-Dimethoxybenzylidene)-1,3-dimethyl-1,3-diazinane-2,4,6-trione

M. Gohain, T. J. Muller and B. C. B. Bezuidenhoudt

### Comment

Barbituric acid is the parent compound of barbiturate drugs, although by itself it is not pharmacologically active (Negwar *et al.*, 2001). Benzylidenobarbituric acids are important building block in the synthesis of pyrazolo-[3,4]-1,3-diazinane derivatives which shows broad-spectrum biological activities (Tanaka *et al.*, 1986 and 1988). We also synthesized some of the benzylidene barbituric acids which were successfully used to prepare pyrano[2,3-*d*]- and furopyrano[2,3-*d*] 1,3-diazinane derivatives (Prajapati *et al.*, 2006). The title compound having molecular formula C<sub>15</sub>H<sub>16</sub>N<sub>2</sub>O<sub>5</sub> can be prepared by the condensation of barbituric acid and 4,5-dimethoxybenzaldehyde. The bond C5—C6 of 1.453 (2) Å is longer than C3—C5 bond of 1.365 (2) Å that indicates C3—C5 as a formally double bond. This is in accordance with the literature (Panchatcharam *et al.* 2009 and Rezende *et al.* 2005).

### Experimental

Mixture of *N,N*-dimethylbarbituric acid (0.50 g, 3.2 mmol) and 4,5-dimethoxy benzaldehyde (0.53 g, 3.2 mmol) in ethanol (10 ml) was stirred at room temperature until completion of the reaction (monitored by TLC). The solids that precipitated during the course of the reaction were filtered and washed with diethyl ether (5 ml). The precipitate was subsequently dissolved in hot acetonitrile. Upon cooling to room temperature with a slow evaporation of the acetonitrile the crystals (mp 229–230 °C) suitable for single-crystal X-ray diffraction were obtained.

<sup>1</sup>H NMR (600 MHz): 3.42 (s, 3H, N—Me), 3.43 (s, 3H, N—Me), 3.99 (s, 3H, OMe), 3.40 (s, 3H, OMe), 6.97 (d, 1H), 7.81 (dd, 1H), 8.41 (d, 1H), 8.51(s, 1H).

<sup>13</sup>C {<sup>1</sup>H} NMR (150Mz): 28.5, 29.1, 56.1, 56.2, 110.4, 114.2, 116.6, 125.9, 132.6, 148.4, 151.4, 154.4, 159.2, 161.1, 163.3.

### Refinement

The aromatic H atoms were positioned geometrically and allowed to ride on their parent atoms, with  $U_{\text{iso}}(\text{H}) = 1.2U_{\text{eq}}(\text{parent})$  of the parent atom with a C—H distance of 0.93. The methyl H atoms were placed in geometrically idealized positions and constrained to ride on its parent atoms with  $U_{\text{iso}}(\text{H}) = 1.5U_{\text{eq}}(\text{C})$  and at a distance of 0.96 Å; their torsion angles were optimized from electron density

## Figures

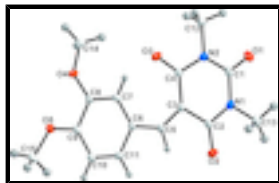


Fig. 1. Diamond representation of the title compound, showing the numbering scheme and displacement ellipsoids (50% probability).

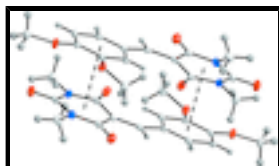


Fig. 2. Diamond representation of the title compound, showing the  $\pi$ - $\pi$  interaction.

## 5-(3,4-Dimethoxybenzylidene)-1,3-dimethyl-1,3-diazinane-2,4,6-trione

### Crystal data

$C_{15}H_{16}N_2O_5$

$M_r = 304.3$

Triclinic,  $P\bar{1}$

Hall symbol: -P 1

$a = 7.3086$  (2) Å

$b = 8.4033$  (3) Å

$c = 11.8705$  (5) Å

$\alpha = 82.5685$  (18)°

$\beta = 77.6686$  (17)°

$\gamma = 71.1469$  (15)°

$V = 672.58$  (4) Å<sup>3</sup>

$Z = 2$

$F(000) = 320$

$D_x = 1.508$  Mg m<sup>-3</sup>

Mo  $K\alpha$  radiation,  $\lambda = 0.71073$  Å

Cell parameters from 3146 reflections

$\theta = 2.6$ – $28.2$ °

$\mu = 0.11$  mm<sup>-1</sup>

$T = 100$  K

Plate, yellow

$0.15 \times 0.12 \times 0.06$  mm

### Data collection

Bruker APEXII CCD  
diffractometer

graphite

$\varphi$  and  $\omega$  scans

Absorption correction: multi-scan  
(*SADABS*; Bruker, 2008)

$T_{\min} = 0.984$ ,  $T_{\max} = 0.994$

12172 measured reflections

3233 independent reflections

2478 reflections with  $I > 2\sigma(I)$

$R_{\text{int}} = 0.032$

$\theta_{\max} = 28$ °,  $\theta_{\min} = 3.2$ °

$h = -9 \rightarrow 9$

$k = -11 \rightarrow 11$

$l = -15 \rightarrow 15$

### Refinement

Refinement on  $F^2$

Least-squares matrix: full

Primary atom site location: structure-invariant direct  
methods

Secondary atom site location: difference Fourier map

$$R[F^2 > 2\sigma(F^2)] = 0.042$$

$$wR(F^2) = 0.113$$

$$S = 1.05$$

3233 reflections

203 parameters

0 restraints

Hydrogen site location: inferred from neighbouring sites

H-atom parameters constrained

$$w = 1/[\sigma^2(F_o^2) + (0.0512P)^2 + 0.239P]$$

$$\text{where } P = (F_o^2 + 2F_c^2)/3$$

$$(\Delta/\sigma)_{\max} = 0.003$$

$$\Delta\rho_{\max} = 0.32 \text{ e } \text{\AA}^{-3}$$

$$\Delta\rho_{\min} = -0.30 \text{ e } \text{\AA}^{-3}$$

### Special details

**Experimental.** The intensity data was collected on a Bruker X8 ApexII 4 K Kappa CCD diffractometer using an exposure time of 15 s/frame. A total of 1821 frames were collected with a frame width of 0.5° covering up to  $\theta = 28.18^\circ$  with 99.7% completeness accomplished.

**Geometry.** All s.u.'s (except the s.u. in the dihedral angle between two l.s. planes) are estimated using the full covariance matrix. The cell s.u.'s are taken into account individually in the estimation of s.u.'s in distances, angles and torsion angles; correlations between s.u.'s in cell parameters are only used when they are defined by crystal symmetry. An approximate (isotropic) treatment of cell s.u.'s is used for estimating s.u.'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  $F^2 > 2\sigma(F^2)$  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 ( $\text{\AA}^2$ )

	<i>x</i>	<i>y</i>	<i>z</i>	$U_{\text{iso}}^*/U_{\text{eq}}$
C1	0.4027 (2)	0.72599 (19)	0.80997 (12)	0.0160 (3)
C2	0.5148 (2)	0.43533 (18)	0.75097 (12)	0.0150 (3)
C3	0.61438 (19)	0.49161 (18)	0.63713 (12)	0.0135 (3)
C4	0.59797 (19)	0.67081 (18)	0.61360 (12)	0.0144 (3)
C5	0.70824 (19)	0.36478 (18)	0.56448 (12)	0.0139 (3)
H5	0.6895	0.2635	0.5979	0.017*
C6	0.82904 (19)	0.34247 (18)	0.44990 (12)	0.0138 (3)
C7	0.89371 (19)	0.46673 (18)	0.37541 (12)	0.0142 (3)
H7	0.8572	0.576	0.3985	0.017*
C8	1.01025 (19)	0.42763 (18)	0.26903 (12)	0.0138 (3)
C9	1.06655 (19)	0.26239 (18)	0.23180 (12)	0.0142 (3)
C10	1.0028 (2)	0.13956 (18)	0.30414 (12)	0.0160 (3)
H10	1.0382	0.0306	0.2807	0.019*
C11	0.8865 (2)	0.17974 (18)	0.41127 (12)	0.0155 (3)
H11	0.8452	0.0963	0.459	0.019*
C12	0.4873 (2)	0.95348 (18)	0.68221 (13)	0.0186 (3)
H12A	0.3864	1.0081	0.6375	0.028*
H12B	0.6109	0.9644	0.6403	0.028*
H12C	0.4551	1.0052	0.7544	0.028*
C13	0.3059 (2)	0.5045 (2)	0.93915 (13)	0.0215 (3)

## supplementary materials

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H13A	0.2832	0.5852	0.9949	0.032*
H13B	0.3826	0.396	0.9664	0.032*
H13C	0.1822	0.498	0.9283	0.032*
C14	0.9864 (2)	0.71432 (18)	0.21141 (14)	0.0203 (3)
H14A	0.8468	0.7402	0.2177	0.03*
H14B	1.0356	0.7815	0.1479	0.03*
H14C	1.0144	0.7384	0.2817	0.03*
C15	1.2176 (2)	0.08217 (19)	0.07552 (14)	0.0215 (3)
H15A	1.29	-0.0087	0.122	0.032*
H15B	1.2928	0.0848	-0.0012	0.032*
H15C	1.0949	0.0657	0.0724	0.032*
N1	0.41279 (17)	0.55732 (16)	0.82856 (10)	0.0161 (3)
N2	0.50181 (17)	0.77434 (15)	0.70439 (10)	0.0151 (3)
O5	1.18050 (15)	0.23929 (13)	0.12565 (9)	0.0181 (2)
O1	0.31031 (16)	0.82697 (14)	0.88192 (9)	0.0229 (3)
O2	0.51855 (15)	0.28912 (13)	0.77753 (9)	0.0216 (3)
O3	0.66023 (15)	0.73491 (13)	0.52101 (9)	0.0210 (3)
O4	1.07951 (14)	0.53932 (13)	0.19183 (9)	0.0174 (2)

### Atomic displacement parameters ( $\text{\AA}^2$ )

	$U^{11}$	$U^{22}$	$U^{33}$	$U^{12}$	$U^{13}$	$U^{23}$
C1	0.0140 (6)	0.0205 (8)	0.0140 (7)	-0.0057 (6)	-0.0022 (5)	-0.0026 (6)
C2	0.0151 (6)	0.0175 (7)	0.0130 (7)	-0.0061 (6)	-0.0020 (5)	-0.0008 (6)
C3	0.0134 (6)	0.0152 (7)	0.0116 (7)	-0.0049 (5)	-0.0022 (5)	0.0006 (5)
C4	0.0129 (6)	0.0161 (7)	0.0135 (7)	-0.0035 (5)	-0.0013 (5)	-0.0022 (6)
C5	0.0138 (6)	0.0149 (7)	0.0135 (7)	-0.0058 (5)	-0.0027 (5)	0.0013 (6)
C6	0.0128 (6)	0.0156 (7)	0.0128 (7)	-0.0041 (5)	-0.0024 (5)	-0.0010 (5)
C7	0.0148 (6)	0.0137 (7)	0.0141 (7)	-0.0048 (5)	-0.0006 (5)	-0.0028 (5)
C8	0.0136 (6)	0.0147 (7)	0.0134 (7)	-0.0061 (5)	-0.0014 (5)	0.0009 (5)
C9	0.0134 (6)	0.0164 (7)	0.0124 (7)	-0.0042 (5)	-0.0015 (5)	-0.0023 (6)
C10	0.0172 (7)	0.0137 (7)	0.0167 (7)	-0.0040 (5)	-0.0024 (5)	-0.0032 (6)
C11	0.0161 (6)	0.0151 (7)	0.0160 (7)	-0.0070 (6)	-0.0023 (5)	0.0012 (6)
C12	0.0216 (7)	0.0140 (7)	0.0201 (8)	-0.0058 (6)	-0.0013 (6)	-0.0034 (6)
C13	0.0228 (8)	0.0279 (9)	0.0138 (8)	-0.0110 (7)	0.0020 (6)	-0.0015 (6)
C14	0.0237 (7)	0.0153 (8)	0.0203 (8)	-0.0067 (6)	0.0005 (6)	-0.0010 (6)
C15	0.0267 (8)	0.0176 (8)	0.0184 (8)	-0.0053 (6)	0.0009 (6)	-0.0067 (6)
N1	0.0171 (6)	0.0192 (7)	0.0118 (6)	-0.0075 (5)	0.0008 (5)	-0.0008 (5)
N2	0.0164 (6)	0.0144 (6)	0.0143 (6)	-0.0055 (5)	-0.0007 (5)	-0.0013 (5)
O5	0.0226 (5)	0.0156 (5)	0.0142 (5)	-0.0063 (4)	0.0041 (4)	-0.0049 (4)
O1	0.0253 (6)	0.0234 (6)	0.0184 (6)	-0.0077 (5)	0.0040 (4)	-0.0085 (5)
O2	0.0284 (6)	0.0173 (6)	0.0177 (6)	-0.0099 (5)	0.0016 (4)	0.0015 (4)
O3	0.0271 (6)	0.0154 (5)	0.0160 (6)	-0.0056 (4)	0.0031 (4)	0.0008 (4)
O4	0.0214 (5)	0.0133 (5)	0.0154 (5)	-0.0067 (4)	0.0033 (4)	-0.0008 (4)

### Geometric parameters ( $\text{\AA}$ , $^\circ$ )

C1—O1	1.2150 (18)	C10—C11	1.385 (2)
C1—N1	1.386 (2)	C10—H10	0.93

C1—N2	1.3897 (19)	C11—H11	0.93
C2—O2	1.2212 (19)	C12—N2	1.467 (2)
C2—N1	1.3826 (19)	C12—H12A	0.96
C2—C3	1.489 (2)	C12—H12B	0.96
C3—C5	1.365 (2)	C12—H12C	0.96
C3—C4	1.466 (2)	C13—N1	1.4716 (19)
C4—O3	1.2235 (17)	C13—H13A	0.96
C4—N2	1.3914 (19)	C13—H13B	0.96
C5—C6	1.453 (2)	C13—H13C	0.96
C5—H5	0.93	C14—O4	1.4331 (19)
C6—C11	1.402 (2)	C14—H14A	0.96
C6—C7	1.413 (2)	C14—H14B	0.96
C7—C8	1.377 (2)	C14—H14C	0.96
C7—H7	0.93	C15—O5	1.4400 (19)
C8—O4	1.3650 (18)	C15—H15A	0.96
C8—C9	1.416 (2)	C15—H15B	0.96
C9—O5	1.3525 (17)	C15—H15C	0.96
C9—C10	1.389 (2)		
O1—C1—N1	121.71 (14)	N2—C12—H12A	109.5
O1—C1—N2	121.60 (14)	N2—C12—H12B	109.5
N1—C1—N2	116.69 (12)	H12A—C12—H12B	109.5
O2—C2—N1	119.59 (13)	N2—C12—H12C	109.5
O2—C2—C3	123.24 (13)	H12A—C12—H12C	109.5
N1—C2—C3	117.16 (13)	H12B—C12—H12C	109.5
C5—C3—C4	127.56 (13)	N1—C13—H13A	109.5
C5—C3—C2	113.69 (13)	N1—C13—H13B	109.5
C4—C3—C2	118.73 (12)	H13A—C13—H13B	109.5
O3—C4—N2	118.37 (14)	N1—C13—H13C	109.5
O3—C4—C3	125.11 (13)	H13A—C13—H13C	109.5
N2—C4—C3	116.51 (13)	H13B—C13—H13C	109.5
C3—C5—C6	138.25 (14)	O4—C14—H14A	109.5
C3—C5—H5	110.9	O4—C14—H14B	109.5
C6—C5—H5	110.9	H14A—C14—H14B	109.5
C11—C6—C7	117.75 (13)	O4—C14—H14C	109.5
C11—C6—C5	115.55 (13)	H14A—C14—H14C	109.5
C7—C6—C5	126.71 (13)	H14B—C14—H14C	109.5
C8—C7—C6	120.66 (13)	O5—C15—H15A	109.5
C8—C7—H7	119.7	O5—C15—H15B	109.5
C6—C7—H7	119.7	H15A—C15—H15B	109.5
O4—C8—C7	124.60 (13)	O5—C15—H15C	109.5
O4—C8—C9	114.77 (12)	H15A—C15—H15C	109.5
C7—C8—C9	120.64 (13)	H15B—C15—H15C	109.5
O5—C9—C10	125.58 (13)	C2—N1—C1	124.99 (13)
O5—C9—C8	115.28 (12)	C2—N1—C13	117.62 (13)
C10—C9—C8	119.14 (13)	C1—N1—C13	117.39 (12)
C11—C10—C9	119.85 (14)	C1—N2—C4	125.56 (13)
C11—C10—H10	120.1	C1—N2—C12	117.23 (12)
C9—C10—H10	120.1	C4—N2—C12	116.91 (12)
C10—C11—C6	121.96 (13)	C9—O5—C15	117.89 (11)

## supplementary materials

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C10—C11—H11	119	C8—O4—C14	116.37 (11)
C6—C11—H11	119		

### *Hydrogen-bond geometry (Å, °)*

<i>D—H···A</i>	<i>D—H</i>	<i>H···A</i>	<i>D···A</i>	<i>D—H···A</i>
C7—H7···O3	0.93	2.08	2.871 (2)	142.



Fig. 1

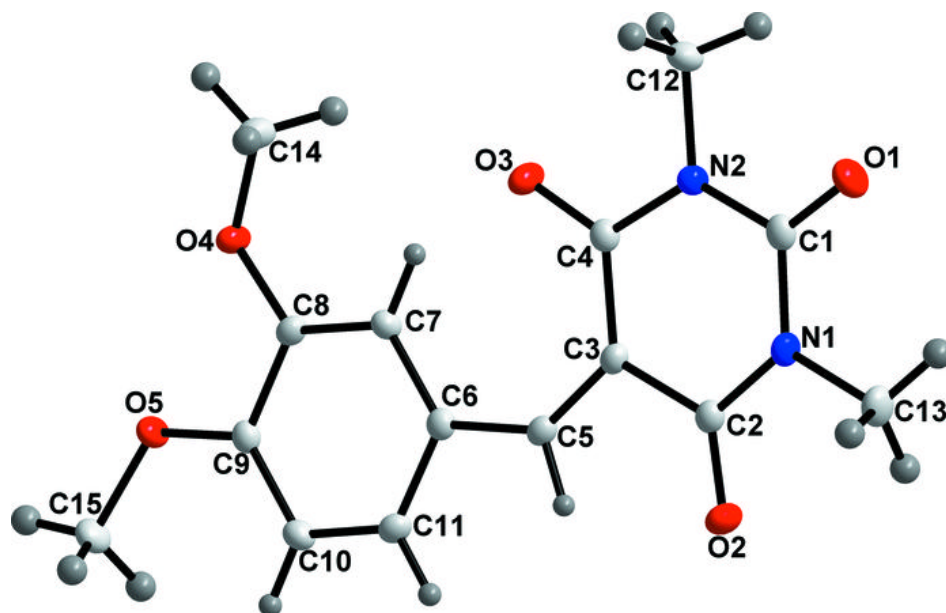


Fig. 2

