

4-*p*-Tolyl-1,3,4,4a,5,6,7,8a-octahydro-2*H*-pyrano[2,3-*d*]pyrimidin-2-oneLei Yan, Yu-Lin Zhu and
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The title compound, C₁₄H₁₈N₂O₂, which was obtained from an extended Biginelli reaction, contains a pyran ring with a chair conformation, a pyrimidine ring with a half-chair conformation and a benzene ring. The crystal structure is stabilized by an intermolecular hydrogen-bond network involving both NH groups and the carbonyl group.

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Key indicators

Single-crystal X-ray study

T = 293 K

Mean σ (C–C) = 0.002 Å

R factor = 0.037

wR factor = 0.112

Data-to-parameter ratio = 13.4

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

Comment

The Biginelli reaction is a classic multicomponent reaction (Biginelli, 1893). It is increasingly attracting the interest of chemists because it shows great advantages of speed, diversity and efficiency in the drug discovery process. Its products, *viz.* functionalized dihydropyrimidines, originally obtained by cyclocondensation of ethyl acetoacetate, benzaldehyde and urea, represent a heterocyclic system of remarkable pharmacological efficiency (Kappe, 2000). The aim of the present work was to study classical and extended Biginelli reactions. Recently, we synthesized a tetrahydropyrimidine scaffold instead of the dihydropyrimidine through an extended Biginelli reaction. We report here the crystal and molecular structure of the title compound, (I).

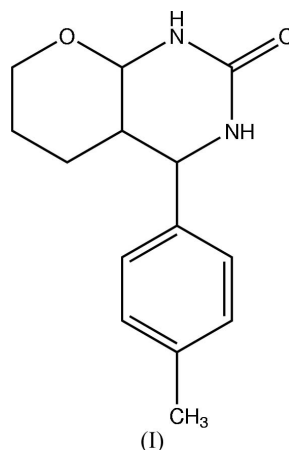


Fig. 1 shows the molecular structure of (I). It contains two fused six-membered rings and a benzene ring. The pyran ring, one of the two fused rings, adopts a chair conformation, while the pyrimidine ring adopts a half-chair conformation due to the two Nsp³ atoms and the C=O at C12. The O8–C9–C10–C5 torsion angle of –54.76 (13)° describes a *cis* configuration of the two fused rings. Also, the C11–C4–C10–C5 torsion angle of –73.58 (14)° suggests a *trans*-C4,C10 configuration. Selected bond distances and angles are listed in Table 1.

The H atoms at both N1 and N3 participate in intermolecular hydrogen bonding, which stabilizes the molecular

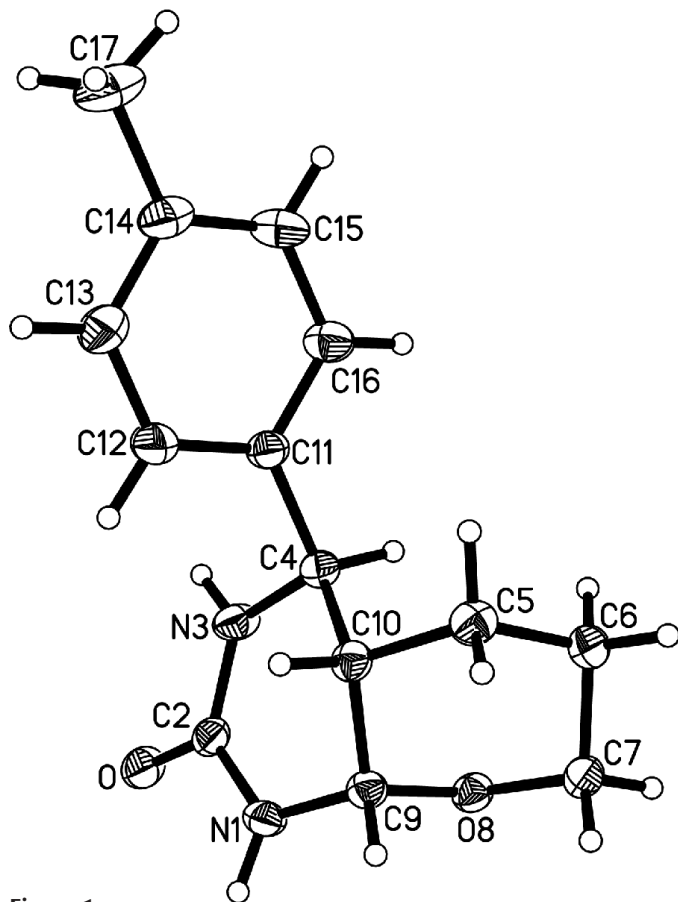


Figure 1

A view of the title molecule, showing the atom-labelling scheme. Displacement ellipsoids are drawn at the 50% probability level. H atoms are represented as small spheres of arbitrary radii.

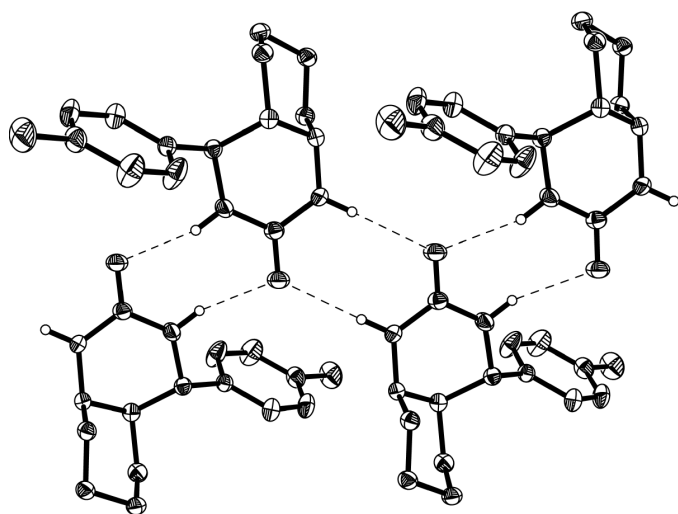


Figure 2

Four molecules of the title compound. For clarity, H atoms bonded to C atoms have been omitted. Dashed lines indicate hydrogen bonds.

packing in the crystal structure of (I) (Fig. 2). In the hydrogen-bond system, the N atoms of the amine group act as hydrogen-bond donors and the O atom of the carbonyl group acts as a hydrogen-bond acceptor (Table 2). A view of the molecular packing down the *b* axis is shown in Fig. 3.

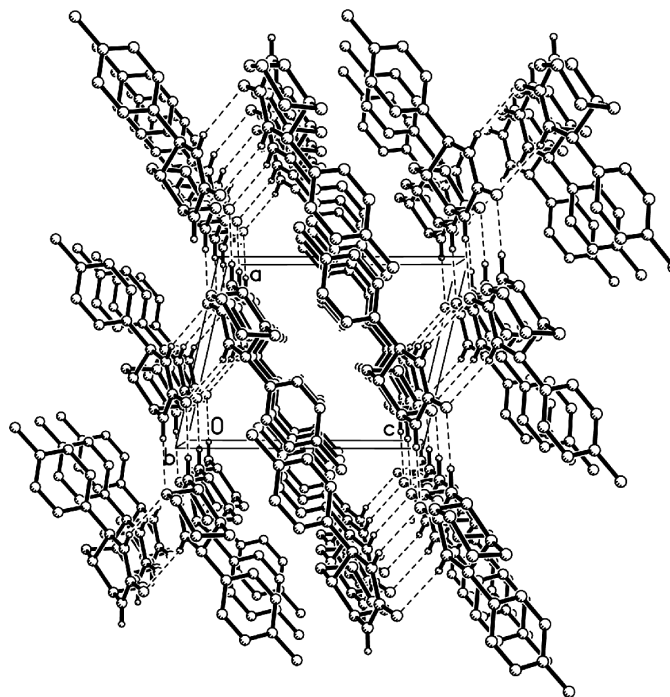


Figure 3

The molecular packing of (I), viewed along the *b* axis. H atoms bonded to C atoms have been omitted for clarity. Dashed lines indicate the hydrogen-bonding interactions.

Experimental

Details of the synthesis and purification of compound (I) will be described elsewhere (Zhu *et al.*, 2005). Crystals appropriate for data collection were obtained by slow evaporation of an *N,N*-dimethyl-formide solution at room temperature over a period of a week.

Crystal data

$C_{14}H_{18}N_2O_2$
 $M_r = 246.30$
 Triclinic, $P\bar{1}$
 $a = 7.7151 (10) \text{ \AA}$
 $b = 8.5541 (11) \text{ \AA}$
 $c = 9.6887 (13) \text{ \AA}$
 $\alpha = 88.516 (2)^\circ$
 $\beta = 76.964 (2)^\circ$
 $\gamma = 83.401 (2)^\circ$
 $V = 618.81 (14) \text{ \AA}^3$

$Z = 2$
 $D_x = 1.322 \text{ Mg m}^{-3}$
 Mo $K\alpha$ radiation
 Cell parameters from 1671 reflections
 $\theta = 2.7\text{--}28.4^\circ$
 $\mu = 0.09 \text{ mm}^{-1}$
 $T = 293 (2) \text{ K}$
 Prism, colourless
 $0.34 \times 0.34 \times 0.30 \text{ mm}$

Data collection

Bruker SMART CCD area-detector diffractometer
 φ and ω scans
 Absorption correction: none
 3223 measured reflections
 2204 independent reflections

1865 reflections with $I > 2\sigma(I)$
 $R_{\text{int}} = 0.013$
 $\theta_{\text{max}} = 25.3^\circ$
 $h = -8 \rightarrow 9$
 $k = -6 \rightarrow 10$
 $l = -11 \rightarrow 11$

Refinement

Refinement on F^2
 $R[F^2 > 2\sigma(F^2)] = 0.037$
 $wR(F^2) = 0.112$
 $S = 1.11$
 2204 reflections
 164 parameters
 H-atom parameters constrained

$w = 1/[\sigma^2(F_o^2) + (0.0609P)^2 + 0.0889P]$
 where $P = (F_o^2 + 2F_c^2)/3$
 $(\Delta/\sigma)_{\text{max}} < 0.001$
 $\Delta\rho_{\text{max}} = 0.20 \text{ e \AA}^{-3}$
 $\Delta\rho_{\text{min}} = -0.14 \text{ e \AA}^{-3}$
 Extinction correction: *SHELXL97*
 Extinction coefficient: 0.032 (7)

Table 1

Selected geometric parameters (Å, °).

| | | | |
|--------------|-------------|---------------|-------------|
| N1—C2 | 1.3573 (18) | N3—C4 | 1.4617 (16) |
| N1—C9 | 1.4251 (17) | C4—C10 | 1.5339 (18) |
| N3—C2 | 1.3453 (17) | C5—C10 | 1.5337 (18) |
| C2—N1—C9 | 122.91 (11) | C5—C10—C4 | 111.88 (11) |
| C2—N3—C4 | 127.05 (11) | C12—C11—C16 | 117.32 (13) |
| N3—C2—N1 | 117.01 (11) | C13—C14—C15 | 116.75 (14) |
| C6—C5—C10 | 112.17 (11) | | |
| O8—C9—C10—C5 | −54.76 (13) | C11—C4—C10—C9 | 163.51 (11) |
| O8—C9—C10—C4 | 69.22 (13) | C11—C4—C10—C5 | −73.58 (14) |

Table 2

Hydrogen-bonding geometry (Å, °).

| <i>D</i> —H... <i>A</i> | <i>D</i> —H | H... <i>A</i> | <i>D</i> ... <i>A</i> | <i>D</i> —H... <i>A</i> |
|-------------------------|-------------|---------------|-----------------------|-------------------------|
| N1—H1...O ⁱ | 0.86 | 2.26 | 3.1177 (16) | 176 |
| N3—H3...O ⁱⁱ | 0.86 | 2.33 | 3.1096 (16) | 151 |

Symmetry codes: (i) 2 − *x*, −*y*, −*z*; (ii) 1 − *x*, −*y*, −*z*.

N-bound H atoms were located in a difference Fourier map and their parameters refined, with N—H distances restrained to 0.86 (1) Å. The positions of the C-bound H atoms were calculated

geometrically and refined using a riding model [C—H = 0.93–0.98 Å], with $U_{\text{iso}}(\text{H}) = 1.2U_{\text{eq}}(\text{C})$.

Data collection: *SMART* (Bruker, 1999); cell refinement: *SAINTE* (Bruker, 1999); data reduction: *SAINTE*; program(s) used to solve structure: *SHELXS97* (Sheldrick, 1997); program(s) used to refine structure: *SHELXL97* (Sheldrick, 1997); molecular graphics: *SHELXTL* (Bruker, 1998); software used to prepare material for publication: *SHELXTL*.

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References

- Biginelli, P. (1893). *Gazz. Chim. Ital.* **23**, 360–413.
- Bruker (1998). *SHELXTL*. Version 5.10. Bruker AXS Inc., Madison, Wisconsin, USA.
- Bruker (1999). *SMART* and *SAINTE*. Bruker AXS Inc., Madison, Wisconsin, USA.
- Kappe, C. O. (2000). *Acc. Chem. Res.* **33**, 879–888.
- Sheldrick, G. M. (1997). *SHELXS97* and *SHELXL97*. University of Göttingen, Germany.
- Zhu, Y.-L., Yan, L., Huang, S.-L. & Pan, Y.-J. (2005). *Eur. J. Org. Chem.* Submitted.