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A second *C*2/*c* polymorph of butobarbitone

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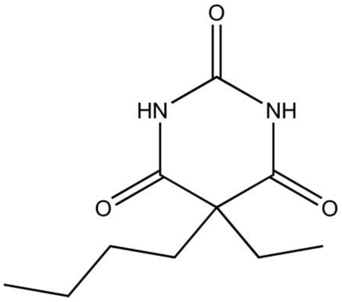
Key indicators: single-crystal X-ray study; T = 150 K; mean σ(C–C) = 0.002 Å; *R* factor = 0.039; *wR* factor = 0.108; data-to-parameter ratio = 17.7.

A second *C*2/*c* polymorph of butobarbitone (5-butyl-5-ethylbarbituric acid), $C_{10}H_{16}N_2O_3$, and a third polymorph overall, has been obtained after prolonged standing of an aqueous solution from which the first polymorph had been crystallized. It has the same space group as the first polymorph at room temperature, but very different cell parameters from both its room-temperature and low-temperature forms (the second polymorph), which are related by a displacive phase transition [Nichol & Clegg (2005). *Acta Cryst.* C**61**, o297–o299] with the low-temperature form having space group $P2_1/n$. The new polymorph shows no phase transition over the temperature range 150–270 K and has a higher density than the first and second polymorphs. N—H···O hydrogen bonds link the molecules into tapes, which interweave in the crystal structure.

Related literature

See Bideau (1971) for the original room-temperature determination of polymorph 1, and Nichol & Clegg (2005) for details of the temperature-induced phase transition to polymorph 2.



Experimental

Crystal data

$C_{10}H_{16}N_2O_3$
M_r = 212.25
Monoclinic, *C*2/*c*
a = 19.519 (4) Å
b = 7.6868 (15) Å
c = 15.117 (3) Å
β = 92.77 (3)°
V = 2265.5 (8) Å^3
Z = 8
Mo $K\alpha$ radiation
μ = 0.09 mm^{-1}
T = 150 (2) K
0.37 × 0.35 × 0.13 mm

Data collection

Nonius KappaCCD diffractometer
Absorption correction: multi-scan (*SADABS*; Sheldrick, 2003)
T_{min} = 0.917, T_{max} = 0.988
12332 measured reflections
2585 independent reflections
1958 reflections with $I > 2\sigma(I)$
R_{int} = 0.034

Refinement

$R[F^2 > 2\sigma(F^2)]$ = 0.039
$wR(F^2)$ = 0.108
S = 1.08
2585 reflections
146 parameters
H atoms treated by a mixture of independent and constrained refinement
$\Delta\rho_{max}$ = 0.28 e Å^{-3}
$\Delta\rho_{min}$ = −0.20 e Å^{-3}

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

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
N1—H1N···O1^{i}	0.882 (19)	2.048 (19)	2.9251 (17)	172.7 (15)
N2—H2N···O2^{ii}	0.883 (16)	1.977 (16)	2.8530 (15)	171.2 (15)

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

Data collection: *COLLECT* (Nonius, 1998); cell refinement: *EVALCCD* (Duisenberg *et al.*, 2003); data reduction: *EVALCCD*; program(s) used to solve structure: *SHELXTL* (Sheldrick, 2001); program(s) used to refine structure: *SHELXTL*; molecular graphics: *DIAMOND* (Brandenburg & Putz, 2004) and *Mercury* (Bruno *et al.*, 2002); software used to prepare material for publication: *SHELXTL* and local programs.

The authors thank Professor Roger Griffin, Newcastle University, for supplying the butobarbitone used in our research.

Supplementary data and figures for this paper are available from the IUCr electronic archives (Reference: FL2163).

References

Bideau, J.-P. (1971). *C. R. Acad. Sc. Paris. Ser. C*, **272**, 757–760.
Brandenburg, K. & Putz, H. (2004). *DIAMOND*. Version 3. University of Bonn, Germany.
Bruno, I. J., Cole, J. C., Edgington, P. R., Kessler, M., Macrae, C. F., McCabe, P., Pearson, J. & Taylor, R. (2002). *Acta Cryst.* B**58**, 389–397.
Duisenberg, A. J. M., Kroon-Batenburg, L. M. J. & Schreurs, A. M. M. (2003). *J. Appl. Cryst.* **36**, 220–229.
Nichol, G. S. & Clegg, W. (2005). *Acta Cryst.* C**61**, o297–o299.
Nonius (1998). *COLLECT*. Nonius BV, Delft, The Netherlands.
Sheldrick, G. M. (2001). *SHELXTL*. Version 6. Bruker AXS Inc., Madison, Wisconsin, USA.
Sheldrick, G. M. (2003). *SADABS*. University of Göttingen, Germany.

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supplementary materials

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A second *C*2/*c* polymorph of butobarbitone

G. S. Nichol and **W. Clegg**

Comment

We have previously reported a temperature-induced phase transition in butobarbitone, (I), with the room-temperature structure determined in space group *C*2/*c* (polymorph 1, originally reported by Bideau, 1971), while at 120 K the space group is $P2_1/n$ (polymorph 2, Nichol & Clegg, 2005). We report here a third polymorph of (I). The space group of this third polymorph is *C*2/*c* with *Z*' = 1 but the unit-cell parameters are quite different from those of the previous structures.

The unit cell volume in this new polymorph is 2265.5 (8) Å^3 and this is lower than the $P2_1/n$ polymorph at low temperature [which is 2356.2 (4) Å^3]. A unit cell determination at 270 K on the same crystal showed only the small expected expansion in the cell parameters and so this third polymorph does not appear to undergo a phase transition within the temperature range from 150 to 270 K.

The molecular structure of (I) is shown in Fig. 1. In common with both reported polymorphs, $R^2_2(6)$ N–H···O hydrogen bonding interactions link the butobarbitone molecules into an infinite hydrogen-bonded tape. Differences are found in the crystal packing. As shown in Fig. 2 the hydrogen-bonded tapes interweave, while in the previously-reported polymorphs the ribbons form roughly parallel stacks.

The higher density of this new polymorph, and the fact that it was formed slowly from a solution from which the first polymorph had already been obtained more quickly, suggest that this is the thermodynamically more stable form.

Experimental

Equimolar amounts of butobarbitone and ammonium carbonate were dissolved in distilled water and heated until boiling. Small colourless crystals of polymorph 1 grew over a period of two days on standing at room temperature in a sealed sample vial. Large colourless crystals of this new polymorph 3 (I) were observed after the sample vial had stood undisturbed for 17 months. Some of the smaller crystals were also still present in the vial.

Refinement

All H atoms were located in a difference map. CH_2 H atoms were then idealized (C—H = 0.99 Å) and refined as riding with $U_{iso}(H) = 1.2U_{eq}(C)$. The methyl H atoms were geometrically positioned (C—H = 0.98 Å) and refined as riding with $U_{iso}(H) = 1.5U_{eq}(C)$ and free rotation about the C—C bond. Nitrogen-bound H atoms were freely refined, giving N—H distances of 0.882 (19) and 0.883 (16) Å.

Figures

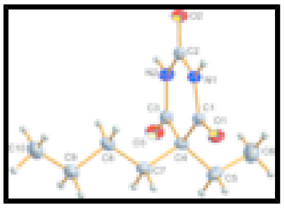

Fig. 1. The molecular structure of (I) with displacement ellipsoids at the 50% probability level and hydrogen atoms as small spheres.

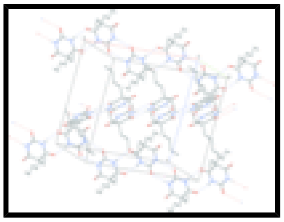

Fig. 2. The interweaving hydrogen-bonded tapes found in (I).

5-butyl-5-ethylbarbituric acid

Crystal data

$C_{10}H_{16}N_2O_3$

M_r = 212.25

Monoclinic, *C*2/*c*

Hall symbol: -C 2yc

a = 19.519 (4) Å

b = 7.6868 (15) Å

c = 15.117 (3) Å

β = 92.77 (3)°

V = 2265.5 (8) $Å^3$

Z = 8

F_{000} = 912

D_x = 1.245 Mg m^{-3}

Mo *K*α radiation
λ = 0.71073 Å

Cell parameters from 6280 reflections

θ = 2.5–27.5°

μ = 0.09 mm^{-1}

T = 150 (2) K

Block, colourless

0.37 × 0.35 × 0.13 mm

Data collection

Nonius KappaCCD
diffractometer

Radiation source: sealed tube

Monochromator: graphite

T = 150(2) K

φ and ω scans

Absorption correction: multi-scan
(SADABS; Sheldrick, 2003)

T_{min} = 0.917, T_{max} = 0.988

12332 measured reflections

2585 independent reflections

1958 reflections with $I > 2\sigma(I)$

R_{int} = 0.034

θ_{max} = 27.5°

θ_{min} = 4.8°

$h = -25 \rightarrow 25$

$k = -9 \rightarrow 9$

$l = -18 \rightarrow 19$

Refinement

Refinement on F^2

Secondary atom site location: difference Fourier map

Least-squares matrix: full

$R[F^2 > 2\sigma(F^2)] = 0.039$

$wR(F^2) = 0.108$

$S = 1.08$

2585 reflections

146 parameters

Primary atom site location: structure-invariant direct methods

Hydrogen site location: difference Fourier map

H atoms treated by a mixture of independent and constrained refinement

$w = 1/[\sigma^2(F_o^2) + (0.0526P)^2 + 1.0348P]$
where $P = (F_o^2 + 2F_c^2)/3$

$(\Delta/\sigma)_{max} < 0.001$

$\Delta\rho_{max} = 0.28$ e Å^{-3}

$\Delta\rho_{min} = -0.20$ e Å^{-3}

Extinction correction: none

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.

Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters (Å^2)

	x	*y*	*z*	U_{iso}*/U_{eq}
O1	0.47791 (4)	0.62343 (12)	0.40519 (6)	0.0242 (2)
O2	0.33382 (5)	0.26565 (12)	0.54285 (6)	0.0262 (2)
O3	0.24583 (5)	0.55071 (13)	0.30083 (6)	0.0296 (3)
N1	0.40634 (5)	0.43550 (14)	0.46817 (7)	0.0191 (2)
H1N	0.4389 (9)	0.411 (2)	0.5089 (11)	0.035 (4)*
N2	0.29072 (6)	0.40867 (15)	0.42063 (7)	0.0218 (3)
H2N	0.2499 (8)	0.364 (2)	0.4287 (10)	0.028 (4)*
C1	0.42106 (6)	0.55569 (16)	0.40434 (8)	0.0176 (3)
C2	0.34256 (6)	0.36347 (16)	0.48085 (8)	0.0188 (3)
C3	0.29594 (6)	0.51903 (16)	0.34869 (8)	0.0199 (3)
C4	0.36613 (6)	0.59673 (16)	0.33256 (8)	0.0191 (3)
C5	0.35829 (7)	0.79708 (18)	0.32390 (10)	0.0275 (3)
H5A	0.4040	0.8482	0.3148	0.033*
H5B	0.3287	0.8230	0.2705	0.033*
C6	0.32798 (10)	0.8851 (2)	0.40337 (12)	0.0442 (4)
H6A	0.2826	0.8362	0.4128	0.066*
H6B	0.3238	1.0104	0.3922	0.066*
H6C	0.3581	0.8655	0.4562	0.066*
C7	0.38930 (7)	0.52140 (18)	0.24327 (8)	0.0237 (3)
H7A	0.3550	0.5555	0.1960	0.028*
H7B	0.4333	0.5768	0.2296	0.028*
C8	0.39867 (7)	0.32317 (18)	0.23944 (9)	0.0251 (3)
H8A	0.4367	0.2882	0.2814	0.030*
H8B	0.3563	0.2653	0.2577	0.030*
C9	0.41447 (8)	0.2637 (2)	0.14602 (9)	0.0295 (3)
H9A	0.3743	0.2895	0.1057	0.035*
H9B	0.4536	0.3324	0.1257	0.035*

C10	0.43166 (9)	0.0710 (2)	0.13864 (10)	0.0362 (4)
H10A	0.4708	0.0433	0.1792	0.054*
H10B	0.4433	0.0444	0.0778	0.054*
H10C	0.3919	0.0014	0.1542	0.054*

Atomic displacement parameters (Å^2)

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
O1	0.0158 (5)	0.0300 (5)	0.0263 (5)	−0.0055 (4)	−0.0038 (4)	0.0070 (4)
O2	0.0219 (5)	0.0330 (5)	0.0233 (5)	−0.0069 (4)	−0.0026 (4)	0.0092 (4)
O3	0.0184 (5)	0.0384 (6)	0.0309 (6)	−0.0024 (4)	−0.0084 (4)	0.0086 (4)
N1	0.0140 (5)	0.0235 (6)	0.0194 (6)	−0.0019 (4)	−0.0039 (4)	0.0042 (4)
N2	0.0128 (5)	0.0286 (6)	0.0237 (6)	−0.0060 (5)	−0.0021 (4)	0.0050 (5)
C1	0.0148 (6)	0.0201 (6)	0.0179 (6)	−0.0002 (5)	0.0002 (5)	0.0002 (5)
C2	0.0161 (6)	0.0200 (6)	0.0201 (6)	−0.0011 (5)	−0.0012 (5)	−0.0006 (5)
C3	0.0165 (6)	0.0219 (6)	0.0209 (6)	−0.0001 (5)	−0.0025 (5)	0.0005 (5)
C4	0.0159 (6)	0.0223 (6)	0.0187 (6)	−0.0015 (5)	−0.0028 (5)	0.0044 (5)
C5	0.0271 (7)	0.0232 (7)	0.0314 (8)	−0.0025 (6)	−0.0083 (6)	0.0070 (6)
C6	0.0592 (11)	0.0287 (8)	0.0438 (10)	0.0079 (8)	−0.0087 (8)	−0.0040 (7)
C7	0.0215 (7)	0.0309 (7)	0.0185 (6)	−0.0037 (6)	−0.0014 (5)	0.0038 (5)
C8	0.0236 (7)	0.0318 (7)	0.0196 (7)	−0.0042 (6)	−0.0012 (5)	0.0006 (6)
C9	0.0304 (8)	0.0358 (8)	0.0222 (7)	−0.0036 (7)	0.0010 (6)	−0.0013 (6)
C10	0.0400 (9)	0.0391 (9)	0.0293 (8)	−0.0026 (7)	0.0004 (7)	−0.0060 (6)

Geometric parameters (Å, °)

O1—C1	1.2253 (15)	C6—H6A	0.980
O2—C2	1.2201 (15)	C6—H6B	0.980
O3—C3	1.2129 (16)	C6—H6C	0.980
N1—H1N	0.882 (19)	C7—H7A	0.990
N1—C1	1.3762 (16)	C7—H7B	0.990
N1—C2	1.3842 (16)	C7—C8	1.536 (2)
N2—H2N	0.883 (16)	C8—H8A	0.990
N2—C2	1.3727 (17)	C8—H8B	0.990
N2—C3	1.3869 (17)	C8—C9	1.5300 (18)
C1—C4	1.5212 (18)	C9—H9A	0.990
C3—C4	1.5249 (17)	C9—H9B	0.990
C4—C5	1.5526 (18)	C9—C10	1.524 (2)
C4—C7	1.5560 (18)	C10—H10A	0.980
C5—H5A	0.990	C10—H10B	0.980
C5—H5B	0.990	C10—H10C	0.980
C5—C6	1.523 (2)		
H1N—N1—C1	117.5 (11)	C5—C6—H6C	109.5
H1N—N1—C2	116.2 (11)	H6A—C6—H6B	109.5
C1—N1—C2	125.81 (11)	H6A—C6—H6C	109.5
H2N—N2—C2	116.7 (10)	H6B—C6—H6C	109.5
H2N—N2—C3	116.7 (10)	C4—C7—H7A	108.2
C2—N2—C3	126.52 (11)	C4—C7—H7B	108.2

O1—C1—N1	119.84 (12)	C4—C7—C8	116.23 (11)
O1—C1—C4	121.74 (11)	H7A—C7—H7B	107.4
N1—C1—C4	118.40 (11)	H7A—C7—C8	108.2
O2—C2—N1	120.86 (12)	H7B—C7—C8	108.2
O2—C2—N2	122.65 (11)	C7—C8—H8A	109.4
N1—C2—N2	116.49 (11)	C7—C8—H8B	109.4
O3—C3—N2	120.19 (12)	C7—C8—C9	111.15 (11)
O3—C3—C4	122.06 (11)	H8A—C8—H8B	108.0
N2—C3—C4	117.75 (11)	H8A—C8—C9	109.4
C1—C4—C3	114.35 (10)	H8B—C8—C9	109.4
C1—C4—C5	109.21 (11)	C8—C9—H9A	108.7
C1—C4—C7	108.50 (10)	C8—C9—H9B	108.7
C3—C4—C5	108.45 (10)	C8—C9—C10	114.38 (12)
C3—C4—C7	107.10 (11)	H9A—C9—H9B	107.6
C5—C4—C7	109.12 (11)	H9A—C9—C10	108.7
C4—C5—H5A	108.6	H9B—C9—C10	108.7
C4—C5—H5B	108.6	C9—C10—H10A	109.5
C4—C5—C6	114.50 (12)	C9—C10—H10B	109.5
H5A—C5—H5B	107.6	C9—C10—H10C	109.5
H5A—C5—C6	108.6	H10A—C10—H10B	109.5
H5B—C5—C6	108.6	H10A—C10—H10C	109.5
C5—C6—H6A	109.5	H10B—C10—H10C	109.5
C5—C6—H6B	109.5		

C2—N1—C1—O1	172.55 (12)	O3—C3—C4—C1	174.50 (12)
C2—N1—C1—C4	−9.21 (18)	O3—C3—C4—C5	52.38 (16)
C3—N2—C2—O2	179.71 (12)	O3—C3—C4—C7	−65.26 (16)
C3—N2—C2—N1	−0.05 (19)	N2—C3—C4—C1	−5.71 (16)
C1—N1—C2—O2	−175.46 (12)	N2—C3—C4—C5	−127.83 (12)
C1—N1—C2—N2	4.30 (18)	N2—C3—C4—C7	114.53 (12)
C2—N2—C3—O3	−178.94 (13)	C1—C4—C5—C6	−68.15 (15)
C2—N2—C3—C4	1.27 (19)	C3—C4—C5—C6	57.05 (16)
O1—C1—C4—C3	−172.44 (12)	C7—C4—C5—C6	173.39 (12)
O1—C1—C4—C5	−50.74 (16)	C1—C4—C7—C8	61.94 (14)
O1—C1—C4—C7	68.10 (15)	C3—C4—C7—C8	−61.97 (14)
N1—C1—C4—C3	9.35 (16)	C5—C4—C7—C8	−179.17 (11)
N1—C1—C4—C5	131.05 (12)	C4—C7—C8—C9	174.37 (11)
N1—C1—C4—C7	−110.10 (12)	C7—C8—C9—C10	174.09 (12)

Hydrogen-bond geometry (Å, °)

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
N1—H1N···O1^{i}	0.882 (19)	2.048 (19)	2.9251 (17)	172.7 (15)
N2—H2N···O2^{ii}	0.883 (16)	1.977 (16)	2.8530 (15)	171.2 (15)
C5—H5B···O3^{iii}	0.99	2.49	3.3341 (19)	143

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

Fig. 1

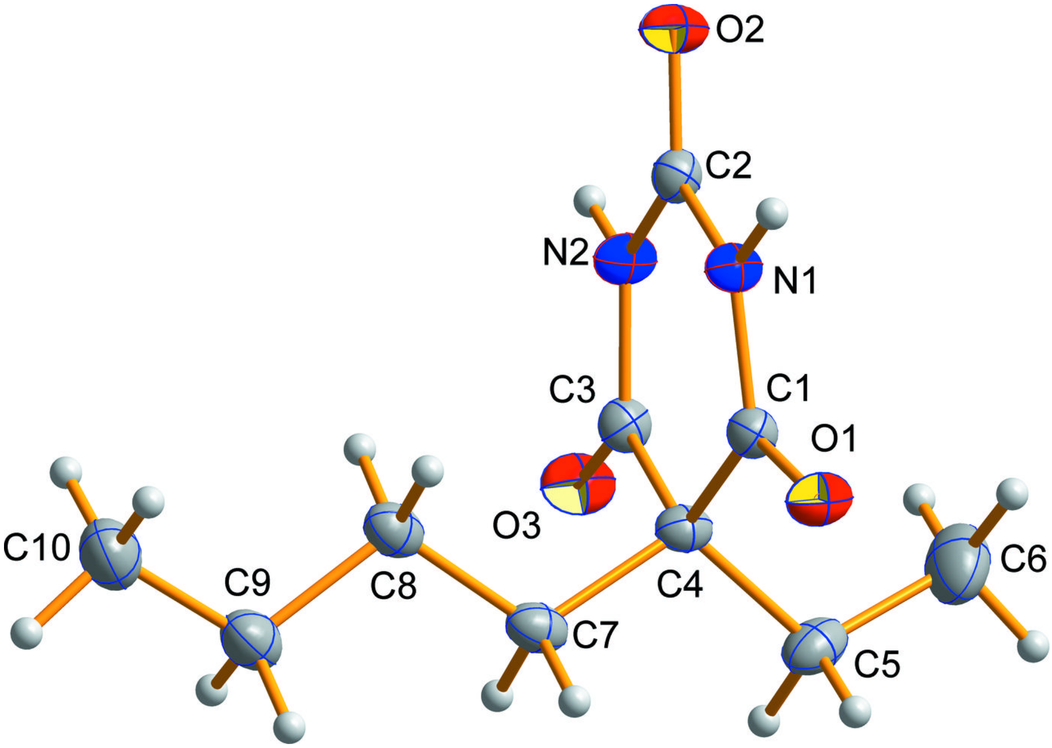

Fig. 2