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3a,6a-Dipropoxycarbonylglycoluril (dipropyl 2,5-dioxoperhydroimidazo-[4,5-*d*]imidazole-3a,6a-dicarboxylate)

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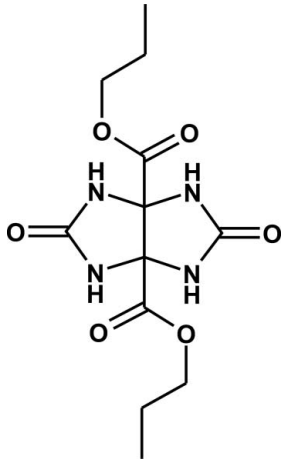
Key indicators: single-crystal X-ray study; T = 300 K; mean σ(C–C) = 0.002 Å; disorder in main residue; R factor = 0.056; wR factor = 0.152; data-to-parameter ratio = 15.6.

The title compound, $C_{12}H_{18}N_4O_6$, exhibits a hydrogen-bonding network, which contains intermolecular classical N—H···O=C(imidazolone rings) hydrogen bonds and, in addition, intermolecular C—H···O interactions that stabilize the crystal structure. Two ethyl groups are each disordered over two positions, with site occupancy factors in a ratio of *ca* 3:2.

Related literature

For related literature, see: Burnett *et al.* (2003); Chen *et al.* (2007); Himes *et al.* (1978); Hof *et al.* (2002); Isaacs & Witt (2002); Kim *et al.* (2000); Li *et al.* (1994); Moon *et al.* (2003); Rowan *et al.* (1999); Wang *et al.* (2006); Wu *et al.* (2002).



Experimental

Crystal data

$C_{12}H_{18}N_4O_6$
M_r = 314.30
Orthorhombic, *Pbca*
a = 13.3013 (7) Å
b = 11.5852 (6) Å
c = 19.9867 (10) Å
V = 3079.9 (3) Å^3
Z = 8
Mo $K\alpha$ radiation
μ = 0.11 mm^{-1}
T = 300 (2) K
0.20 × 0.20 × 0.10 mm

Data collection

Bruker SMART 4K CCD area-detector diffractometer
Absorption correction: none
20966 measured reflections
3752 independent reflections
2887 reflections with $I > 2\sigma(I)$
R_{int} = 0.060

Refinement

$R[F^2 > 2\sigma(F^2)]$ = 0.056
$wR(F^2)$ = 0.152
S = 1.04
3752 reflections
241 parameters
45 restraints
H-atom parameters constrained
$\Delta\rho_{max}$ = 0.41 e Å^{-3}
$\Delta\rho_{min}$ = −0.25 e Å^{-3}

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

D—H···A	D—H	H···A	D···A	D—H···A
C10—H10B···O3^{i}	0.97	2.56	3.494 (3)	163
N4—H4···O1^{ii}	0.86	2.09	2.8459 (16)	146
N3—H3···O1^{iii}	0.86	2.20	3.0134 (17)	157
N2—H2···O2^{iv}	0.86	2.09	2.9265 (16)	163
N1—H1···O2^{v}	0.86	1.98	2.8320 (17)	174

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

Data collection: *SMART* (Bruker, 1997); cell refinement: *SAINT* (Bruker, 1999); data reduction: *SAINT*; program(s) used to solve structure: *SHELXS97* (Sheldrick, 1997); program(s) used to refine structure: *SHELXL97* (Sheldrick, 1997); molecular graphics: *PLATON* (Spek, 2003); software used to prepare material for publication: *SHELXTL* (Bruker, 1997).

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Supplementary data and figures for this paper are available from the IUCr electronic archives (Reference: RK2021).

References

Bruker (1997). *SMART* (Version 5.054) and *SHELXTL* (Version 5.10). Bruker AXS Inc., Madison, Wisconsin, USA.
Bruker (1999). *SAINT*. Version 6.01. Bruker AXS Inc., Madison, Wisconsin, USA.
Burnett, C. A., Lagona, J., Wu, A. X., Shaw, J. A., Coady, D., Fettinger, J. C., Day, A. I. & Isaacs, L. (2003). *Tetrahedron*, **59**, 1961–1970.
Chen, Y. F., She, N. F., Meng, X. G., Yin, G. D., Wu, A. X. & Isaacs, L. (2007). *Org. Lett.* **9**, 1899–1902.
Himes, V. L., Hubbard, C. R., Mighell, A. D. & Fatiadi, A. J. (1978). *Acta Cryst.* B**34**, 3102–3104.
Hof, F., Craig, S. L., Nuckolls, C. & Rebek, J. Jr (2002). *Angew. Chem. Int. Ed.* **41**, 1488–1508.
Isaacs, L. & Witt, D. (2002). *Angew. Chem. Int. Ed.* **41**, 1905–1907.
Kim, J., Jung, I.-S., Kim, S.-Y., Lee, E., Kang, J.-K., Sakamoto, S., Yamaguchi, K. & Kim, K. (2000). *J. Am. Chem. Soc.* **122**, 540–541.
Li, N., Maluendes, S., Blessing, R. H., Dupuis, M., Moss, G. R. & Detitta, G. T. (1994). *J. Am. Chem. Soc.* **116**, 6494–6507.
Moon, K., Chen, W. Z., Ren, T. & Kaifer, A. E. (2003). *CrystEngComm*, **5**, 451–453.
Rowan, A. E., Elemans, J. A. A. W. & Nolte, R. J. M. (1999). *Acc. Chem. Res.* **32**, 995–1006.
Sheldrick, G. M. (1997). *SHELXL97* and *SHELXS97*. University of Göttingen, Germany.
Spek, A. L. (2003). *J. Appl. Cryst.* **36**, 7–13.
Wang, Z. G., Zhou, B. H., Chen, Y. F., Yin, G. D., Li, Y. T., Wu, A. X. & Isaacs, L. (2006). *J. Org. Chem.* **71**, 4502–4508.
Wu, A. X., Fettinger, J. C. & Isaacs, L. (2002). *Tetrahedron*, **58**, 9769–9777.

supplementary materials

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3a,6a-Dipropoxycarbonylglycoluril (dipropyl 2,5-dioxoperhydroimidazo[4,5-*d*]imidazole-3a,6a-dicarboxylate)

Y.-Z. Wang, M. Gao and L.-P. Cao

Comment

Glycoluril –$C_4H_6N_4O_2$– skeleton moiety of the title compound (scheme 1) is an important building block for both molecular and supramolecular chemistry. Its derivatives have been used as the basis for molecular capsules (Hof *et al.*, 2002), molecular clips (Rowan *et al.*, 1999), self-complementary facial amphiphiles (Isaacs & Witt, 2002), and the cucurbit[*n*]uril (CB[*n*]) family (Kim *et al.*, 2000), and its utilization has been explored as a platform for studies of crystal engineering (Wang *et al.*, 2006; Chen *et al.*, 2007). Despite a variety of crystal structures reported for a number of its derivatives, relatively few crystal structures are known for glycoluril derivatives without N-substituents, which exhibit two different H-bonded types (scheme 2). The mode A have been found for (*R* = H) (Li *et al.*, 1994), (*R* = CH_3) (Himes *et al.*, 1978), (*R* = Ph) (Wu *et al.*, 2002), and so on, but the mode B, so far was only observed in the (*R* = Ph) (Moon *et al.*, 2003). Herein, we report the crystal structure of the title compound with *R* = –COO–*n*-C_3H_7) (Fig. 1), which exhibits the scarce mode B of hydrogen bonding (scheme 2).

In the crystal structure, the two-dimension hydrogen bonding network, shown in Fig. 2, is based on the formation of eight-membered rings and ten-membered rings H-bonding motifs. This is fully according with the scarce mode B of hydrogen bonding (scheme 2). While the mode A, reported in the crystal structure of the (scheme 2, *R* = H, CH_3, Ph), is only made up of eight-membered rings H-bonding motifs. In addition, intermolecular C10–H10B···O3 interactions (see table) can stabilize the crystal structure.

Experimental

The title compound was synthesized according to literature procedure (Burnett *et al.*, 2003) in 52% isolated yield. Crystals appropriate for data collection were obtained by slow evaporation of CH_3OH solution at room temperature.

Refinement

Two ethyl group were found to be disordered over two orientations. The occupancies of the disordered positions C7/ C7' and C8/ C8' refined to 0.60 (4) / 0.40 (4), C11 / C11' and C12 / C12', refined to 0.609 (7) / 0.391 (7). In addition, there is also some disorder in the H atoms of C6 and C10 oweing to disorder of its nearby ethyl group. All other H-atoms bound to carbon were positioned geometrically idealized positions and constrained to ride on their parent atoms, with d(C–H) = 0.97 Å, U_{iso} = 1.2U_{eq} (C) for CH_2 and 0.96 Å, U_{iso} = 1.5U_{eq} (C) for CH_3 atoms, and with d(N–H) = 0.86 Å, U_{iso} (H) = 1.2U_{eq} (N).

Figures

Fig. 1. The molecular structure of the title compound with the atom-labelling scheme. Atoms of the minor disorder components are omitted for clarity.

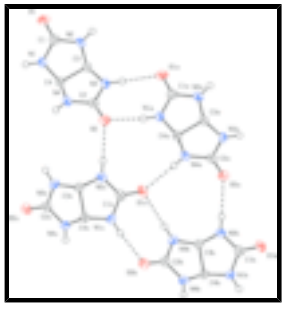


Fig. 2. The unique hydrogen bonding network in the crystal structure of the title compound. H-bonds drawn as dashed lines. *R* = –COO–*n*-C_3H_7) moities are omitted for simplicity.

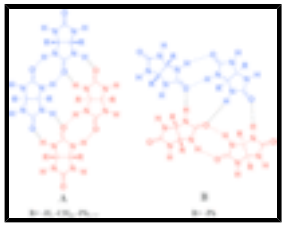


Fig. 3. The two different hydrogen bonding types for glycoluril derivatives without N-substituents.

dipropyl 2,5-dioxoperhydroimidazo[4,5-*d*]imidazole-3a,6a-dicarboxylate

Crystal data

$C_{12}H_{18}N_4O_6$	F_{000} = 1328
M_r = 314.30	D_x = 1.356 Mg m^{-3}
Orthorhombic, *Pbca*	Mo *K*α radiation λ = 0.71073 Å
Hall symbol: -P 2ac 2ab	Cell parameters from 5455 reflections
a = 13.3013 (7) Å	θ = 2.5–27.1º
b = 11.5852 (6) Å	μ = 0.11 mm^{-1}
c = 19.9867 (10) Å	*T* = 300 (2) K
V = 3079.9 (3) Å^3	Plate, colorless
Z = 8	0.20 × 0.20 × 0.10 mm

Data collection

Bruker SMART 4K CCD area-detector diffractometer	2887 reflections with $I > 2\sigma(I)$
Radiation source: fine-focus sealed tube	R_{int} = 0.060
Monochromator: graphite	θ_{max} = 28.3º
T = 300(2) K	θ_{min} = 2.0º

φ and ω scans
Absorption correction: none
20966 measured reflections
3752 independent reflections

$h = -11 \rightarrow 17$
$k = -15 \rightarrow 15$
$l = -26 \rightarrow 26$

Refinement

Refinement on F^2
Least-squares matrix: full
$R[F^2 > 2\sigma(F^2)] = 0.056$
$wR(F^2) = 0.152$
$S = 1.04$
3752 reflections
241 parameters
45 restraints
Primary atom site location: structure-invariant direct methods

Secondary atom site location: difference Fourier map
Hydrogen site location: inferred from neighbouring sites
H-atom parameters constrained
?
$(\Delta/\sigma)_{max} = 0.001$
$\Delta\rho_{max} = 0.41$ e Å^{-3}
$\Delta\rho_{min} = -0.25$ 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.

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 (Å^2)

	x	*y*	*z*	U_{iso}*/U_{eq}	Occ. (<1)
C1	0.75770 (11)	−0.07104 (12)	0.27047 (7)	0.0268 (3)	
C2	0.99691 (11)	0.11085 (12)	0.25548 (8)	0.0287 (3)	
C3	0.91169 (11)	−0.02387 (12)	0.31998 (8)	0.0251 (3)	
C4	0.84418 (10)	0.08686 (12)	0.31140 (8)	0.0260 (3)	
C5	0.93005 (12)	−0.06780 (13)	0.39124 (8)	0.0332 (4)	
C6	0.85324 (15)	−0.09772 (19)	0.49676 (9)	0.0519 (5)	
H6A	0.9205	−0.0884	0.5148	0.062*	0.60 (4)
H6B	0.8390	−0.1798	0.4947	0.062*	0.60 (4)
H6C	0.9199	−0.0914	0.5161	0.062*	0.40 (4)
H6D	0.8322	−0.1779	0.4975	0.062*	0.40 (4)
C7	0.7798 (6)	−0.0418 (10)	0.5431 (5)	0.051 (2)	0.60 (4)
H7A	0.7146	−0.0361	0.5216	0.061*	0.60 (4)
H7B	0.8023	0.0356	0.5540	0.061*	0.60 (4)
C8	0.7710 (8)	−0.1140 (18)	0.6072 (3)	0.086 (3)	0.60 (4)

H8A	0.7477	−0.1901	0.5962	0.130*	0.60 (4)
H8B	0.7242	−0.0780	0.6372	0.130*	0.60 (4)
H8C	0.8357	−0.1191	0.6284	0.130*	0.60 (4)
C7'	0.7807 (13)	−0.0185 (17)	0.5315 (9)	0.084 (4)	0.40 (4)
H7'1	0.7175	−0.0168	0.5074	0.101*	0.40 (4)
H7'2	0.8078	0.0592	0.5327	0.101*	0.40 (4)
C8'	0.7635 (12)	−0.063 (2)	0.6031 (7)	0.085 (4)	0.40 (4)
H8'1	0.7223	−0.1306	0.6019	0.127*	0.40 (4)
H8'2	0.7304	−0.0039	0.6289	0.127*	0.40 (4)
H8'3	0.8270	−0.0808	0.6233	0.127*	0.40 (4)
C9	0.82776 (12)	0.16353 (13)	0.37327 (9)	0.0332 (4)	
C10	0.91527 (18)	0.27569 (18)	0.45434 (11)	0.0592 (6)	
H10A	0.8515	0.3163	0.4550	0.071*	0.609 (7)
H10B	0.9227	0.2355	0.4966	0.071*	0.609 (7)
H10C	0.9666	0.2577	0.4872	0.071*	0.391 (7)
H10D	0.8503	0.2768	0.4763	0.071*	0.391 (7)
C11	0.9967 (4)	0.3581 (3)	0.4464 (2)	0.0615 (14)	0.609 (7)
H11A	1.0587	0.3163	0.4385	0.074*	0.609 (7)
H11B	1.0044	0.4012	0.4877	0.074*	0.609 (7)
C12	0.9801 (6)	0.4412 (5)	0.3901 (3)	0.104 (3)	0.609 (7)
H12A	0.9778	0.3997	0.3485	0.157*	0.609 (7)
H12B	1.0342	0.4960	0.3889	0.157*	0.609 (7)
H12C	0.9176	0.4811	0.3967	0.157*	0.609 (7)
C11'	0.9364 (6)	0.3945 (7)	0.4212 (4)	0.078 (3)	0.391 (7)
H11C	0.8867	0.4092	0.3868	0.093*	0.391 (7)
H11D	0.9311	0.4550	0.4546	0.093*	0.391 (7)
C12'	1.0388 (8)	0.3958 (10)	0.3912 (6)	0.120 (5)	0.391 (7)
H12D	1.0881	0.3863	0.4258	0.180*	0.391 (7)
H12E	1.0496	0.4681	0.3688	0.180*	0.391 (7)
H12F	1.0447	0.3339	0.3595	0.180*	0.391 (7)
N1	0.75121 (9)	0.04142 (11)	0.28776 (7)	0.0301 (3)	
H1	0.6968	0.0813	0.2848	0.036*	
N2	0.85239 (9)	−0.10854 (10)	0.28403 (7)	0.0286 (3)	
H2	0.8747	−0.1753	0.2725	0.034*	
N3	1.00441 (9)	0.00930 (11)	0.28903 (7)	0.0313 (3)	
H3	1.0588	−0.0307	0.2914	0.038*	
N4	0.90195 (10)	0.15112 (10)	0.26255 (7)	0.0298 (3)	
H4	0.8784	0.2087	0.2404	0.036*	
O1	0.68919 (9)	−0.12847 (9)	0.24606 (6)	0.0383 (3)	
O2	1.06484 (9)	0.15783 (9)	0.22415 (7)	0.0411 (3)	
O3	1.00591 (10)	−0.11525 (15)	0.40724 (7)	0.0622 (4)	
O4	0.85052 (9)	−0.05045 (11)	0.42944 (6)	0.0417 (3)	
O5	0.74722 (10)	0.19561 (12)	0.39100 (7)	0.0535 (4)	
O6	0.91566 (9)	0.19233 (11)	0.39968 (7)	0.0468 (4)	

Atomic displacement parameters (Å^2)

U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}

C1	0.0263 (8)	0.0219 (7)	0.0322 (8)	−0.0029 (6)	0.0012 (6)	0.0022 (6)
C2	0.0241 (8)	0.0195 (7)	0.0426 (9)	−0.0010 (6)	0.0011 (7)	−0.0021 (6)
C3	0.0197 (7)	0.0196 (6)	0.0361 (8)	0.0011 (5)	−0.0002 (6)	−0.0005 (5)
C4	0.0198 (7)	0.0201 (6)	0.0380 (8)	0.0008 (5)	−0.0004 (6)	−0.0031 (6)
C5	0.0284 (8)	0.0320 (8)	0.0391 (9)	−0.0012 (7)	−0.0036 (7)	0.0003 (6)
C6	0.0544 (12)	0.0682 (13)	0.0332 (10)	−0.0090 (10)	−0.0026 (9)	0.0028 (9)
C7	0.048 (3)	0.075 (4)	0.030 (3)	−0.008 (2)	0.006 (2)	−0.011 (2)
C8	0.103 (5)	0.123 (7)	0.034 (2)	−0.011 (5)	0.009 (2)	0.011 (3)
C7'	0.094 (7)	0.116 (8)	0.043 (5)	−0.031 (6)	0.007 (5)	0.004 (5)
C8'	0.087 (6)	0.110 (9)	0.057 (5)	−0.019 (6)	0.000 (4)	0.000 (5)
C9	0.0315 (9)	0.0251 (7)	0.0430 (9)	0.0028 (6)	0.0002 (7)	−0.0062 (6)
C10	0.0708 (15)	0.0514 (12)	0.0553 (12)	−0.0015 (10)	−0.0096 (11)	−0.0270 (10)
C11	0.079 (3)	0.0429 (19)	0.063 (2)	−0.0114 (19)	−0.031 (2)	−0.0083 (16)
C12	0.144 (7)	0.084 (4)	0.085 (4)	−0.031 (4)	−0.028 (4)	0.036 (3)
C11'	0.088 (5)	0.067 (4)	0.077 (5)	0.007 (4)	−0.025 (4)	−0.025 (4)
C12'	0.139 (8)	0.102 (7)	0.118 (8)	−0.038 (6)	−0.003 (7)	−0.012 (6)
N1	0.0187 (6)	0.0230 (6)	0.0487 (8)	0.0023 (5)	−0.0036 (6)	−0.0057 (5)
N2	0.0277 (7)	0.0174 (6)	0.0408 (7)	0.0018 (5)	−0.0026 (5)	−0.0035 (5)
N3	0.0199 (6)	0.0240 (6)	0.0501 (8)	0.0047 (5)	0.0040 (6)	0.0052 (5)
N4	0.0253 (7)	0.0194 (6)	0.0448 (8)	0.0035 (5)	0.0018 (6)	0.0048 (5)
O1	0.0330 (7)	0.0269 (6)	0.0550 (7)	−0.0087 (5)	−0.0095 (5)	−0.0014 (5)
O2	0.0289 (6)	0.0265 (6)	0.0680 (9)	−0.0009 (5)	0.0115 (6)	0.0067 (5)
O3	0.0410 (8)	0.0915 (12)	0.0542 (8)	0.0207 (8)	−0.0054 (7)	0.0231 (8)
O4	0.0395 (7)	0.0493 (7)	0.0363 (7)	0.0050 (6)	0.0028 (5)	0.0045 (5)
O5	0.0361 (7)	0.0624 (9)	0.0619 (9)	0.0122 (6)	0.0054 (6)	−0.0249 (7)
O6	0.0367 (7)	0.0439 (7)	0.0597 (8)	0.0001 (6)	−0.0070 (6)	−0.0257 (6)

Geometric parameters (Å, °)

C1—O1	1.2291 (18)	C7'—H7'2	0.9700
C1—N1	1.3507 (19)	C8'—H8'1	0.9600
C1—N2	1.3596 (19)	C8'—H8'2	0.9600
C2—O2	1.2268 (18)	C8'—H8'3	0.9600
C2—N4	1.3538 (19)	C9—O5	1.1880 (19)
C2—N3	1.3577 (19)	C9—O6	1.326 (2)
C3—N3	1.4323 (19)	C10—C11	1.452 (4)
C3—N2	1.4494 (18)	C10—O6	1.458 (2)
C3—C5	1.532 (2)	C10—C11'	1.552 (9)
C3—C4	1.5752 (19)	C10—H10A	0.9700
C4—N1	1.4247 (18)	C10—H10B	0.9700
C4—N4	1.4485 (19)	C10—H10C	0.9700
C4—C9	1.538 (2)	C10—H10D	0.9700
C5—O3	1.193 (2)	C11—C12	1.498 (5)
C5—O4	1.320 (2)	C11—H11A	0.9700
C6—O4	1.453 (2)	C11—H11B	0.9700
C6—C7	1.494 (4)	C12—H12A	0.9600
C6—C7'	1.501 (8)	C12—H12B	0.9600
C6—H6A	0.9700	C12—H12C	0.9600
C6—H6B	0.9700	C11'—C12'	1.489 (8)

C6—H6C	0.9700	C11'—H11C	0.9700
C6—H6D	0.9700	C11'—H11D	0.9700
C7—C8	1.535 (7)	C12'—H12D	0.9600
C7—H7A	0.9700	C12'—H12E	0.9600
C7—H7B	0.9700	C12'—H12F	0.9600
C8—H8A	0.9600	N1—H1	0.8600
C8—H8B	0.9600	N2—H2	0.8600
C8—H8C	0.9600	N3—H3	0.8600
C7'—C8'	1.538 (9)	N4—H4	0.8600
C7'—H7'1	0.9700		
O1—C1—N1	125.19 (14)	O5—C9—O6	126.72 (15)
O1—C1—N2	126.36 (14)	O5—C9—C4	123.27 (15)
N1—C1—N2	108.44 (12)	O6—C9—C4	109.88 (13)
O2—C2—N4	125.99 (14)	C11—C10—O6	110.5 (2)
O2—C2—N3	125.61 (14)	C11—C10—C11'	40.2 (3)
N4—C2—N3	108.40 (13)	O6—C10—C11'	105.5 (3)
N3—C3—N2	115.85 (12)	C11—C10—H10A	109.5
N3—C3—C5	110.70 (12)	O6—C10—H10A	109.5
N2—C3—C5	108.84 (12)	C11'—C10—H10A	74.6
N3—C3—C4	103.02 (11)	C11—C10—H10B	109.5
N2—C3—C4	100.78 (11)	O6—C10—H10B	109.5
C5—C3—C4	117.55 (12)	C11'—C10—H10B	141.1
N1—C4—N4	115.26 (13)	H10A—C10—H10B	108.1
N1—C4—C9	110.90 (12)	C11—C10—H10C	72.0
N4—C4—C9	108.70 (11)	O6—C10—H10C	111.2
N1—C4—C3	103.30 (11)	C11'—C10—H10C	110.6
N4—C4—C3	100.92 (11)	H10A—C10—H10C	135.3
C9—C4—C3	117.63 (13)	H10B—C10—H10C	40.1
O3—C5—O4	126.36 (16)	C11—C10—H10D	134.9
O3—C5—C3	122.50 (15)	O6—C10—H10D	110.6
O4—C5—C3	111.07 (13)	C11'—C10—H10D	110.0
O4—C6—C7	113.2 (5)	H10A—C10—H10D	37.6
O4—C6—C7'	100.5 (8)	H10B—C10—H10D	72.7
C7—C6—C7'	13.6 (7)	H10C—C10—H10D	108.9
O4—C6—H6A	108.9	C10—C11—C12	113.3 (4)
C7—C6—H6A	108.9	C10—C11—H10C	38.7
C7'—C6—H6A	110.6	C12—C11—H10C	151.9
O4—C6—H6B	108.9	C10—C11—H11A	108.9
C7—C6—H6B	108.9	C12—C11—H11A	108.9
C7'—C6—H6B	119.6	H10C—C11—H11A	85.8
H6A—C6—H6B	107.7	C10—C11—H11B	108.9
O4—C6—H6C	111.3	C12—C11—H11B	108.9
C7—C6—H6C	108.5	H10C—C11—H11B	88.0
C7'—C6—H6C	110.9	H11A—C11—H11B	107.7
H6A—C6—H6C	2.6	C12'—C11'—C10	110.3 (7)
H6B—C6—H6C	105.6	C12'—C11'—H11C	109.6
O4—C6—H6D	111.6	C10—C11'—H11C	109.6
C7—C6—H6D	102.6	C12'—C11'—H11D	109.6
C7'—C6—H6D	113.2	C10—C11'—H11D	109.6

H6A—C6—H6D	111.5	H11C—C11'—H11D	108.1
H6B—C6—H6D	6.4	C11'—C12'—H12D	109.5
H6C—C6—H6D	109.2	C11'—C12'—H12E	109.5
C6—C7—C8	109.3 (5)	H12D—C12'—H12E	109.5
C6—C7—H7A	109.8	C11'—C12'—H12F	109.5
C8—C7—H7A	109.8	H12D—C12'—H12F	109.5
C6—C7—H7B	109.8	H12E—C12'—H12F	109.5
C8—C7—H7B	109.8	C1—N1—C4	112.70 (12)
H7A—C7—H7B	108.3	C1—N1—H1	123.7
C6—C7'—C8'	108.9 (8)	C4—N1—H1	123.7
C6—C7'—H7'1	109.9	C1—N2—C3	112.75 (12)
C8'—C7'—H7'1	109.9	C1—N2—H2	123.6
C6—C7'—H7'2	109.9	C3—N2—H2	123.6
C8'—C7'—H7'2	109.9	C2—N3—C3	112.49 (12)
H7'1—C7'—H7'2	108.3	C2—N3—H3	123.8
C7'—C8'—H8'1	109.5	C3—N3—H3	123.8
C7'—C8'—H8'2	109.5	C2—N4—C4	112.84 (12)
H8'1—C8'—H8'2	109.5	C2—N4—H4	123.6
C7'—C8'—H8'3	109.5	C4—N4—H4	123.6
H8'1—C8'—H8'3	109.5	C5—O4—C6	117.28 (14)
H8'2—C8'—H8'3	109.5	C9—O6—C10	117.51 (14)
N3—C3—C4—N1	133.40 (12)	O1—C1—N1—C4	−176.74 (14)
N2—C3—C4—N1	13.43 (14)	N2—C1—N1—C4	2.66 (18)
C5—C3—C4—N1	−104.61 (14)	N4—C4—N1—C1	98.59 (15)
N3—C3—C4—N4	13.91 (14)	C9—C4—N1—C1	−137.38 (13)
N2—C3—C4—N4	−106.06 (12)	C3—C4—N1—C1	−10.49 (16)
C5—C3—C4—N4	135.91 (13)	O1—C1—N2—C3	−173.07 (14)
N3—C3—C4—C9	−104.09 (14)	N1—C1—N2—C3	7.54 (17)
N2—C3—C4—C9	135.94 (13)	N3—C3—N2—C1	−123.43 (14)
C5—C3—C4—C9	17.90 (19)	C5—C3—N2—C1	111.10 (14)
N3—C3—C5—O3	−30.6 (2)	C4—C3—N2—C1	−13.12 (16)
N2—C3—C5—O3	97.77 (18)	O2—C2—N3—C3	−179.24 (15)
C4—C3—C5—O3	−148.60 (16)	N4—C2—N3—C3	0.77 (18)
N3—C3—C5—O4	152.17 (13)	N2—C3—N3—C2	99.33 (15)
N2—C3—C5—O4	−79.41 (15)	C5—C3—N3—C2	−136.16 (13)
C4—C3—C5—O4	34.22 (18)	C4—C3—N3—C2	−9.65 (16)
O4—C6—C7—C8	167.4 (6)	O2—C2—N4—C4	−170.19 (15)
C7'—C6—C7—C8	−171 (5)	N3—C2—N4—C4	9.81 (18)
O4—C6—C7'—C8'	173.8 (12)	N1—C4—N4—C2	−125.35 (14)
C7—C6—C7'—C8'	14 (4)	C9—C4—N4—C2	109.47 (14)
N1—C4—C9—O5	−12.1 (2)	C3—C4—N4—C2	−14.86 (16)
N4—C4—C9—O5	115.65 (18)	O3—C5—O4—C6	−2.8 (3)
C3—C4—C9—O5	−130.59 (17)	C3—C5—O4—C6	174.24 (14)
N1—C4—C9—O6	171.75 (13)	C7—C6—O4—C5	160.1 (5)
N4—C4—C9—O6	−60.55 (16)	C7'—C6—O4—C5	155.0 (9)
C3—C4—C9—O6	53.21 (18)	O5—C9—O6—C10	−2.8 (3)
O6—C10—C11—C12	71.3 (6)	C4—C9—O6—C10	173.21 (15)
C11'—C10—C11—C12	−18.6 (5)	C11—C10—O6—C9	−138.0 (2)
C11—C10—C11'—C12'	39.3 (7)	C11'—C10—O6—C9	−96.0 (3)

O6—C10—C11'—C12'	−64.4 (8)

Hydrogen-bond geometry (Å, °)

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
C10—H10B···O3^{i}	0.97	2.56	3.494 (3)	163
N4—H4···O1^{ii}	0.86	2.09	2.8459 (16)	146
N3—H3···O1^{iii}	0.86	2.20	3.0134 (17)	157
N2—H2···O2^{iv}	0.86	2.09	2.9265 (16)	163
N1—H1···O2^{v}	0.86	1.98	2.8320 (17)	174

Symmetry codes: (i) −*x*+2, −*y*, −*z*+1; (ii) −*x*+3/2, *y*+1/2, *z*; (iii) *x*+1/2, *y*, −*z*+1/2; (iv) −*x*+2, *y*−1/2, −*z*+1/2; (v) *x*−1/2, *y*, −*z*+1/2.

Fig. 1

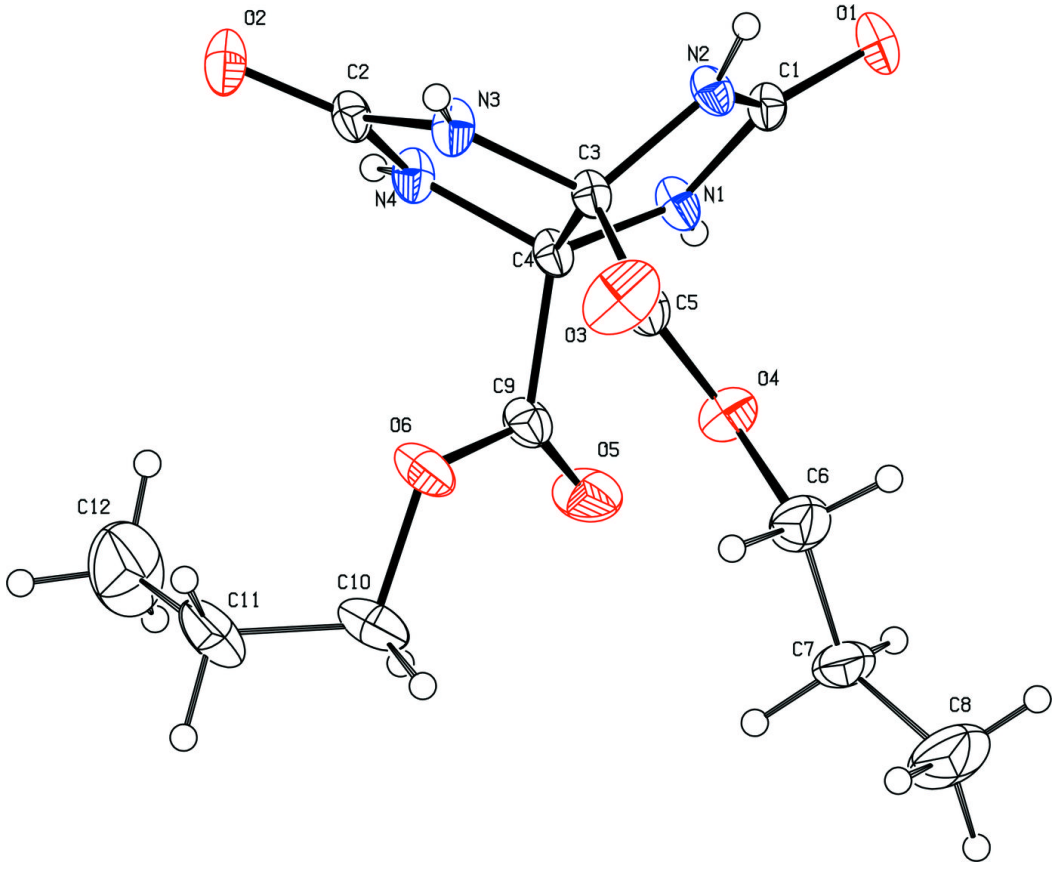


Fig. 2

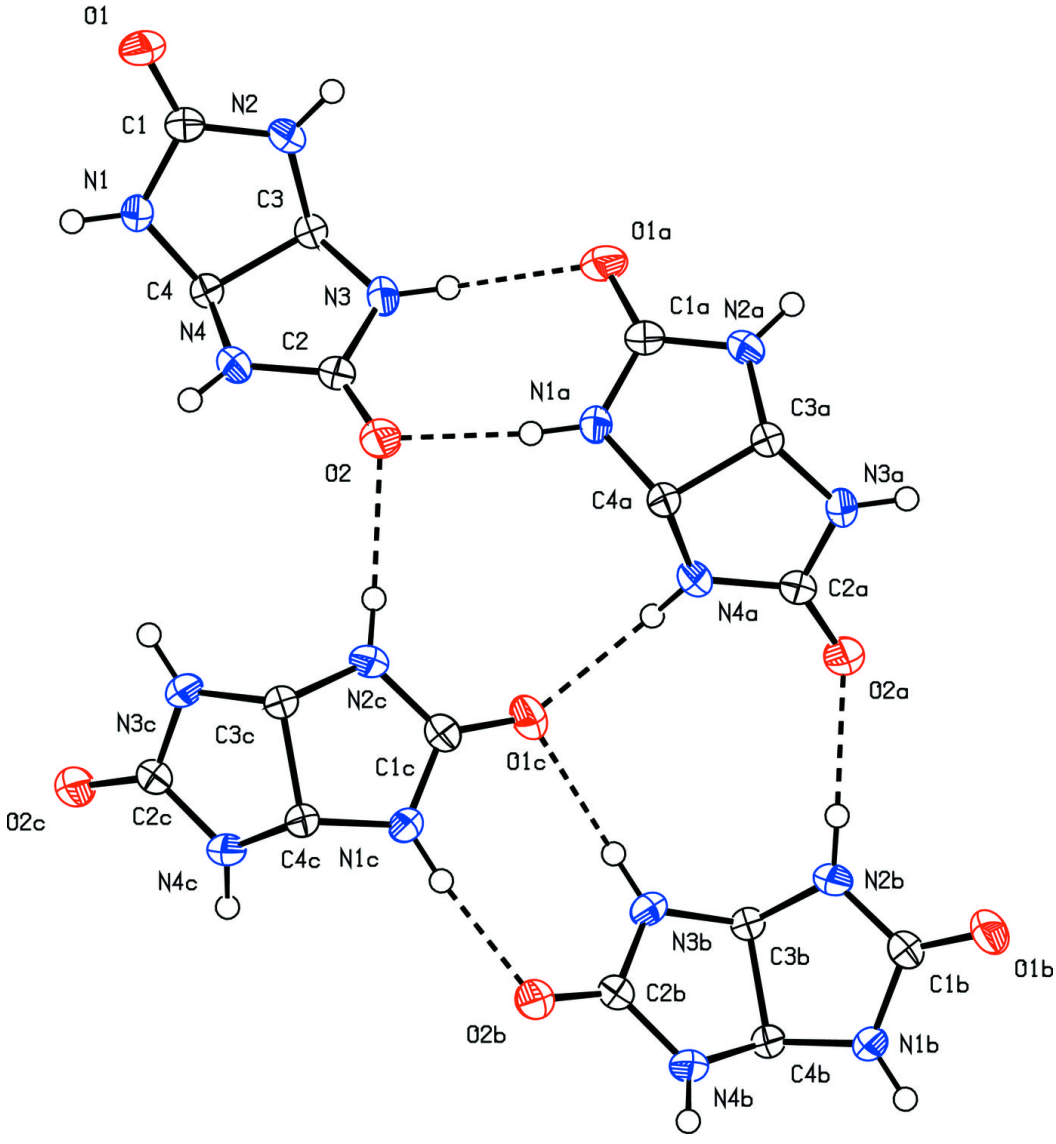
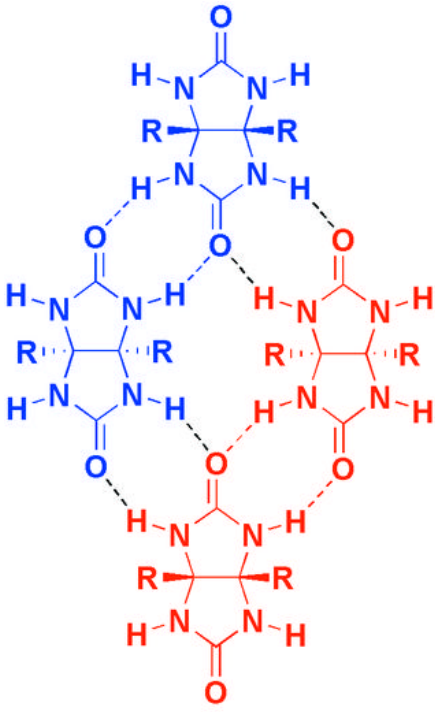
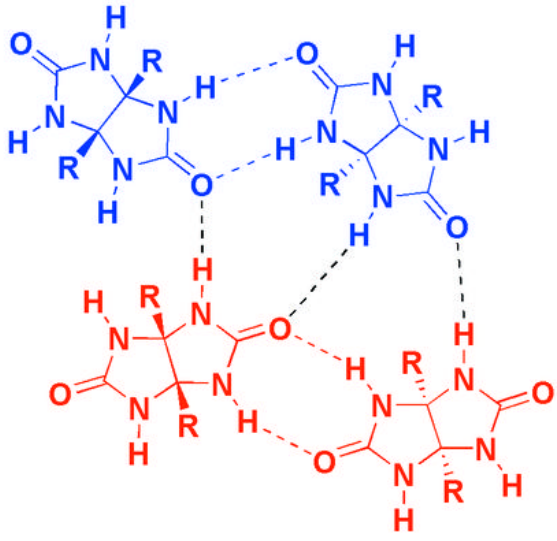

O1
N2
C1
N1
C3
C4
N3
N4
C2
O2
O1a
C1a
N2a
N1a
C3a
N3a
C4a
C2a
N4a
O2a
N2c
N3c
C3c
C1c
O1c
O2c
C2c
C4c
N1c
N4c
N2b
N3b
C3b
C1b
O1b
C2b
O2b
N1b
C4b
N4b

Fig. 3



A

R= -H, -CH_3, -Ph, ...



B

R= -Ph