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5,6-Dichloro-1-propylbenzimidazole

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The title compound, $C_{10}H_{10}Cl_2N_2$, forms centrosymmetric head-to-tail face-to-face dimers with the *n*-propyl groups *exo* to the dichlorobenzimidazole groups, which are effectively planar. Within a dimer, the least-squares planes are necessarily parallel and exhibit an interplanar spacing of 3.412 (2) Å, which lies near the short end of the range (3.3–3.8 Å) generally accepted to indicate π–π interactions. When viewed normal to the (100) planes, the structure appears as slanted columns of dimers arranged so as to form alternating –aromatic–aliphatic– layers parallel to the (001) planes.

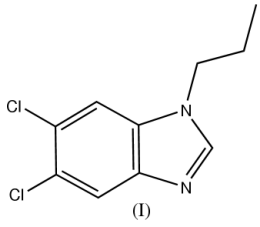
Key indicators

Single-crystal X-ray study
T = 297 K
Mean σ(C–C) = 0.005 Å
R factor = 0.064
wR factor = 0.195
Data-to-parameter ratio = 12.9

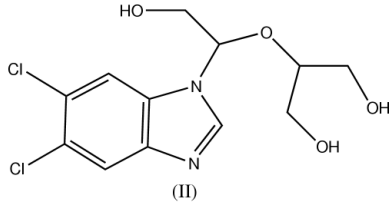
For details of how these key indicators were automatically derived from the article, see http://journals.iucr.org/e.

Comment

Our exploration of the chemistry of bis(benzimidazoles) has led to species which behave as geometrically constraining ligands (Stibrany *et al.*, 2004), ionic salts (Stibrany, Potenza & Schugar, 2002), catalysts (Patil *et al.*, 2003; Stibrany *et al.*, 2003; Stibrany, 2001), and proton sponges (Stibrany, Schugar & Potenza, 2002). Preparation of these species often involves condensation and *N*-alkylation steps to generate a benzimidazole building block, as in the present study, followed by elaboration at the 2-position of the imidazole ring and coupling to yield the appropriate bis(benzimidazole) species. We report here the structure of the building block (I).



(I)



(II)

The structure of (I) contains discrete molecules (Fig. 1) in which the dichlorobenzimidazole fragments are effectively planar. A search of the Cambridge Structural Database

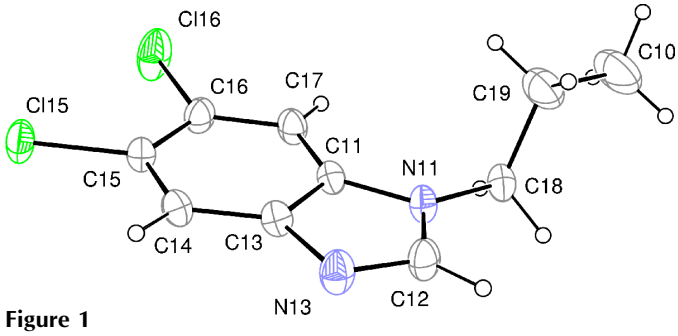


Figure 1
View of the molecule of (I), showing 25% probability displacement ellipsoids and the atom-numbering scheme. H atoms are represented by small spheres of arbitrary radius.

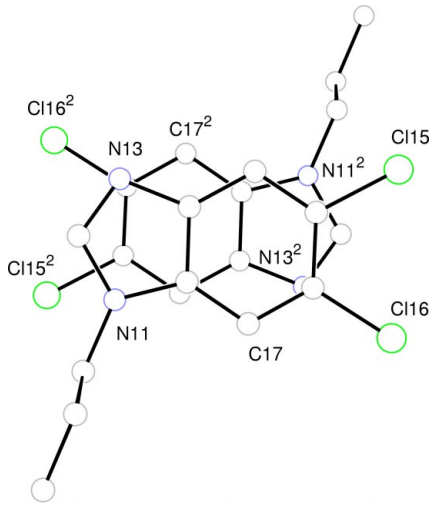


Figure 2
View, approximately normal to the benzimidazole plane, of a dimer of (I). Atom labels without superscripts correspond to symmetry code (x, y, z), while atom labels with the superscript '2' correspond to the symmetry code ($\frac{1}{2}-x$, $\frac{1}{2}-y$, $\frac{1}{2}-z$).

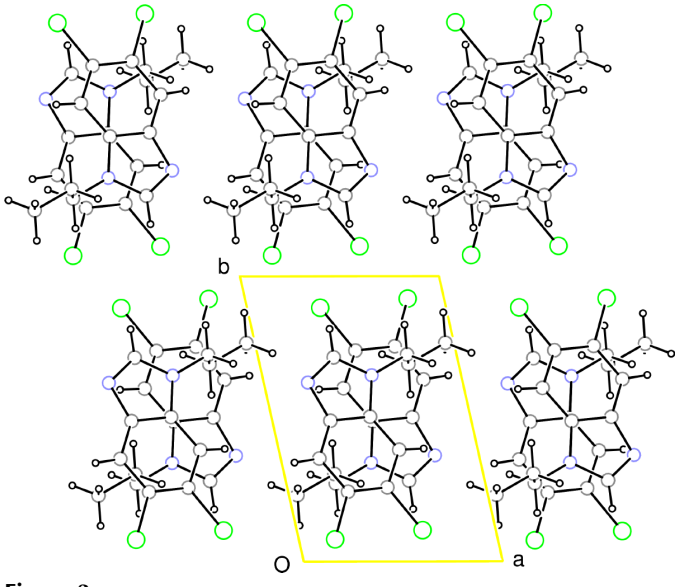


Figure 3
Projection, along the c axis, of the structure of (I) down [001].

(Version 5.24; Allen, 2002) reveals only one entry [(II), refcode CITSUT10; Birnbaum *et al.*, 1985] for a neutral species which contains an *N*-alkylated 5,6-dichlorobenzimidazole unit. Metric parameters for (I) and (II), which contains two independent molecules in the asymmetric unit, compare well (Table 1); in particular, in all three molecules, the C12—N13(imine) distances are considerably shorter than the corresponding C12—N11(amine) distances, as expected.

In the crystal structure, molecules of (I), related by centers of symmetry at ($\frac{1}{2}$, $\frac{1}{2}$, $\frac{1}{2}$), form head-to-tail face-to-face dimers with the *n*-propyl groups *exo* to the dichlorobenzimidazole planes, which are necessarily parallel (Fig. 2). The interplanar spacing of 3.412 (2) Å in the dimers lies near the short end of the range (3.3–3.8 Å; Janiak, 2000) generally accepted to indicate π–π interactions. When viewed along the c axis (Fig. 3), the structure appears as columns of dimers related to each other by translations along the a and b unit-cell directions. In profile (Fig. 4), the columns are seen to be slanted and the structure can be described as consisting of alternating –aromatic–aliphatic– layers parallel to the (001) planes.

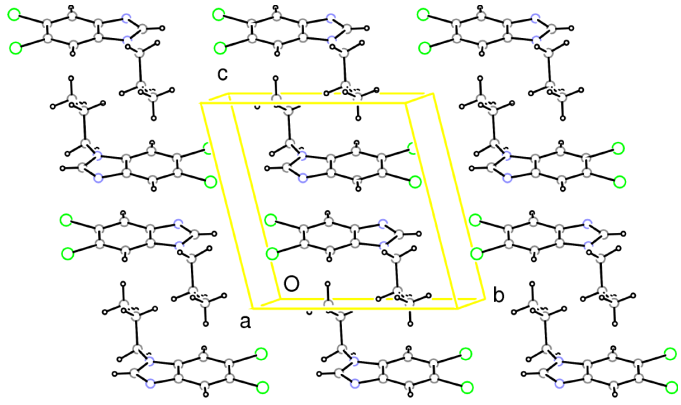


Figure 4
View, normal to (100), of the structure of (I).

Experimental

The precursor 5,6-dichlorobenzimidazole, first reported by Davies *et al.* (1951), was prepared by the Phillips condensation method from 4,5-dichloro-1,2-phenylenediamine and formic acid. The title compound, (I), was prepared by alkylation of this product with *n*-propyl iodide, using a procedure described previously (Stibrany *et al.*, 2004). Alkylation afforded an oil initially, from which crystals of (I) deposited upon standing. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.89 (*s*, 1H), 7.50 (*s*, 1H), 7.27 (*s*, 1H), 4.10 (*t*, J = 7.1 Hz, 2H), 1.91 (sextet, J = 7.3 Hz, 2H), 0.96 (*t*, J = 7.4 Hz, 3H).

Crystal data

$C_{10}H_{10}Cl_2N_2$
M_r = 229.11
Triclinic, $P\overline{1}$
a = 6.2342 (6) Å
b = 9.4516 (8) Å
c = 9.8012 (9) Å
α = 104.207 (7)°
β = 91.138 (8)°
γ = 101.849 (7)°
V = 546.42 (9) Å^3
Z = 2
D_x = 1.392 Mg m^{-3}
Mo $K\alpha$ radiation
Cell parameters from 1011 reflections
θ = 3.4–25.0°
μ = 0.56 mm^{-1}
T = 297 (2) K
Rod, colorless
0.41 × 0.22 × 0.12 mm

Data collection

Bruker SMART APEX CCD area-detector diffractometer
ω scans
Absorption correction: multi-scan (*SADABS*; Blessing, 1995)
T_{min} = 0.663, T_{max} = 0.936
5615 measured reflections
1905 independent reflections
1690 reflections with $I > 2\sigma(I)$
R_{int} = 0.045
θ_{max} = 25.1°
$h = -7 \rightarrow 7$
$k = -11 \rightarrow 11$
$l = -10 \rightarrow 11$

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)]$ = 0.064
$wR(F^2)$ = 0.195
S = 1.00
1905 reflections
148 parameters
All H-atom parameters refined
$w = 1/[\sigma^2(F_o^2) + (0.1415P)^2 + 0.179P]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{max} < 0.001$
$\Delta\rho_{max}$ = 0.60 e Å^{-3}
$\Delta\rho_{min}$ = −0.28 e Å^{-3}

Table 1
Bond lengths (Å) in the 5,6-dichlorobenzimidazole fragments of structures (I) and (II) (molecules *a* and *b*).

	(I)	(II), molecule *a*	(II), molecule *b*
C15—Cl15	1.738 (3)	1.735	1.731
C16—Cl16	1.737 (3)	1.726	1.734
N11—C12	1.363 (4)	1.367	1.356
N13—C12	1.305 (4)	1.308	1.311
N11—C11	1.380 (3)	1.378	1.386
N13—C13	1.391 (4)	1.391	1.393
C11—C13	1.396 (4)	1.405	1.408
C13—C14	1.385 (4)	1.387	1.400
C14—C15	1.375 (4)	1.375	1.371
C15—C16	1.403 (4)	1.405	1.400
C16—C17	1.375 (4)	1.369	1.390
C11—C17	1.381 (4)	1.387	1.386

Note: for molecules *a* and *b* of (II), s.u. values are reported in the range 0.003–0.005 Å.

The coordinates and isotropic displacement parameters of all H atoms were refined.

Data collection: *SMART-WNT*/2000 (Bruker, 2000); cell refinement: *SAINT-Plus* (Bruker, 2000); data reduction: *SAINT-Plus*; program(s) used to solve structure: *SHELXS*97 (Sheldrick, 1990); program(s) used to refine structure: *SHELXL*97 (Sheldrick, 1997); molecular graphics: *ORTEP*III (Burnett & Johnson, 1996) and *ORTEP*-32 (Farrugia, 1997); software used to prepare material for publication: *SHELXTL* (Bruker, 2000).

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