

(1-Phenyl-2,3-dihydro-1*H*-benzimidazol-2-ylidene)malonodinitrile

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Key indicators

Single-crystal X-ray study

$T = 293\text{ K}$

Mean $\sigma(\text{C}-\text{C}) = 0.004\text{ \AA}$

R factor = 0.059

wR factor = 0.130

Data-to-parameter ratio = 12.5

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

The title compound, $\text{C}_{16}\text{H}_{10}\text{N}_4$, has been designed and synthesized for use as a new potential organic molecular electronic material. There are hydrogen bonds in the structure, which are responsible for the formation of linked pairs of molecules. Weak interactions are observed between antiparallel cyano substituents.

Comment

We have designed and synthesized the title compound, (I), as a new potential organic molecular electronic material with high thermal stability. The compound was synthesized by the reaction of *N*-phenyl-*o*-phenylenediamine and 2-cyano-3,3-bis(methylthio)acrylonitrile, which was prepared *in situ* by a modification of the literature method of El-Shafei *et al.* (1995).

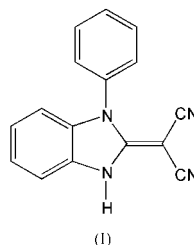
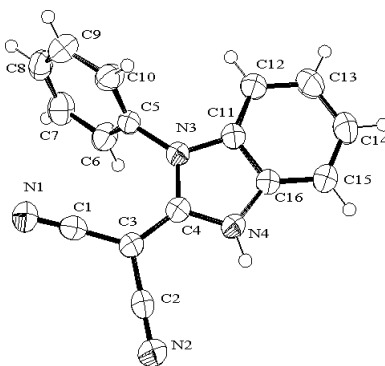


Fig. 1 depicts the structure of (I). As can be seen, the angle $\text{N1}-\text{C1}-\text{C3}$ [$176.3(3)^\circ$] is different from $\text{N2}-\text{C2}-\text{C3}$ [$179.2(3)^\circ$], and $\text{C4}-\text{C3}-\text{C1}$ [$125.5(2)^\circ$] is larger than $\text{C4}-\text{C3}-\text{C2}$ [$117.7(2)^\circ$]. The distance between C1 and the centroid of the C5–C10 phenyl ring is $3.524(4)\text{ \AA}$. The $\text{C1}\cdots\text{C5}$ distance is remarkably shorter [$2.992(4)\text{ \AA}$] than the sum of the van der Waals radii (3.40 \AA); this is indicative of $\pi-\pi$ interaction.

**Figure 1**

The molecular structure of the title compound. Displacement ellipsoids are drawn at the 50% probability level and H atoms are shown as small spheres of arbitrary radii.

Inspection of the packing structure of (I) reveals $N4-H4 \cdots N2^i$ hydrogen bonds (Table 1). These are responsible for the formation of two-membered aggregates (Fig. 2) (Nesterov *et al.*, 2000). In addition, weak intermolecular interactions are also observed between antiparallel cyano substituents (Bock *et al.*, 1996). Such interactions exist either within or between linked pairs of molecules. For example, the antiparallel interaction involving $C2 \equiv N2$ and $N2 \equiv C2^i$ has a $N2 \cdots N2^i$ separation of 3.248 (5) Å and a $C2-N2 \cdots N2^i$ angle of 93.8 (2)° [symmetry code: (i) $x, -y, -1-z$]; on the other hand, the interactions involving $C1 \equiv N1$ and $N1 \equiv C1^{iii}$ [symmetry code: (iii) $1-x, -y, -z$] has a larger $N1 \cdots N1^{iii}$ separation of 3.614 (5) Å and a $C1-N1 \cdots N1^{iii}$ angle of 79.13 (19)°.

Experimental

The title compound was synthesized by the reaction of *N*-phenyl-*o*-phenylenediamine and 2-cyano-3,3-bis(methylthio)acrylonitrile according to the method of El-Shafei *et al.* (1995). Single crystals of (I) were grown by slow evaporation, in air, of an ethanol solution. Selected analytical data: pale yellow solid, yield 79.4%; m.p. 555–557 K; 1H NMR (DMSO- d_6 , 500 MHz): δ 6.80–7.64 (*m*, 9H), 13.03 (*s*, 1H); IR (KBr): ν 3148, 3103, 3001, 2946, 2212, 2187, 1624, 1573, 1481, 1256, 1195, 758, 690 cm^{-1} .

Crystal data

$C_{16}H_{10}N_4$	$D_x = 1.333 \text{ Mg m}^{-3}$
$M_r = 258.28$	Mo $K\alpha$ radiation
Monoclinic, $P2_1/c$	Cell parameters from 950 reflections
$a = 10.377$ (3) Å	$\theta = 2.3\text{--}27.1^\circ$
$b = 17.876$ (5) Å	$\mu = 0.08 \text{ mm}^{-1}$
$c = 7.226$ (2) Å	$T = 293$ (2) K
$\beta = 106.235$ (4)°	Parallelepiped, colourless
$V = 1286.9$ (7) Å ³	$0.15 \times 0.10 \times 0.05 \text{ mm}$
$Z = 4$	

Data collection

Bruker SMART CCD area-detector diffractometer	1556 reflections with $I > 2\sigma(I)$
φ and ω scans	$R_{\text{int}} = 0.051$
Absorption correction: none	$\theta_{\text{max}} = 25.0^\circ$
5318 measured reflections	$h = -11 \rightarrow 12$
2260 independent reflections	$k = -21 \rightarrow 17$
	$l = -8 \rightarrow 8$

Refinement

Refinement on F^2	$w = 1/[\sigma^2(F_o^2) + (0.0503P)^2 + 0.2084P]$
$R[F^2 > 2\sigma(F^2)] = 0.059$	where $P = (F_o^2 + 2F_c^2)/3$
$wR(F^2) = 0.130$	$(\Delta/\sigma)_{\text{max}} < 0.001$
$S = 1.13$	$\Delta\rho_{\text{max}} = 0.16 \text{ e \AA}^{-3}$
2260 reflections	$\Delta\rho_{\text{min}} = -0.18 \text{ e \AA}^{-3}$
181 parameters	
H-atom parameters constrained	

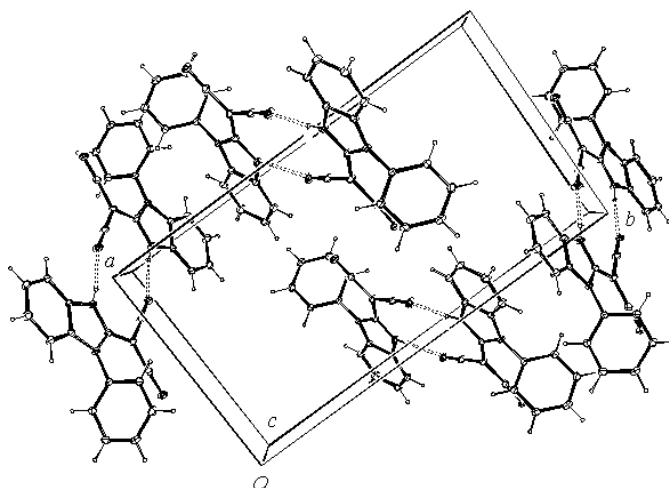


Figure 2

Packing of the molecules in the unit cell, viewed along the *c* axis. Dashed lines indicate N–H···N hydrogen bonds.

Table 1

Hydrogen-bonding geometry (Å, °).

$D-H \cdots A$	$D-H$	$H \cdots A$	$D \cdots A$	$D-H \cdots A$
$N4-H4 \cdots N2^i$	0.86	2.03	2.875 (3)	166
$C9-H9 \cdots N1^{ii}$	0.93	2.58	3.390 (4)	145

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

The H atoms were included using a riding model and were constrained to have C–H = 0.93 and N–H = 0.86 Å and $U_{\text{iso}} = 1.2U_{\text{eq}}$ of their parent atom.

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

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References

- Bock, H., Ziemer, K., Nather, C., Schodel, H., Kleine, M. & Sievert, M. (1996). *Z. Naturforsch. Teil B*, **51**, 1538–1554.
- Bruker (1999). SMART and SAINT. Bruker AXS Inc., Madison, Wisconsin, USA.
- El-Shafei, A. K., Soliman, A. M., Sultan, A. A.-R. & El-Saghier, A. M. M. (1995). *Gazz. Chim. Ital.* **125**, 115–118.
- Nesterov, V. N., Deng, X., Timofeeva, T. V., Antipin, M. Yu., Clark, R. D., Frazier, D. O. & Penn, B. (2000). *J. Mol. Struct.* **523**, 309–318.
- Sheldrick, G. M. (1997a). SHELXS97 and SHELXL97. University of Göttingen, Germany.
- Sheldrick, G. M. (1997b). SHELXTL. Bruker AXS Inc., Madison, Wisconsin, USA.