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4-(2,6-Dichlorobenzylamino)-3,5-diphenyl-4*H*-1,2,4-triazole

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Key indicators

Single-crystal X-ray study
T = 293 K
Mean σ(C–C) = 0.004 Å
R factor = 0.036
wR factor = 0.079
Data-to-parameter ratio = 14.9

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

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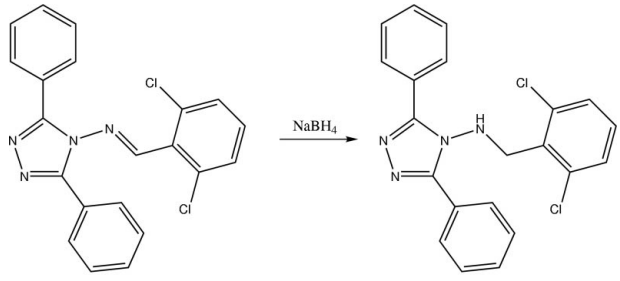
The title compound, $C_{21}H_{16}N_4Cl_2$, was prepared by the reaction of 4-(2,6-dichlorobenzylidenamino)-3,5-diphenyl-4*H*-1,2,4-triazole and $NaBH_4$. The supramolecular structure is defined by N—H···N and C—H···N hydrogen bonds, π–π stacking and C—H···π interactions.

Comment

In recent years, several articles have been devoted to the synthesis and pharmacological investigation of certain compounds (Yüksel *et al.*, 1997; Demirayak *et al.*, 1998; Küçükgüzel *et al.*, 1999; Ikizler *et al.*, 1999; Lonning *et al.*, 1998). We have investigated the possible antimicrobial activity of 4-(2,6-dichlorobenzylamino)-3,5-diphenyl-4*H*-1,2,4-triazole, (I), towards eight standard organisms, including bacterial and fungal strains.



(I)

Compound (I) contains three benzene rings and one 1,2,4-triazole ring. For simplicity, the four rings will be called *A*, *B*, *C* and *D*, corresponding to N1/N2/C3/N4/C5, C6–C11, C12–C17 and C19–C24, respectively. The molecular structure is shown in Fig.1, and selected geometric parameters and the hydrogen-bond geometry are reported in Tables 1 and 2, respectively.

The dihedral angles between the planes of the rings are *A*/*B* = 41.46 (7)°, *A*/*C* = 31.15 (8)°, *A*/*D* = 38.56 (7)°, *B*/*C* = 20.56 (9)°, *B*/*D* = 27.93 (8)° and *C*/*D* = 9.77 (9)°. In the triazole ring, the maximum deviation from planarity is 0.0095 (2) Å for atom C3.

The N═C, N—N and Cl—C bond lengths [N4═C3 = 1.369 (3) Å, N4═C5 = 1.376 (3) Å, N4—N3 = 1.410 (2) Å, N1—N2 = 1.393 (3) Å, Cl1═C24 = 1.730 (2) Å and Cl2═C20 = 1.739 (2) Å] agree with literature values (Palmer & Parsons, 1996; Ocak, Çoruh *et al.*, 2003; Ocak, Kahvecib *et al.*, 2003; McCarrick *et al.*, 1999; Atalay *et al.*, 2003, 2004).

The crystal structure is stabilized by N—H···N and C—H···N intra- and intermolecular hydrogen bonds (Fig. 2), π–π stacking and C—H···π interactions.

The crystal structure contains three intermolecular C—H···π interactions. In the first of these interactions, phenyl

atom C7 acts as a donor and interacts with the symmetry-related benzene ring *B* of the molecule at $(\frac{1}{2}-x, \frac{1}{2}+y, z)$ [C7···*Cg*4 = 3.669 (3) Å, H1···*Cg*4 = 3.17 Å and C7—H1···*Cg*4 = 114°; *Cg*4 is the centroid of ring *B* at $(\frac{1}{2}-x, \frac{1}{2}+y, z)$]. In the second interaction, atom C15 interacts with the symmetry-related benzene ring *C* of the molecule at $(\frac{1}{2}+x, \frac{1}{2}-y, -z)$ [C15···*Cg*2 = 3.987 (4) Å, H12···*Cg*2 = 3.17 Å and C15—H12···*Cg*2 = 141°; *Cg*2 is the centroid of ring *C* at $(\frac{1}{2}+x, \frac{1}{2}-y, -z)$]. In the third of the interactions, atom C21 interacts with the symmetry-related benzene ring *D* of the molecule at $(\frac{1}{2}-x, -\frac{1}{2}+y, z)$ [C18···*Cg*1 = 3.601 (3) Å, H18···*Cg*1 = 2.99 Å and C21—H18···*Cg*1 = 124°; *Cg*1 is the centroid of ring *D* at $(\frac{1}{2}-x, -\frac{1}{2}+y, z)$].

In the structure of (I), the π-π stacking interactions involve rings *C* and *D*. Ring *C* in the molecule at (*x*, *y*, *z*) stacks with ring *D* in the same molecule, with a distance of 3.81 (3) Å between the ring centroids (Fig. 1).

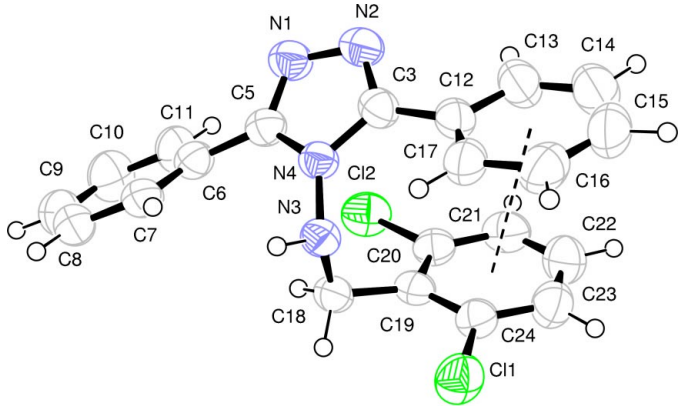


Figure 1
The molecular structure of (I), showing the atomic numbering scheme. Displacement ellipsoids are drawn at the 50% probability level for all non-H atoms. The dashed line indicates π–π stacking.

Experimental

4-(2,6-Dichlorobenzylideneamino)-3,5-diphenyl-4*H*-1,2,4-triazole (0.005 mol) was dissolved in dried methanol (50 ml), and $NaBH_4$ (0.005 mol) was added in small portions to this solution. The mixture was refluxed for 20 min and then allowed to cool. After evaporation at 298–300 K under reduced pressure, the solid residue was washed with cold water. After drying *in vacuo*, the solid product was recrystallized from ethyl acetate (yield 92%, m.p. 475–476 K). IR (KBr, cm^{-1}): 3248, 1592, 771, 768, 717, 692. ^{1}H NMR ($CDCl_3$, p.p.m.): 4.08 (*d*, 2H, CH_2), 6.10 (*t*, 1H, NH), 6.90 (*m*, 3H, Ar-H), 7.40 (*m*, 6H, Ar-H), 7.80 (*m*, 4H, Ar-H). ^{13}H NMR ($CDCl_3$): 153.61 (2C), 136.04 (2C), 130.36, 129.86, 129.70 (2C), 128.55 (4C), 127.96 (4C), 127.88 (2C), 126.28 (2C), 50.85. UV [ν_{max}, nm ($\varepsilon \times 10^{-3}$)]: 259 (21.7), 208 (31.3) Analysis calculated for $C_{21}H_{16}Cl_2N_4$: C 63.81, H 4.08, N 14.17%; found: C 63.93, H 4.01, N 14.33%.

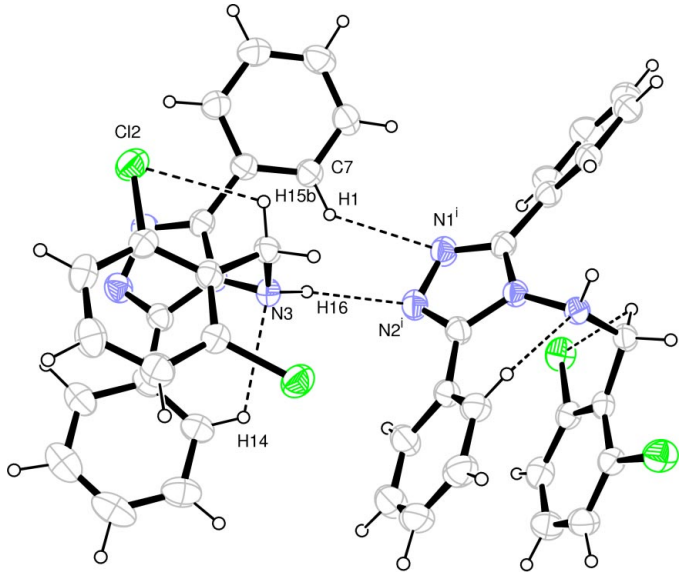


Figure 2
The hydrogen-bonding network (dashed lines) in the title compound. [Symmetry code: (i) $x, \frac{1}{2}-y, \frac{1}{2}+z$.]

Crystal data

$C_{21}H_{16}Cl_2N_4$
M_r = 395.28
Orthorhombic, *Pbca*
a = 23.5075 (13) Å
b = 13.9199 (10) Å
c = 11.531 (8) Å
V = 3773 (3) Å^3
Z = 8
D_x = 1.392 Mg m^{-3}
Mo *K*α radiation
Cell parameters from 14 312 reflections
θ = 1.7–25.7°
μ = 0.36 mm^{-1}
T = 293 (2) K
Plate, colorless
0.42 × 0.25 × 0.10 mm

Data collection

Stoe IPDS-II diffractometer
ω scans
Absorption correction: integration (*X-RED*32; Stoe & Cie 2002)
T_{min} = 0.882, T_{max} = 0.968
36 036 measured reflections
3705 independent reflections
1822 reflections with $I > 2\sigma(I)$
R_{int} = 0.096
θ_{max} = 26.0°
h = −28 → 28
k = −17 → 17
l = −13 → 14

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)]$ = 0.036
$wR(F^2)$ = 0.079
S = 0.75
3705 reflections
249 parameters
H atoms treated by a mixture of independent and constrained refinement
$w = 1/[\sigma^2(F_o^2) + (0.0403P)^2]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{max}$ = 0.001
$\Delta\rho_{max}$ = 0.14 e Å^{-3}
$\Delta\rho_{min}$ = −0.20 e Å^{-3}
Extinction correction: *SHELXL*97
Extinction coefficient: 0.00149 (16)

Table 1
Selected bond lengths (Å).

Cl1—C24	1.730 (2)	C3—N2	1.313 (3)
N3—N4	1.410 (2)	C5—N1	1.309 (3)
N3—C18	1.465 (3)	N2—N1	1.393 (3)
N4—C3	1.369 (3)	C20—C21	1.385 (3)

Table 2
Hydrogen-bond geometry (Å, °).

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
N3—H3···N2^i	0.89 (2)	2.30 (2)	3.148 (3)	159.1 (19)
C7—H7···N1^i	0.93	2.54	3.241 (3)	132
C17—H17···N3	0.93	2.59	3.068 (3)	113
C18—H18*A*···Cl2	0.97	2.59	3.089 (2)	112

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

All C-bound H atoms were placed in calculated positions and refined using a riding model, with C—H distances of 0.93 (aromatic) and 0.97 Å (CH_2), an N—H distance of 0.89 Å and with $U_{iso}(H) = 1.2U_{eq}(C,N)$.

Data collection: *X-AREA* (Stoe & Cie, 2002); cell refinement: *X-AREA*; data reduction: *X-RED*32 (Stoe & Cie, 2002); program(s) used to solve structure: *SHELXS*97 (Sheldrick, 1997); program(s) used to refine structure: *SHELXL*97 (Sheldrick, 1997); molecular graphics: *ORTEP*III (Burnett & Johnson, 1996); software used to prepare material for publication: *WinGX* (Farrugia, 1999) and *PARST* (Nardelli, 1995).

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