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N,*N*′-Bis[(4-ethoxyphenyl)aminoacetyl]-4,5-dimethyl-*o*-phenylenediamine

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Key indicators

Single-crystal X-ray study
T = 293 K
Mean σ(C–C) = 0.004 Å
R factor = 0.048
wR factor = 0.175
Data-to-parameter ratio = 17.0

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

All interatomic distances in the title compound, $C_{28}H_{34}N_4O_4$, are normal. The (4-ethoxyphenyl)amino parts of the side branches are almost ideally planar. The overall arrangement of the side branches seems to be imposed by hydrogen bonds. The substituents are stabilized by interbranch N—H···O hydrogen bonds. The structure of the title compound is assembled by intermolecular N—H···O hydrogen bonds, forming a two-dimensional framework.

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Comment

The present work is a continuation of our previous studies on derivatives of 2,3,4,5,6,7-hexahydro-1*H*-1,4,7-benzotriazone-2,5-dione. Upon introduction of pharmacophoric substituents for a desired activity into those systems, it is expected that derivatives will be obtained exhibiting numerous favourable properties, such as analeptic activity or possible anticancer and anti-HIV activities (Bartczak *et al.*, 1995). Within this area of research, preliminary results of the crystal structure of the title compound have been previously published [Mikiciuk–Olasik *et al.*, 1993; Cambridge Structural Database (CSD; Allen and Kennard, 1993) refcode: WEWPIX], but the complete structure was never determined. Thus, we now present the full structure determination of *N*,*N*′-bis-[(4-ethoxyphenyl)aminoacetyl]-4,5-dimethyl-*o*-phenylenediamine.

A perspective view of the title compound, (III), together with the atom-numbering scheme is shown in Fig. 1. All interatomic distances can be considered normal. Atoms C7, C8 and C30 show signs of disorder (particularly high values of $U_{\rm eq}$), but all attempts to model the disorder failed. It can be suggested that the disorder is dynamical in character. All atoms of the (4-ethoxyphenyl)amino parts of the respective side branches are almost coplanar. The maximum deviation in the N11 branch is 0.058 (2) Å for O12 (branch *A* hereafter) and the maximum deviation in the N21 branch is 0.046 (2) Å for O22 (branch *B* hereafter). The next C atom of each branch, C*x*2, deviates by 0.740 (2) and 0.425 (2) Å from the above planes, respectively (where $x = 1$ for the atoms of the branch *A* and $x = 2$ for the atoms of the branch *B*). These planes make a dihedral angle of 27.37 (5)°. The main difference between branches *A* and *B* is the conformation of the chain consisting of N*x*1, C*x*1, C*x*2 and N*x*2 (best described by the torsion angles, see Table 1 for details). The overall arrangement of the side branches seems to be imposed partly by the N12—H12···O11 hydrogen bond and the C5—H5···O21 weak hydrogen bond (Desiraju & Steiner, 1999) (branch *A*), and by the N21—H21···N22 hydrogen bond (branch *B*); for details, see Table 2. The substituents are stabilized by an interbranch N21—H21···O11 hydrogen bond (Jeffrey & Saenger, 1994).

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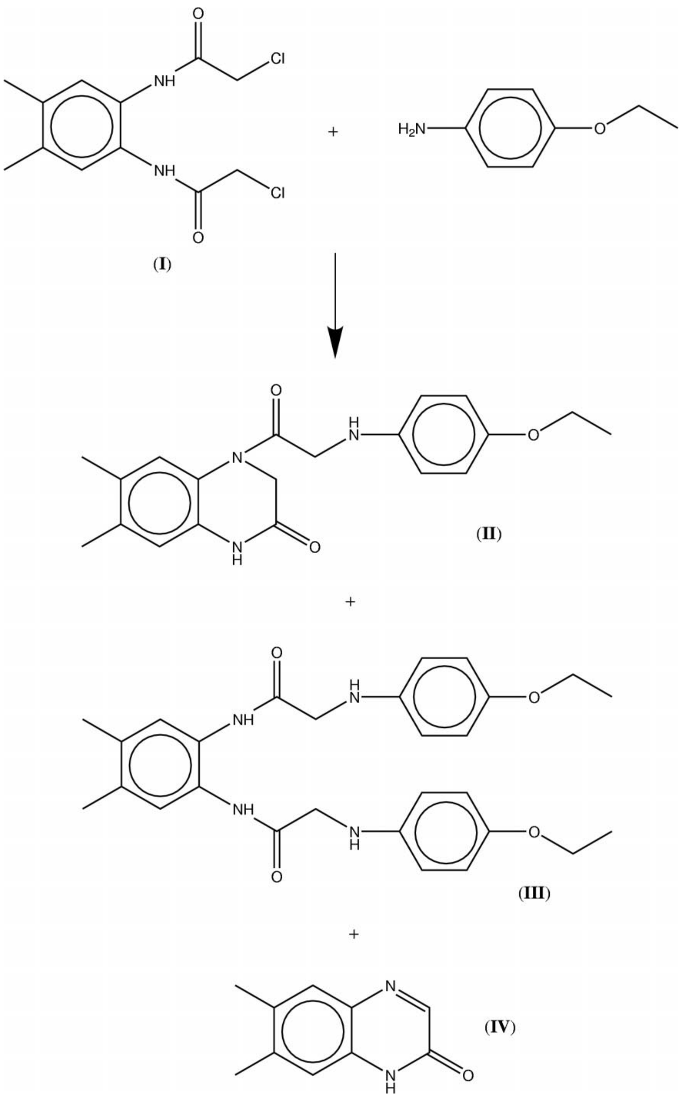
The N21 atom acts as a donor for two intramolecular hydrogen bonds and these hydrogen bonds are created *via* the same H atom (H21). The structure of the title compound is assembled by intermolecular N—H···O hydrogen bonds, to form a two-dimensional framework (Fig. 2 and Table 2). There are no unusual intermolecular short contacts, except for the hydrogen bonds described in Table 2.



Experimental

Compound (III) was prepared according to the method depicted in the above Scheme. A mixture 0.015 mol (4.00 g) of (I) and 0.030 mol (4.20 g) of *p*-phenetidine in 600 ml of anhydrous ethanol in the presence of anhydrous sodium carbonate (0.075 mol, 8.00 g) was heated under reflux for 10 h (Mikiciuk-Olasik *et al.*, 1993, 1994). The mixture was filtered and the solvent was distilled off. Compounds (II), (III) and a small amount of (IV) were isolated by fractional crystallization of the solid residue. Data for (III): yield 20%; m.p. 454.2 K. Elemental analysis (calculated/found): C 68.55/68.53%, H 6.98/7.09%, N 11.42/11.24%. IR (cm^{-1}): 3200, 1590 (NH); 1670, 1520 (NH–CO). ^{1}H NMR (in DMSO/TMS, chemical shifts in p.p.m.): 1.4 (*t*, 6H, $2CH_3CH_2$, *J* = 7 Hz), 2.35 (*s*, 6H, $2CH_3Ph$), 3.7 (*d*, 4H, $2CH_2NH$, *J* = 6 Hz), 4.05 (*q*, 4H, $2CH_2CH_3$, *J* = 7 Hz), 5.65 (*t*, 2H, $2NHCH_2$, *J* = 6 Hz) 6.9 (*m*, $10H_{ar}$), 9.6 (*s*, 2H, NHCO).

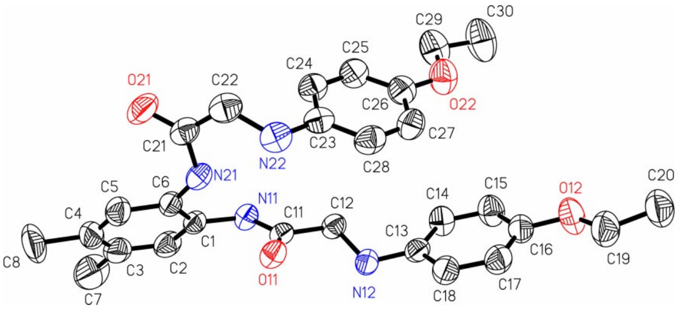


Figure 1
The molecular structure of title compound (III). Displacement ellipsoids are drawn at the 50% probability level. H atoms have been omitted for clarity.

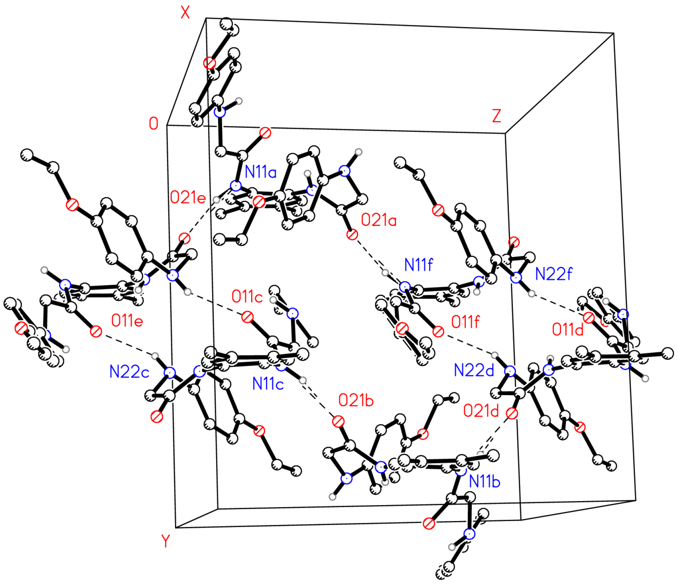


Figure 2
Molecular packing of (III) showing intermolecular hydrogen bonds creating a two-dimensional net structure. Carbon-bonded H atoms have been omitted for clarity. Hydrogen bonds are indicated by dashed lines.

Crystal data

$C_{28}H_{34}N_4O_4$
$M_r = 490.59$
Monoclinic, $P2_1/c$
$a = 14.6542$ (11) Å
$b = 14.3344$ (14) Å
$c = 12.7220$ (12) Å
$\beta = 95.606$ (9)°
$V = 2659.6$ (4) Å^3
$Z = 4$
$D_x = 1.225$ Mg m^{-3}
Cu *Kα* radiation
Cell parameters from 99 reflections
$\theta = 5–60°$
$\mu = 0.67$ mm^{-1}
$T = 293$ (2) K
Sphere, colourless
0.46 mm (radius)

Data collection

Kuma KM-4 diffractometer
$\omega–2\theta$ scans
Absorption correction: numerical *X-RED* (Stoe & Cie, 1999)
$T_{\min} = 0.713$, $T_{\max} = 0.789$
6127 measured reflections
5857 independent reflections
3352 reflections with $I > 2\sigma(I)$
$R_{\rm int} = 0.040$
$\theta_{\max} = 80.8°$
$h = -18 \rightarrow 18$
$k = -18 \rightarrow 0$
$l = -16 \rightarrow 0$
2 standard reflections every 100 reflections intensity decay: −3.1%

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)] = 0.048$
$wR(F^2) = 0.175$
$S = 1.05$
5857 reflections
345 parameters
H atoms treated by a mixture of independent and constrained refinement
$w = 1/[\sigma^2(F_o^2) + (0.1016P)^2]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{\max} = 0.001$
$\Delta\rho_{\max} = 0.35$ e Å^{-3}
$\Delta\rho_{\min} = -0.42$ e Å^{-3}

Table 1
Selected torsion angles (°).

C6—C1—N11—C11	57.6 (3)	N21—C21—C22—N22	5.8 (3)
C1—N11—C11—C12	177.75 (18)	C21—C22—N22—C23	98.6 (3)
N11—C11—C12—N12	−158.31 (18)	C22—N22—C23—C24	−18.6 (3)
C11—C12—N12—C13	−149.63 (19)	C15—C16—O12—C19	172.0 (3)
C12—N12—C13—C14	−34.7 (3)	C16—O12—C19—C20	−176.3 (3)
C1—C6—N21—C21	136.3 (2)	C25—C26—O22—C29	−6.2 (4)
C6—N21—C21—C22	−179.5 (2)	C26—O22—C29—C30	−179.3 (3)

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

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
N11—H11···O21^{i}	0.91 (3)	1.89 (3)	2.783 (2)	166 (2)
N12—H12···O11	0.91 (3)	2.13 (3)	2.675 (2)	118 (2)
N22—H22···O11^{ii}	0.85 (3)	2.27 (3)	3.025 (2)	148 (3)
N21—H21···N22	0.92 (3)	2.12 (3)	2.674 (3)	118 (2)
N21—H21···O11	0.92 (3)	2.15 (3)	2.819 (2)	129 (2)
C5—H5···O21	0.93	2.60	2.974 (3)	105

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

Because the diffraction data were relatively weak, there is a large number of reflections with small intensities; these were flagged as unobserved. This affects the fraction of unique observed reflections (out to $\theta = 79°$) which is equal to 96%.

All H atoms, except those bonded to N atoms, were placed at calculated positions. The H atoms bonded to N atoms were located in difference Fourier syntheses, calculated after four cycles of anistropic refinement. All H atoms were treated as riding on the parent C atom. The methyl groups were allowed to rotate about their local threefold axis.

Data collection: *KM*-4 *Software* (Kuma, 1993); cell refinement: *KM*-4 *Software*; data reduction: *DATAPROC* (Gałdecki *et al.*, 1998); program(s) used to solve structure: *SHELXS*97 (Sheldrick, 1990*a*); program(s) used to refine structure: *SHELXL*97 (Sheldrick, 1997); molecular graphics: *XP* in *SHELXTL*/*PC* (Sheldrick, 1990*b*) and *ORTEP*-3 (Farrugia, 1997).

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