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Key indicators

Single-crystal X-ray study
 $T = 296$ K
Mean $\sigma(\text{C}-\text{C}) = 0.004$ Å
 R factor = 0.049
 wR factor = 0.125
Data-to-parameter ratio = 15.0For details of how these key indicators were
automatically derived from the article, see
<http://journals.iucr.org/e>.

2-(1-Phenylethylideneamino)guanidine

The title compound, $\text{C}_9\text{H}_{12}\text{N}_4$, is planar. The crystal packing is
mainly stabilized by intramolecular and intermolecular $\text{N}-\text{H}\cdots\text{N}$
and $\text{C}-\text{H}\cdots\text{N}$ hydrogen-bonding interactions.Received 28 July 2006
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Comment

Guanylhyazones (alkylenaminoguanidines, carboximide-amidehydrazones, diaminomethylene hydrazones) are condensation products of oxo compounds with aminoguanidines. This class of compounds has been known for a long time (Thiele, 1892) and is of considerable interest due to a wide variety of different pharmacological activities found for many representatives (Richter *et al.*, 1993; Schleuder *et al.*, 1993). Further, guanylhyazones are valuable synthetic building blocks for ring closure reactions leading to several nitrogen-containing heterocycles (Sarıpinar *et al.*, 2005). Protomeric tautomerism is of much interest in experimental as well as theoretical chemistry, since it is an important reaction in biological processes. Guanylhyazones can exist in two tautomeric forms (Zoltan *et al.*, 1999). They may undergo proton shifts (tautomerism) rapidly and easily, and the chemical reactivity of the two isomers may be quite different. In this study, the compound 2-(1-phenylethylideneamino)guanidine, (I), is in the *E* configuration at the $\text{C}=\text{N}$ double bond.

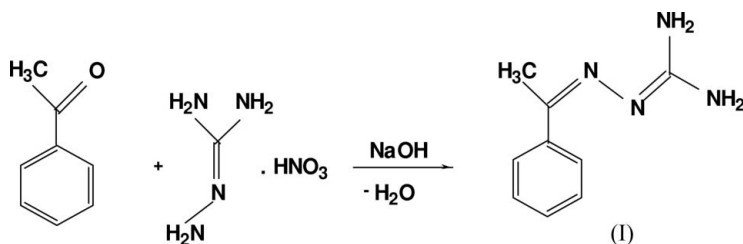


Fig. 1 shows the molecular structure and conformation of (I), with the atom-labelling scheme. The $\text{C}-\text{N}$ bond distances involving the two NH_2 groups, *viz.* $\text{C}1-\text{N}4$ and $\text{C}1-\text{N}3$, as well as the $\text{N}4-\text{C}1-\text{N}3$ angle (Table 1), indicate considerable π -character in the bonding.

In the aminoguanidine group, the $\text{N}-\text{N}$ bond length is $1.401(2)$ Å, approximating to a pure single bond (1.41 Å; Burke-Laing & Laing, 1976). That the lone-pair electrons on atom $\text{N}2$ are delocalized through conjugation with the amidine group, rather than the $\text{N}1=\text{C}2$ double bond, is also seen in the $\text{N}2-\text{C}1$ bond length of $1.317(2)$ Å, which is intermediate between a single and a double bond, and similar to the two $\text{C}-\text{N}$ bonds in the amidine group. The $\text{C}9-\text{C}4-\text{C}5$ angle is $117.8(2)^\circ$, while the rest of the internal angles are almost

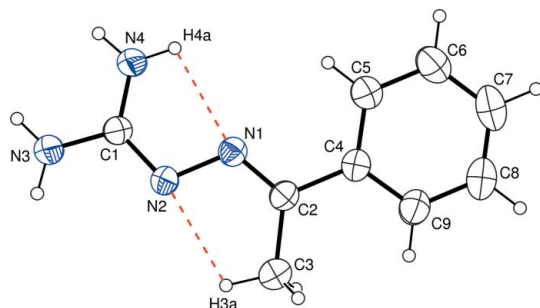


Figure 1

A view of the title compound, (I), showing the atomic numbering scheme. Displacement ellipsoids of non-H atoms are drawn at the 40% probability level and H atoms are shown as small spheres of arbitrary radius. Dashed lines indicate hydrogen bonds.

equal to the normal value of 120° for the intra-ring bond angles of benzene rings due to the steric effect of the methyl group bonded to atom C2.

An intramolecular N4—H4A...N1 hydrogen bond leads to the formation of a five-membered ring in the aminoguanidine group (Fig. 1). The crystal structure of (I) is stabilized by a network of N—H...N hydrogen bonds (Table 2).

Experimental

Acetophenone (2.4 g) and aminoguanidine nitrate (2.74 g) were stirred in water (20 ml) at room temperature for approximately 12 h. NaOH (3 g) was added to neutralize the mixture, which was cooled to 273 K and kept at that temperature overnight. Precipitated (I) was filtered and recrystallized from water and allowed to dry on P_2O_5 (yield 2.30 g, 75%; m.p. 464 K). Analysis calculated for $C_9H_{12}N_4$: C 61.17, H 6.82, N 31.82%; found: C 61.20, H 6.91, N 31.75%.

Crystal data

$C_9H_{12}N_4$	$Z = 8$
$M_r = 176.23$	$D_x = 1.182 \text{ Mg m}^{-3}$
Orthorhombic, <i>Pbca</i>	Mo $K\alpha$ radiation
$a = 8.2673 (9) \text{ \AA}$	$\mu = 0.08 \text{ mm}^{-1}$
$b = 9.0198 (7) \text{ \AA}$	$T = 296 \text{ K}$
$c = 26.565 (2) \text{ \AA}$	Needle, colorless
$V = 1981.0 (3) \text{ \AA}^3$	$0.76 \times 0.35 \times 0.11 \text{ mm}$

Data collection

Stoe IPDS-2 diffractometer	8590 measured reflections
ω scans	1741 independent reflections
Absorption correction: integration	1085 reflections with $I > 2\sigma(I)$
(<i>X-RED32</i> ; Stoe & Cie, 2002)	$R_{\text{int}} = 0.081$
$T_{\text{min}} = 0.960$, $T_{\text{max}} = 0.992$	$\theta_{\text{max}} = 25.0^\circ$

Refinement

Refinement on F^2	$w = 1/[\sigma^2(F_o^2) + (0.0617P)^2]$
$R[F^2 > 2\sigma(F^2)] = 0.049$	where $P = (F_o^2 + 2F_c^2)/3$
$wR(F^2) = 0.125$	$(\Delta/\sigma)_{\text{max}} < 0.001$
$S = 1.03$	$\Delta\rho_{\text{max}} = 0.15 \text{ e \AA}^{-3}$
1741 reflections	$\Delta\rho_{\text{min}} = -0.12 \text{ e \AA}^{-3}$
116 parameters	Extinction correction: <i>SHELXL97</i>
H atoms treated by a mixture of independent and constrained refinement	Extinction coefficient: 0.018 (2)

Table 1

Selected geometric parameters (\AA , $^\circ$).

C1—N2	1.317 (3)	C2—N1	1.286 (2)
C1—N4	1.335 (3)	N1—N2	1.401 (2)
C1—N3	1.354 (3)		
N4—C1—N3	117.1 (2)	C2—N1—N2	116.41 (17)
N1—C2—C4	116.4 (2)		

Table 2

Hydrogen-bond geometry (\AA , $^\circ$).

<i>D</i> —H... <i>A</i>	<i>D</i> —H	H... <i>A</i>	<i>D</i> ... <i>A</i>	<i>D</i> —H... <i>A</i>
N3—H33 <i>A</i> ...N4 ⁱ	0.89 (3)	2.41 (3)	3.227 (3)	153 (2)
N3—H33 <i>B</i> ...N1 ⁱⁱ	0.90 (3)	2.40 (3)	3.277 (3)	165 (3)
N4—H4 <i>A</i> ...N2 ⁱⁱⁱ	0.92 (3)	2.49 (3)	3.197 (3)	134 (2)
N4—H4 <i>B</i> ...N2 ⁱⁱ	0.87 (3)	2.16 (3)	3.026 (3)	173 (2)
N4—H4 <i>A</i> ...N1	0.92 (3)	2.25 (3)	2.585 (3)	100.6 (19)
C3—H3 <i>A</i> ...N2	0.96	2.31	2.751 (3)	107

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

Methyl H atoms were refined in riding mode, with C—H = 0.96 \AA and $U_{\text{iso}}(\text{H}) = 1.5U_{\text{eq}}(\text{C})$. All other H atoms were refined freely; C—H = 0.93 (4)–0.98 (3) and N—H = 0.87 (3)–0.92 (3) \AA . The displacement parameters of the H atoms were constrained as $U_{\text{iso}}(\text{H}) = 1.2U_{\text{eq}}$ of the carrier atom.

Data collection: *X-AREA* (Stoe & Cie, 2002); cell refinement: *X-AREA*; data reduction: *X-RED32* (Stoe & Cie, 2002); program(s) used to solve structure: *SHELXS97* (Sheldrick, 1997); program(s) used to refine structure: *SHELXL97* (Sheldrick, 1997); molecular graphics: *ORTEP-3 for Windows* (Farrugia, 1997); software used to prepare material for publication: *WinGX* (Farrugia, 1999) and *PLATON* (Spek, 2003).

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