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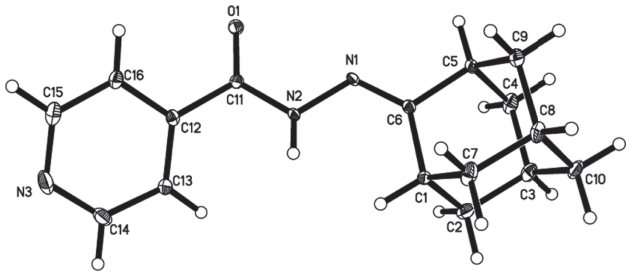
Crystal structure of N′-(adamantan-2-ylidene)-isonicotinohydrazide, $C_{16}H_{19}N_3O$



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Abstract

$C_{16}H_{19}N_3O$, orthorhombic, $Pna2_1$ (no. 33), $a = 8.0251(7)$ Å, $b = 10.1627(9)$ Å, $c = 17.0686(16)$ Å, $V = 1392.1(2)$ Å^3, $Z = 4$, $R_{gt}(F) = 0.064$, $wR(F^2) = 0.109$, $T = 100$ K.

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The crystal structure is shown in the figure. Tables 1–3 contain details of the measurement method and a list of the atoms including atomic coordinates and displacement parameters.

Source of material

Pyridine-4-carbohydrazide (1.37 g, 0.01 mol) was added to a solution of 2-adamantanone (1.5 g, 0.01 mol) in ethanol (8 mL) and the mixture was heated under reflux for 4 hours. On cooling, the precipitated crystalline solid was filtered, dried and recrystallized from ethanol to yield 2.21 g (82%) of the title compound as colourless crystals. M.P.: 239–241 °C. Single crystals suitable for X-ray analysis were obtained by slow evaporation of $CHCl_3$:EtOH (1:1) at room temperature. ^{1}H NMR ($CDCl_3$, 500.13 MHz): δ 1.92–2.09 (m, 14H, Adamantane-H), 7.76 (d, 2H, pyridine-H, $J = 5.5$ Hz), 8.75 (d, 2H, pyridine-H, $J = 5.5$ Hz), 9.09 (s, 1H, NH). ^{13}C NMR ($CDCl_3$, 125.76 MHz): δ 27.66, 32.13, 36.12, 38.01, 39.53, 161.75 (adamantane-C), 122.85, 137.87, 148.95 (pyridine-C), 163.15 (C = O).

Table 1: Data collection and handling.

Crystal:	Colourless, needle, size 0.08×0.12×0.85 mm
Wavelength:	Mo K_α radiation (0.71073 Å)
μ:	0.83 cm^{-1}
Diffractometer, scan mode:	Bruker APEX-II CCD, φ and ω scans
$2\theta_{max}$:	66.32°
$N(hkl)_{measured}$, $N(hkl)_{unique}$:	12950, 4046
Criterion for I_{obs}, $N(hkl)_{gt}$:	$I_{obs} > 2\,\sigma(I_{obs})$, 3230
$N(param)_{refined}$:	185
Programs:	SHELX [17], Bruker data collection and reduction programs [18]

Table 2: Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters (Å^2).

Atom	Site	x	y	z	U_{iso}
H(1A)	4a	0.5392	0.8881	0.5245	0.015
H(2A)	4a	0.5575	1.1227	0.5435	0.023
H(2B)	4a	0.4222	1.0914	0.4769	0.023
H(3A)	4a	0.3123	1.2488	0.5671	0.019
H(4A)	4a	0.1174	1.1028	0.5077	0.021
H(4B)	4a	0.0564	1.1383	0.5946	0.021
H(5A)	4a	0.0353	0.9046	0.5732	0.015
H(7A)	4a	0.4879	0.8270	0.6589	0.022
H(7B)	4a	0.5988	0.9582	0.6564	0.022
H(8A)	4a	0.3788	0.9852	0.7491	0.019
H(9A)	4a	0.1859	0.8369	0.6878	0.020
H(9B)	4a	0.0959	0.9750	0.7045	0.020
H(10A)	4a	0.4621	1.1796	0.6808	0.020
H(10B)	4a	0.2668	1.1865	0.7002	0.020
H(13A)	4a	0.6134	0.6856	0.3413	0.016
H(14A)	4a	0.8185	0.5468	0.2921	0.025
H(15A)	4a	0.5446	0.2345	0.3317	0.023
H(16A)	4a	0.3313	0.3615	0.3838	0.016
H(1N2)	4a	0.466(2)	0.736(2)	0.458(1)	0.008(4)

Discussion

Isonicotinic acid hydrazide (INH) and its *N*′-arylidene derivatives were early recognized as effective antitubercular drugs

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Table 3: Atomic displacement parameters (Å^2).

Atom	Site	*x*	*y*	*z*	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
O(1)	4*a*	0.1651(1)	0.5878(1)	0.39484(7)	0.0099(5)	0.0170(6)	0.0168(6)	−0.0013(4)	−0.0003(5)	−0.0056(5)
N(1)	4*a*	0.2385(2)	0.7871(1)	0.49212(8)	0.0077(6)	0.0117(6)	0.0089(6)	0.0010(5)	0.0022(5)	−0.0008(5)
N(2)	4*a*	0.3619(2)	0.7209(1)	0.45016(8)	0.0061(6)	0.0114(6)	0.0123(6)	0.0007(5)	0.0010(5)	−0.0037(5)
N(3)	4*a*	0.7027(2)	0.3775(2)	0.3083(1)	0.0186(7)	0.0236(8)	0.0245(9)	0.0091(6)	−0.0005(7)	−0.0087(7)
C(1)	4*a*	0.4522(2)	0.9352(2)	0.5557(1)	0.0080(6)	0.0132(7)	0.0160(8)	−0.0007(6)	0.0006(6)	−0.0052(6)
C(2)	4*a*	0.4473(2)	1.0823(2)	0.5334(1)	0.0201(8)	0.0176(8)	0.0187(9)	−0.0085(7)	0.0027(7)	−0.0025(7)
C(3)	4*a*	0.3138(2)	1.1535(2)	0.5816(1)	0.0183(8)	0.0087(7)	0.0194(9)	0.0009(6)	−0.0031(7)	−0.0004(6)
C(4)	4*a*	0.1437(2)	1.0925(2)	0.5640(1)	0.0159(8)	0.0151(8)	0.0223(9)	0.0059(6)	−0.0062(7)	−0.0051(7)
C(5)	4*a*	0.1456(2)	0.9455(2)	0.5855(1)	0.0077(6)	0.0143(7)	0.0144(8)	0.0004(6)	0.0013(6)	−0.0052(6)
C(6)	4*a*	0.2822(2)	0.8785(2)	0.5393(1)	0.0090(6)	0.0091(7)	0.0101(7)	0.0012(6)	0.0007(6)	0.0001(6)
C(7)	4*a*	0.4875(2)	0.9213(2)	0.6443(1)	0.0189(8)	0.0164(8)	0.0190(9)	0.0047(7)	−0.0083(7)	−0.0053(7)
C(8)	4*a*	0.3544(2)	0.9941(2)	0.6919(1)	0.0212(8)	0.0155(8)	0.0118(7)	−0.0002(7)	−0.0022(7)	−0.0026(6)
C(9)	4*a*	0.1841(2)	0.9312(2)	0.6734(1)	0.0199(8)	0.0175(8)	0.0127(8)	−0.0033(7)	0.0033(7)	−0.0041(7)
C(10)	4*a*	0.3524(2)	1.1396(2)	0.6692(1)	0.0152(7)	0.0138(7)	0.0207(9)	−0.0018(6)	−0.0040(7)	−0.0080(7)
C(11)	4*a*	0.3116(2)	0.6175(2)	0.4051(1)	0.0103(7)	0.0108(7)	0.0094(7)	−0.0005(5)	−0.0002(6)	−0.0007(6)
C(12)	4*a*	0.4501(2)	0.5380(2)	0.3698(1)	0.0106(6)	0.0131(7)	0.0087(6)	0.0016(6)	−0.0016(6)	−0.0023(6)
C(13)	4*a*	0.5968(2)	0.5930(2)	0.3409(1)	0.0134(7)	0.0140(7)	0.0138(7)	−0.0011(6)	0.0014(6)	−0.0041(6)
C(14)	4*a*	0.7183(2)	0.5089(2)	0.3113(1)	0.0126(7)	0.028(1)	0.0210(9)	0.0000(7)	0.0042(7)	−0.0073(8)
C(15)	4*a*	0.5594(2)	0.3271(2)	0.3345(1)	0.0231(8)	0.0144(7)	0.0196(8)	0.0041(7)	−0.0042(7)	−0.0044(7)
C(16)	4*a*	0.4307(2)	0.4022(2)	0.3657(1)	0.0154(7)	0.0120(7)	0.0131(7)	0.0009(6)	−0.0020(7)	−0.0027(6)

[1–5]. In addition, derivatives of adamantane have long been known for their diverse chemotherapeutic activities such as antiviral [6–10], anticancer [11], antibacterial [12–14] and antitubercular [15, 16] agents. In continuation to our ongoing interest in the structural and pharmacological properties of adamantane derivatives [9, 12–14], we synthesized the title compound as hybrid INH-adamantane molecule with potential chemotherapeutic activity.

The crystal structure of title compound contains one molecule in the asymmetric unit. The molecule is a functionalized hydrazine with 4-pyridinoyl and adamantyl substituents attached to the two hydrazine nitrogen atoms, N1 and N2. The molecules packing in the crystal structure is stabilized by four intermolecular hydrogen bonds, of which O1 and N1 work as hydrogen bond acceptors and N2, C1 and C13 work as hydrogen bond donors. The distance of the interactions between N2—H1···N1 is 2.27 (2) Å, C1—H1A···N1 is 2.45 Å, C1—H1A···O1 is 2.46 Å and C13—H13A···O1 is 2.51 Å and the angles are 163°, 157°, 136°, and 159°, respectively.

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