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Crystal structure of 2-hydroxybenzylidene-5-methylisoxazole-4-carbohydrazide monohydrate, $C_{12}H_{13}N_3O_4$

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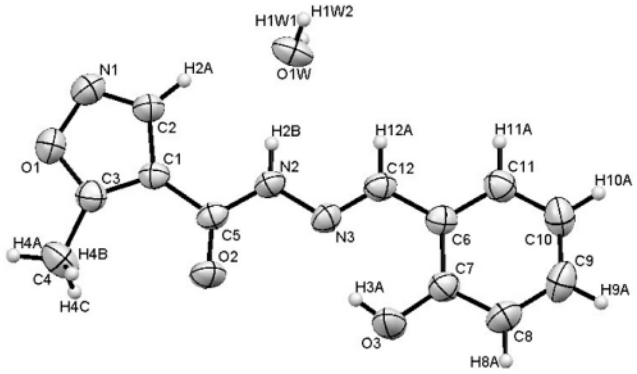

Abstract

$C_{12}H_{13}N_3O_4$, orthorhombic, *Pna*2$_1$ (no. 33), a = 12.8783(6) Å, b = 11.3108(6) Å, c = 8.6535(4) Å, V = 1260.5 Å^3, Z = 4, $R_{gt}(F)$ = 0.0323, $wR_{ref}(F^2)$ = 0.0922, T = 296 K.

Table 1. Data collection and handling.

Crystal:	white blocks, size 0.28×0.39×0.48 mm
Wavelength:	Mo K_α radiation (0.71073 Å)
μ:	1.06 cm^{-1}
Diffractometer, scan mode:	Bruker SMART CCD, φ and ω
$2\theta_{max}$:	54°
$N(hkl)_{measured}$, $N(hkl)_{unique}$:	12295, 1432
Criterion for I_{obs}, $N(hkl)_{gt}$:	$I_{obs} > 2\ \sigma(I_{obs})$, 1381
$N(param)_{refined}$:	182
Programs:	SHELX [10]

Source of material

Salicylaldehyde (1.50 g, 0.01 mol) was added into a solution of 5-methyl-4-isoxazole formyl hydrazine (2.0 g, 0.014 mol) in anhydrous ethanol (40 mL). The mixture was refluxed for 2 h, and then the precipitate formed was collected by filtration and washed with water, chloroform and ethanol. The product was recrystallized from ethanol and dried in vacuum at 65°C to give the title compound in 84.5% yield.

m.p.: 190.8-191.8 °C. **^{1}H-NMR** (200 MHz, CD_3COCD_3, ppm), δ: 2.68 (s, 3H, –CH_3) 6.8-7.4 (m, 4H, Ar–H), 8.4 (s, 1H, –N=CH–), 8.8 (s, 1H, 3–isoxazole–H), 11.2 (s, 1H, –CO–NH–), 11.4 (s, 1H, –OH). **IR** (KBr, cm^{-1}): 3172.7(w) (N–H) , 1654.8(s) (C=O) , 1620.1(s) (C=N). **MS**(ESI):m/z 246.2(M+1).

Experimental details

H atoms bonded to N atoms were located in a difference map and refined with distance restraints of N–H = 0.86Å, and with U_{iso}(H)=1.2Ueq(N). Other H atoms were positioned geometrically and refined using a riding model (including free rotation about the ethanol C–C bond), with C–H = 0.93-0.98 Å and with U_{iso}(H) = 1.2 (for methyl groups) times U_{eq}.

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Discussion

Hydrazones, which are usually prepared via the condensation of aldehydes or ketones with hydrazines, are well known to be biologically important and interest in studying this class of compound lies in its antibacterial, antitumour and antitubercular activities [1]. In addition, it has been used as an analytical reagent [2] and as polymer-coating, ink, pigment [3] and fluorescent materials [4]. Previous studies have focused on their extraordinary ability to form complexes with metal ions , as well as exploring the development of new drugs based on their biological properties [5]. For instance, Ainscough *et al.* [5] had extensively studied the coordination chemistry of acylhydrazones as well as a number of copper(II) complexes. Similarly, Singh *et al.* [5] synthesized the cobalt(II) complexes with some acylhydrazones, which showed a fair antifungal and antibacterial activity against a number of fungi and bacteria. Otherwise, photoluminescence, photoabsorption and photoemission studies of hydrazone thin films used as hole transporting material in organic light emitting diodes (OLEDs) were reported [6]. On the other hand, isoxazole compounds have a rich history with both pharmaceutical and agricultural applications due to their apoptotic activity [7], plant-growth regulating activity [8] and antibacterial activity [9]. In the title compound, $C_{12}H_{11}N_3O_3$, the molecule is almost planar, the dihedral angle formed by the benzene and isoxazole rings is 2.03 (8)° and in the crystal structure screw-related molecules are linked by O–H···N, O–H···O and N–H···O hydrogen bonds and π-π packing to form chains along the a-axis. The chains are interlinked by the water molecules *via* N–HO and C–HO hydrogen bonds. In the structure of the title complex, a three-dimensional (3-D) network supramolecule is formed by the hydrogen bonds interactions and π-π interactions [centroid–centroid separation = 3.663 (2) Å].

Table 2. Atomic coordinates and displacement parameters (in Å^2).

Atom	Site	x	y	z	U_{iso}
H(2B)	4*a*	0.1744	0.1523	0.7368	0.051
H(1W1)	4*a*	–0.026(2)	0.123(2)	0.776(4)	0.084
H(1W2)	4*a*	–0.019(2)	0.226(2)	0.704(4)	0.084
H(2A)	4*a*	0.1002	0.0704	0.5294	0.056
H(4A)	4*a*	0.4309	–0.0251	0.2427	0.107
H(4B)	4*a*	0.4575	–0.0367	0.4189	0.107
H(4C)	4*a*	0.4555	0.0882	0.3404	0.107
H(8A)	4*a*	0.4563	0.3500	1.3347	0.069
H(9A)	4*a*	0.3336	0.4143	1.5086	0.070
H(10A)	4*a*	0.1598	0.3912	1.4576	0.068
H(11A)	4*a*	0.1075	0.3080	1.2277	0.059
H(12A)	4*a*	0.1437	0.2176	0.9776	0.051
H(3A)	4*a*	0.403(2)	0.224(2)	0.994(3)	0.070(9)

Table 3. Atomic coordinates and displacement parameters (in Å^2).

Atom	Site	*x*	*y*	*z*	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
N(1)	4*a*	0.1439(2)	–0.0028(2)	0.3335(3)	0.048(1)	0.066(1)	0.056(1)	–0.0047(9)	–0.0081(9)	–0.012(1)
N(2)	4*a*	0.2403(1)	0.1571(2)	0.7518(2)	0.0321(8)	0.0543(9)	0.0399(9)	–0.0018(7)	–0.0076(7)	–0.0024(8)
N(3)	4*a*	0.2788(1)	0.2006(2)	0.8883(2)	0.0401(8)	0.0488(9)	0.0358(9)	0.0008(7)	–0.0086(8)	–0.0013(7)
O(1W)	4*a*	0.0171(1)	0.1679(1)	0.7294(2)	0.0322(7)	0.0646(9)	0.071(1)	0.0027(6)	0.0057(8)	0.0108(9)
O(1)	4*a*	0.2453(1)	–0.0196(1)	0.2774(2)	0.0562(9)	0.0595(9)	0.0491(9)	–0.0023(7)	0.0009(8)	–0.0155(8)
O(2)	4*a*	0.4016(1)	0.1320(1)	0.6549(2)	0.0307(7)	0.0630(9)	0.061(1)	0.0010(6)	–0.0087(7)	–0.0070(8)
O(3)	4*a*	0.4316(1)	0.2552(2)	1.0747(2)	0.0354(8)	0.109(2)	0.0502(9)	0.0052(8)	–0.0047(7)	–0.014(1)
C(1)	4*a*	0.2606(1)	0.0686(2)	0.5035(3)	0.0325(9)	0.0358(9)	0.043(1)	0.0001(7)	–0.0015(9)	0.0017(8)
C(2)	4*a*	0.1553(2)	0.0488(2)	0.4659(3)	0.035(1)	0.056(1)	0.049(1)	–0.0004(8)	–0.0036(9)	–0.008(1)
C(3)	4*a*	0.3127(2)	0.0233(2)	0.3808(3)	0.043(1)	0.042(1)	0.047(1)	0.0001(8)	0.001(1)	–0.0017(9)
C(4)	4*a*	0.4239(2)	0.0114(3)	0.3423(4)	0.048(1)	0.087(2)	0.080(2)	0.007(1)	0.017(1)	–0.013(2)
C(5)	4*a*	0.3073(1)	0.1221(2)	0.6423(3)	0.0350(9)	0.0360(9)	0.042(1)	0.0000(7)	–0.0068(8)	0.0030(9)
C(6)	4*a*	0.2500(2)	0.2771(2)	1.1395(3)	0.0377(9)	0.0405(9)	0.039(1)	0.0009(7)	–0.0030(8)	0.0058(9)
C(7)	4*a*	0.3562(2)	0.2908(2)	1.1731(3)	0.039(1)	0.055(1)	0.039(1)	0.0012(8)	–0.0051(9)	0.002(1)
C(8)	4*a*	0.3861(2)	0.3415(2)	1.3120(3)	0.049(1)	0.072(2)	0.053(1)	–0.004(1)	–0.014(1)	–0.008(1)
C(9)	4*a*	0.3126(2)	0.3794(2)	1.4164(3)	0.075(2)	0.058(1)	0.042(1)	0.004(1)	–0.011(1)	–0.009(1)
C(10)	4*a*	0.2090(2)	0.3664(2)	1.3859(3)	0.064(2)	0.062(1)	0.044(1)	0.014(1)	0.005(1)	–0.006(1)
C(11)	4*a*	0.1780(2)	0.3161(2)	1.2483(3)	0.043(1)	0.055(1)	0.049(1)	0.0040(9)	0.000(1)	0.002(1)
C(12)	4*a*	0.2145(2)	0.2285(2)	0.9941(3)	0.0355(9)	0.049(1)	0.043(1)	–0.0002(8)	–0.0068(9)	0.0030(9)

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