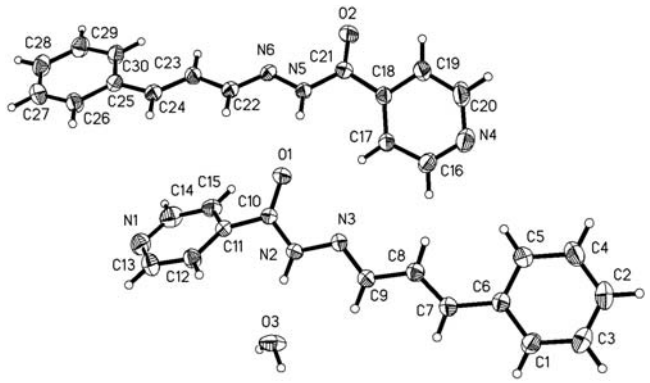
Refinement of the crystal structure of bis[*N′*-(3-phenylallylidene)-isonicotinohydrazide] hydrate, $2C_{15}H_{13}N_3O \cdot H_2O$

Liu-Shui Yan*, De-He Huang and Yuan Chen

Nanchang Hangkong University, School of Environment and Chemical Engineering, Nanchang 330063, Jiangxi, P. R. China

Received September 4, 2010, accepted and available on-line January 25, 2011; CCDC no. 1267/3200



Abstract

$C_{30}H_{28}N_6O_3$, monoclinic, *P*1*c*1 (no. 7), a = 12.704(1) Å, b = 11.012(1) Å, c = 10.071(1) Å, β = 105.438(1)°, V = 1357.9 Å^3, Z = 2, $R_{gt}(F)$ = 0.037, $wR_{ref}(F^2)$ = 0.078, T = 296 K.

Source of material

The mixture of cinnamaldehyde (0.01 mol, 1.37 g) and anhydrous methanol (20 ml) was stirred for dissolution absolutely, then isoniazid (0.01 mol) was added dropwise and the solution was stirred at room temperature for 5 h. After the reaction was completed, the solution was filtered and poured into a beaker, and kept at room temperature for 3 d. Natural evaporation gave colorless crystals of the title compound suitable for X-ray analysis (yield 79.5 %, mp. 473 - 476 K).

Experimental details

The hydrogen atoms bound to carbon atoms were positioned geometrically and allowed to ride on their parent atoms with d(C—H) = 0.93 Å, d(N—H) = 0.86 Å and U_{iso}(H) = 1.2 U_{eq}(C,N). The Flack parameter is 0.0(10), confirming the determined absolute structure.

Discussion

Schiff base (or azomethine), named after Hugo Schiff, is a functional group that contains a carbon-nitrogen double bond. Schiff bases are of the general formula R1R2C=N–R3, where R3 is an aryl or alkyl group that makes the Schiff base a stable imine. Up to date, Schiff bases and their complexes have been extensively studied because of their interesting and various properties, e.g., the ability to bind oxygen reversibly, catalytic activity [1,2], photochromic properties [3], complexing ability towards certain toxic metals [4], chemotherapeutic properties and unusual physiochemical properties [5].

The crystal structure of the title compound has been determined previously [6] with a reliability factor $R_{gt}(F)$ = 0.044. The present redetermination converges with lower reliability factors [$R_{gt}(F)$ = 0.037] and the quality of the data allowed the determination of the positions of the lattice water molecules.

The title compound crystallizes in the monoclinic space group *Pc*, the same as the space group in [6]. The isomer of *N′*-(3-phenylallylidene)isonicotinohydrazide monohydrate [7] crystallizes in the monoclinic space group $P2_1/c$.

The bond lengths of C=N are 1.278 and 1.281(3) Å, comparing with 1.273(5) and 1.287(6) Å [6] and 1.285 Å [7] are all in the normal range of C=N double bond [8]. The bond angles ∠C22–N6–N5 and ∠C9–N3–N2 are 115.8° and 116.1° for the title molecule, 108.8° and 114.5° in [6] and 114.7° in [7]. The dihedral angles between the pyridine and benzene rings are 56.2(7)° and 48.6(7)°, also comparable with 56.6(2)° and 45.2(2)° [6] and 35.5(7)° [7]. Intramolecular N2–H2A···O3 hydrogen bond and intermolecular O3–H1W···N3, O3–H2W···.N6 and N5–H5A···O1 hydrogen bonds link the molecules of the title compound to a layer in (100). There are also various hydrogen bonds O–H···O, O–H···N, N–H···O, C–H···O and C–H···π interactions in the crystal structure of isomer [7]. In [6] only intermolecular N–H···O hydrogen bonds are reported, owing to the non-found water molecules.

Table 1. Data collection and handling.

Crystal:	colorless block, size 0.07 × 0.16 × 0.34 mm
Wavelength:	Mo K_α radiation (0.71073 Å)
μ:	0.85 cm^{-1}
Diffractometer, scan mode:	Bruker APEX-II CCD, φ/ω
$2\theta_{max}$:	55°
$N(hkl)_{measured}$, $N(hkl)_{unique}$:	12065, 3112
Criterion for I_{obs}, $N(hkl)_{gt}$:	$I_{obs} > 2\,\sigma(I_{obs})$, 2275
$N(param)_{refined}$:	352
Programs:	SHELXS-97, SHELXL-97 [9]

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

Atom	Site	x	y	z	U_{iso}
H(2)	2*a*	0.4506	0.2045	–0.5137	0.085
H(1)	2*a*	0.5855	0.2030	–0.3100	0.070
H(5)	2*a*	0.6677	0.5427	–0.3833	0.071
H(4)	2*a*	0.5350	0.5402	–0.5903	0.084
H(7)	2*a*	0.7299	0.3007	–0.1396	0.061
H(2A)	2*a*	1.0103	0.4668	0.2123	0.054
H(12)	2*a*	1.1980	0.4681	0.3549	0.062
H(13)	2*a*	1.3085	0.4654	0.5759	0.075
H(3)	2*a*	0.4257	0.3719	–0.6553	0.087
H(14)	2*a*	1.1714	0.7544	0.6698	0.084

* Correspondence author (e-mail: lsyan2010@sohu.com)

Table 2. Continued.

Atom	Site	x	y	z	U_{iso}
H(15)	2*a*	1.0593	0.7697	0.4513	0.066
H(16)	2*a*	–0.2880	–0.2983	–0.0972	0.070
H(17)	2*a*	–0.1630	–0.1894	0.0647	0.060
H(19)	2*a*	–0.2032	0.0840	–0.2052	0.071
H(20)	2*a*	–0.3194	–0.0377	–0.3602	0.091
H(22)	2*a*	0.1401	–0.0679	0.3257	0.058
H(23)	2*a*	0.1638	0.1525	0.4804	0.062
H(24)	2*a*	0.2839	–0.0569	0.5322	0.061
H(26)	2*a*	0.4383	–0.0710	0.7255	0.071
H(27)	2*a*	0.5529	0.0035	0.9245	0.084
H(28)	2*a*	0.5199	0.1920	1.0074	0.084
H(29)	2*a*	0.3712	0.3037	0.8865	0.080
H(30)	2*a*	0.2586	0.2314	0.6846	0.068
H(8)	2*a*	0.8037	0.5212	–0.1991	0.058
H(9)	2*a*	0.8792	0.3820	0.0463	0.057
H(5A)	2*a*	–0.0064	–0.0849	0.1507	0.055
H(1W)	2*a*	0.9895	0.3190	0.3964	0.109
H(2W)	2*a*	0.9809	0.2497	0.2811	0.109

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}
C(2)	2*a*	0.4950(3)	0.2723(3)	–0.4881(3)	0.060(2)	0.085(2)	0.064(2)	–0.020(2)	0.008(2)	–0.018(2)
C(1)	2*a*	0.5756(2)	0.2716(3)	–0.3658(3)	0.060(2)	0.057(2)	0.053(2)	–0.010(1)	0.010(1)	–0.003(1)
C(6)	2*a*	0.6425(2)	0.3722(2)	–0.3246(3)	0.049(2)	0.049(2)	0.044(2)	–0.002(1)	0.008(1)	–0.003(1)
C(5)	2*a*	0.6252(2)	0.4736(3)	–0.4096(3)	0.053(2)	0.054(2)	0.064(2)	0.000(1)	0.004(2)	0.005(1)
C(4)	2*a*	0.5449(3)	0.4725(3)	–0.5332(3)	0.062(2)	0.078(2)	0.063(2)	0.011(2)	0.003(2)	0.014(2)
C(7)	2*a*	0.7272(2)	0.3698(2)	–0.1934(3)	0.056(2)	0.046(2)	0.046(2)	–0.001(1)	0.004(1)	0.000(1)
N(3)	2*a*	0.9331(2)	0.5442(2)	0.0374(2)	0.047(1)	0.042(1)	0.044(1)	–0.001(1)	–0.001(1)	0.0026(9)
N(2)	2*a*	0.9998(2)	0.5356(2)	0.1705(2)	0.050(1)	0.034(1)	0.044(1)	–0.0012(9)	0.001(1)	0.0042(9)
C(10)	2*a*	1.0477(2)	0.6361(2)	0.2326(3)	0.043(1)	0.038(1)	0.054(2)	–0.001(1)	0.002(1)	0.003(1)
C(11)	2*a*	1.1166(2)	0.6210(2)	0.3772(3)	0.043(1)	0.039(1)	0.045(2)	–0.004(1)	0.007(1)	–0.001(1)
C(12)	2*a*	1.1917(2)	0.5285(2)	0.4169(3)	0.054(2)	0.053(2)	0.046(2)	0.005(1)	0.006(1)	–0.003(1)
C(13)	2*a*	1.2573(3)	0.5273(3)	0.5506(3)	0.058(2)	0.074(2)	0.049(2)	0.008(2)	0.004(2)	0.004(2)
C(3)	2*a*	0.4797(2)	0.3721(4)	–0.5723(3)	0.050(2)	0.102(3)	0.056(2)	0.001(2)	–0.002(2)	–0.003(2)
C(14)	2*a*	1.1774(3)	0.6965(3)	0.6050(3)	0.093(2)	0.068(2)	0.052(2)	–0.014(2)	0.025(2)	–0.015(2)
C(15)	2*a*	1.1093(2)	0.7064(2)	0.4740(3)	0.064(2)	0.044(2)	0.058(2)	0.001(1)	0.018(2)	–0.003(1)
C(16)	2*a*	–0.2696(2)	–0.2208(3)	–0.1203(3)	0.052(2)	0.054(2)	0.064(2)	–0.006(1)	0.008(2)	–0.007(1)
C(17)	2*a*	–0.1950(2)	–0.1553(2)	–0.0211(3)	0.047(2)	0.047(2)	0.049(2)	0.002(1)	0.000(1)	–0.001(1)
C(18)	2*a*	–0.1685(2)	–0.0392(2)	–0.0508(3)	0.038(1)	0.046(1)	0.045(2)	0.002(1)	0.006(1)	–0.000(1)
C(19)	2*a*	–0.2177(2)	0.0056(3)	–0.1806(3)	0.054(2)	0.069(2)	0.050(2)	–0.004(1)	0.005(1)	0.012(1)
C(20)	2*a*	–0.2885(3)	–0.0684(4)	–0.2724(3)	0.064(2)	0.108(3)	0.045(2)	–0.011(2)	–0.005(2)	0.010(2)
C(21)	2*a*	–0.0965(2)	0.0449(2)	0.0501(3)	0.046(2)	0.043(2)	0.053(2)	–0.000(1)	0.008(1)	0.002(1)
C(22)	2*a*	0.1208(2)	0.0106(2)	0.3435(3)	0.047(2)	0.041(1)	0.054(2)	–0.003(1)	0.006(1)	–0.002(1)
C(23)	2*a*	0.1830(2)	0.0734(2)	0.4644(3)	0.051(2)	0.044(1)	0.054(2)	–0.000(1)	0.002(1)	–0.004(1)
C(24)	2*a*	0.2670(2)	0.0215(2)	0.5537(3)	0.051(2)	0.043(1)	0.054(2)	–0.003(1)	0.008(1)	–0.004(1)
C(25)	2*a*	0.3363(2)	0.0718(3)	0.6814(3)	0.044(2)	0.052(2)	0.049(2)	–0.006(1)	0.008(1)	0.001(1)
C(26)	2*a*	0.4249(2)	0.0054(3)	0.7568(3)	0.057(2)	0.060(2)	0.054(2)	0.005(1)	0.003(1)	0.003(1)
C(27)	2*a*	0.4934(3)	0.0494(3)	0.8767(3)	0.065(2)	0.077(2)	0.057(2)	0.004(2)	–0.002(2)	0.009(2)
C(28)	2*a*	0.4740(3)	0.1617(3)	0.9263(3)	0.062(2)	0.090(2)	0.050(2)	–0.013(2)	0.002(2)	–0.007(2)
C(29)	2*a*	0.3854(2)	0.2283(3)	0.8534(3)	0.063(2)	0.067(2)	0.065(2)	–0.006(2)	0.011(2)	–0.017(2)
C(30)	2*a*	0.3176(2)	0.1848(3)	0.7326(3)	0.047(2)	0.056(2)	0.061(2)	0.001(1)	0.005(1)	–0.006(1)
N(1)	2*a*	1.2515(2)	0.6089(3)	0.6444(2)	0.076(2)	0.080(2)	0.042(1)	–0.009(2)	0.005(1)	0.001(1)
C(8)	2*a*	0.8001(2)	0.4549(2)	–0.1433(3)	0.051(2)	0.045(2)	0.044(2)	0.001(1)	0.007(1)	–0.002(1)
C(9)	2*a*	0.8742(2)	0.4513(2)	–0.0079(3)	0.049(2)	0.042(1)	0.047(2)	0.001(1)	0.005(1)	0.001(1)
N(4)	2*a*	–0.3164(2)	–0.1806(3)	–0.2459(3)	0.060(2)	0.086(2)	0.057(2)	–0.012(2)	0.003(1)	–0.007(2)
N(5)	2*a*	–0.0191(2)	–0.0083(2)	0.1528(2)	0.047(1)	0.032(1)	0.051(1)	–0.001(1)	–0.000(1)	–0.004(1)
N(6)	2*a*	0.0389(2)	0.0622(2)	0.2603(2)	0.050(1)	0.039(1)	0.051(1)	–0.008(1)	0.001(1)	–0.005(1)
O(1)	2*a*	1.0365(2)	0.7364(2)	0.1771(2)	0.075(1)	0.039(1)	0.076(1)	–0.0060(9)	–0.015(1)	0.010(1)
O(2)	2*a*	–0.1085(2)	0.1543(2)	0.0411(2)	0.079(2)	0.038(1)	0.081(1)	0.001(1)	–0.007(1)	0.006(1)
O(3)	2*a*	0.9910(2)	0.3197(2)	0.3141(2)	0.123(2)	0.038(1)	0.062(1)	–0.004(1)	0.034(1)	–0.0016(9)

Acknowledgment. This work was supported by the National Natural Science Foundation of China (grant no. 20961007).

References

1. Zhou, M. D.; Zhao, J.; Li, J.; Yue, S.; Bao, C. N.; Mink, J.; Zang, S. L.; Kühn, F. E.: MTO Schiff-Base Complexes: Synthesis, Structures and Catalytic Applications in Olefin Epoxidation. Chem. Eur. J. **13** (2007) 158-166.
2. Sun, B.; Chen, J. R.; Hu, J. Y.; Li, X. J.: Dioxygen affinities and catalytic oxidation activities of cobalt complexes with Schiff bases containing crown ether. J. Inorg. Biochem. **100** (2006) 1308-1313.
3. Koichi, T.; Ryota, S.; Caira, M. R.: Synthesis, crystal structures and photochromic properties of novel chiral Schiff base macrocycles. Tetrahedron Lett. **51** (2010) 449-452.
4. Soliman, A. A.; Linert, W.: Investigations on new transition metal chelates of the 3-methoxysalicylidene-2-aminothiophenol Schiff base. Thermochim. Acta. **338** (1999) 67-75.

5. Charette, J. J.; De Hoffmann, E.: Physicochemical properties of Schiff bases. 4. Tautomeric equilibrium and kinetics of hydrolysis of *N*-benzylideneaniline derivatives. J. Org. Chem. **44** (1979) 2256-2262.
6. Bao, F.-Y.: *N'*-(3-Phenylallylidene)isonicotinohydrazide. Acta Crystallogr. **E64** (2008) o1789.
7. Archana, R.; Saradhadevi, N.; Manimekalai, A.; Thiruvalluvar, A.; Butcher, R. J.: *N'*-(3-Phenylallylidene)nicotinohydrazide Monohydrate. Acta Crystallogr. **E65** (2009) o2833.
8. Yin, Z. G.; Fei, Z. L.; Qian, H. Y.; Zhang, C. X.: Crystal structure of *N*-(benzo[*d*][1,3]dioxol-5-ylmethyl)-4-methylaniline, $C_{15}H_{13}NO_2$. Z. Kristallogr. NCS **225** (2010) 251-252.
9. Sheldrick, G. M.: A short history of SHELX. Acta Crystallogr. **A64** (2008) 112-122.