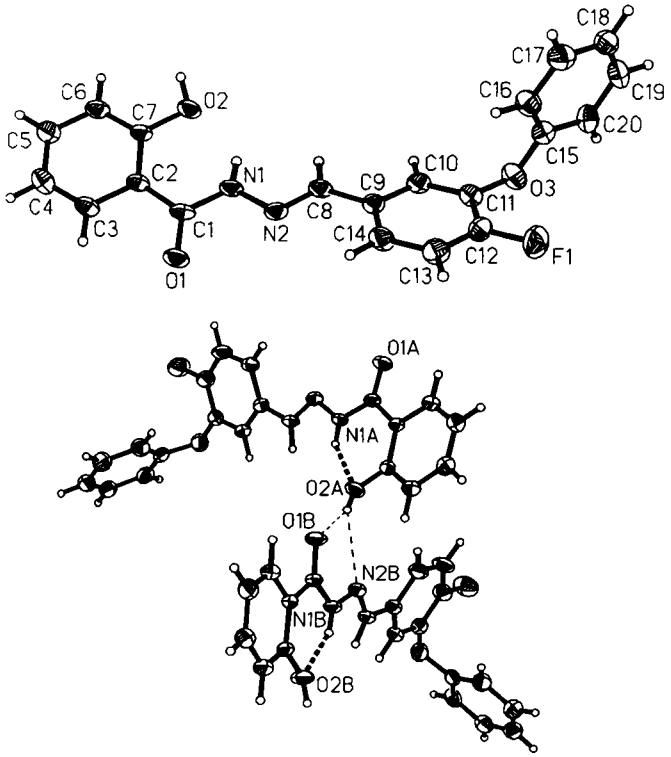
Crystal structure of 2′-(3-benzoxy-4-fluorobenzylidene)-2-hydroxybenzoylhydrazide, $C_{20}H_{15}FN_2O_3$

H.-D. Liang[I], J.-G. Yang[II], H. Yan[I] and F.-Y. Pan*[,I]

[I] Taizhou University, Department of Chemistry, Taizhou, Zhejiang, 317000 P. R. China
[II] Taizhou University, Office of Assets Administration, Taizhou, Zhejiang, 317000 P. R. China

Received September 27, 2005, accepted and available on-line December 11, 2005; CCDC no. 1267/1651



Abstract

$C_{20}H_{15}FN_2O_3$, orthorhombic, $Pna2_1$ (no. 33), $a = 25.765(4)$ Å, $b = 11.185(2)$ Å, $c = 5.9115(8)$ Å, $V = 1703.6$ Å^3, $Z = 4$, $R_{gt}(F) = 0.052$, $wR_{ref}(F^2) = 0.125$, $T = 293$ K.

Source of material

2-Hydroxybenzoylhydrazine (0.02 mol, 3.04 g) was dissolved in anhydrous ethanol (50 ml) and 3-benzoxy-4-fluorobenzaldehyde (0.02 mol, 4.32 g) was added to the solution. The mixture was refluxed for 2.5 h and the precipitate formed was collected by filtration and washed with ethanol. The product was recrystallized from ethanol and dried under reduced pressure to give the title compound. The compound (2.0 mmol, 0.70 g) was dissolved in dimethylformamide (30 ml) and kept at room temperature for 35 d to obtain yellow block-shaped crystals, which were collected and washed with distilled water.

Experimental details

While the H atoms bonded to C atoms were fixed geometrically and treated as riding, the H atoms bonded to O atoms were refined freely due to their contribution to the intermolecular bonding.

* Correspondence author (e-mail: panfy@tzc.edu.cn)

Discussion

Some benzoylhydrazone compounds possess bacteriostatic activity. This type of compound has wide application in tuberculosis treatment and exhibits also fungicidal activity [1]. Some hydrazonecarbonyl compounds show bioactivity [2]. In order to explore more effective antibacterial medicines, we have synthesized the title compound.

The title molecule (figure, top) is non-planar; the dihedral angle between the [C2 – C7] and [C9 – C14] aromatic rings is 30.5(1)°, the respective angle between the [C9 – C14] and [C15 – C20] aromatic rings is 81.9(1)°. As a result of conjugation, the C1=O1 distance (1.238(4) Å) is longer than the normal value of 1.20 Å, and the C1—N1 bond distance (1.341(4) Å) is longer than the C=N double-bond distance and shorter than the C—N single-bond distance [3]. The values of angles C7–C2–C1, C3–C2–C1, C8–C9–C14 and C8–C9–C10 close to 120° and of the distances C1—C2 (1.464(4) Å), C8—C9 (1.443(5) Å) confirm sp^2 hybridization of carbon atoms [4]. An intramolecular N1–H1···O2 hydrogen bond which forms a six-membered ring is observed (figure, bottom). In addition, two intermolecular hydrogen bonds occur (O2–H2···O1^i and O2–H2···N2^i, symmetry code (i) $-x+½,y+½,z-½$), which link the hydroxyl H with the keto group and amide NH of an adjacent molecule and form a 5-membered ring (figure, bottom).

Table 1. Data collection and handling.

Crystal:	yellow block, size 0.09 × 0.48 × 0.52 mm
Wavelength:	Mo K_α radiation (0.71073 Å)
μ:	1.00 cm^{-1}
Diffractometer, scan mode:	Bruker SMART CCD, φ/ω
$2\theta_{max}$:	54°
$N(hkl)_{measured}$, $N(hkl)_{unique}$:	9691, 3677
Criterion for I_{obs}, $N(hkl)_{gt}$:	$I_{obs} > 2\,\sigma(I_{obs})$, 1773
$N(param)_{refined}$:	244
Programs:	SHELXS-97 [5], SHELXL-97 [6]

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

Atom	Site	x	y	z	U_{iso}
H(3)	4*a*	0.3330	–0.1220	–0.0874	0.063
H(4)	4*a*	0.3747	–0.0528	–0.4036	0.077
H(5)	4*a*	0.3647	0.1433	–0.5162	0.073
H(6)	4*a*	0.3187	0.2739	–0.2892	0.067
H(8)	4*a*	0.1967	0.1595	0.5927	0.058
H(10)	4*a*	0.1509	0.2139	0.9322	0.061
H(13)	4*a*	0.0760	–0.1600	0.9764	0.088
H(14)	4*a*	0.1383	–0.1231	0.7065	0.075

Table 2. Continued.

Atom	Site	x	y	z	U_{iso}
H(16)	4a	0.0208	0.1920	0.9265	0.076
H(17)	4a	–0.0569	0.2946	0.8962	0.085
H(18)	4a	–0.0815	0.4287	1.1699	0.080
H(19)	4a	–0.0324	0.4518	1.4846	0.079
H(20)	4a	0.0448	0.3457	1.5274	0.073
H(1)	4a	0.240(1)	0.120(3)	0.306(5)	0.04(1)
H(2)	4a	0.260(2)	0.287(4)	–0.026(9)	0.10(3)

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}
O(1)	4a	0.27660(8)	–0.1296(2)	0.2417(5)	0.069(2)	0.030(1)	0.082(2)	0.006(1)	0.009(2)	0.011(1)
O(2)	4a	0.2673(1)	0.2310(2)	0.0701(5)	0.072(2)	0.035(2)	0.085(2)	0.015(1)	0.025(2)	0.020(2)
O(3)	4a	0.08704(9)	0.2020(2)	1.2646(5)	0.056(2)	0.065(2)	0.073(2)	0.001(1)	–0.003(2)	–0.018(2)
N(1)	4a	0.2407(1)	0.0482(3)	0.3211(5)	0.050(2)	0.025(2)	0.068(2)	0.002(1)	0.006(2)	0.013(2)
N(2)	4a	0.2101(1)	0.0020(2)	0.4898(5)	0.048(2)	0.033(2)	0.059(2)	–0.002(1)	0.003(2)	0.014(2)
F(1)	4a	0.04272(9)	–0.0186(2)	1.2804(5)	0.104(2)	0.077(2)	0.113(2)	–0.011(1)	0.052(2)	0.011(2)
C(1)	4a	0.2716(1)	–0.0221(3)	0.1965(6)	0.043(2)	0.028(2)	0.068(3)	0.001(2)	–0.009(2)	0.011(2)
C(2)	4a	0.2993(1)	0.0333(3)	0.0072(6)	0.036(2)	0.030(2)	0.060(2)	–0.000(1)	–0.004(2)	0.004(2)
C(3)	4a	0.3295(1)	–0.0420(3)	–0.1277(6)	0.052(2)	0.033(2)	0.071(3)	0.002(2)	0.000(2)	0.001(2)
C(4)	4a	0.3544(1)	–0.0009(3)	–0.3184(7)	0.052(2)	0.056(3)	0.083(3)	0.001(2)	0.009(2)	–0.011(2)
C(5)	4a	0.3493(1)	0.1163(3)	–0.3835(7)	0.052(2)	0.053(2)	0.078(3)	–0.001(2)	0.012(2)	0.012(2)
C(6)	4a	0.3209(1)	0.1936(3)	–0.2496(7)	0.052(2)	0.039(2)	0.075(3)	0.002(2)	0.004(2)	0.015(2)
C(7)	4a	0.2959(1)	0.1537(3)	–0.0593(6)	0.038(2)	0.032(2)	0.062(2)	0.003(2)	0.004(2)	0.013(2)
C(8)	4a	0.1883(1)	0.0795(3)	0.6146(6)	0.052(2)	0.027(2)	0.067(2)	–0.002(2)	0.003(2)	0.009(2)
C(9)	4a	0.1513(1)	0.0497(3)	0.7890(6)	0.044(2)	0.038(2)	0.061(2)	–0.003(2)	0.006(2)	0.009(2)
C(10)	4a	0.1364(1)	0.1379(3)	0.9422(6)	0.048(2)	0.037(2)	0.066(2)	–0.007(2)	0.003(2)	0.006(2)
C(11)	4a	0.1007(1)	0.1140(3)	1.1068(6)	0.047(2)	0.049(2)	0.062(3)	0.002(2)	–0.001(2)	–0.003(2)
C(12)	4a	0.0791(1)	0.0031(4)	1.1181(7)	0.063(2)	0.055(3)	0.072(3)	–0.005(2)	0.019(2)	0.014(2)
C(13)	4a	0.0919(2)	–0.0855(3)	0.9682(8)	0.080(3)	0.038(2)	0.103(4)	–0.010(2)	0.030(3)	0.010(2)
C(14)	4a	0.1285(1)	–0.0628(3)	0.8062(7)	0.064(2)	0.038(2)	0.086(3)	–0.002(2)	0.016(2)	0.000(2)
C(15)	4a	0.0396(1)	0.2597(3)	1.2300(7)	0.047(2)	0.046(2)	0.059(2)	–0.010(2)	0.000(2)	–0.003(2)
C(16)	4a	0.0101(2)	0.2438(3)	1.0406(7)	0.065(2)	0.059(3)	0.066(3)	0.003(2)	–0.001(2)	–0.013(2)
C(17)	4a	–0.0359(2)	0.3062(4)	1.0222(8)	0.069(3)	0.072(3)	0.073(3)	0.001(2)	–0.006(2)	0.006(3)
C(18)	4a	–0.0510(2)	0.3851(4)	1.1868(8)	0.061(3)	0.058(3)	0.080(3)	0.003(2)	0.012(3)	0.010(2)
C(19)	4a	–0.0217(2)	0.3992(4)	1.3720(8)	0.058(3)	0.056(3)	0.082(3)	–0.006(2)	0.021(2)	–0.015(2)
C(20)	4a	0.0246(1)	0.3360(3)	1.3983(7)	0.061(2)	0.061(3)	0.060(3)	–0.014(2)	0.009(2)	–0.020(2)

Acknowledgments. The authors acknowledge financial support by the Zhejiang Provincial Natural Science Foundation of China (grant no. M203115) and Scientific Research Fund of Zhejiang Provincial Education Department (grant no. 20050057).

References

1. Edwards, E. I.; Epton, R.; Marr, G.: Organometallic derivatives of penicillins and cephalosporins a new class of semi-synthetic antibiotics. J. Organomet. Chem. **85** (1975) C23-C25.
2. Fan, Z. J.; Zhong, B.; Wang, S. H.; Li, Z. M.: Synthesis and Fungicidal Activity of Derivatives of 1,3-Dimethyl-5-methylthio-4-phenylhydrazonocarboxyl Pyrazole. Chin. J. Appl. Chem. **4** (2003) 365-367.
3. John, A. D.: Lang's Handbook of Chemistry. McGraw-Hill, New York 1998.
4. Liu, S. L.; Dai, J. F.; Chen, Y.; Cao, G. B.; Liu, H. W.: Synthesis and Crystal Structure of 3-(2-Hydroxy-phenyl)-5-phenyl-6-ethoxycarbonyl-cyclohex-2-enone. Chin. J. Org. Chem. **12** (2004) 1583-1586.
5. Sheldrick, G. M.: Phase Annealing in SHELX-90: Direct Methods for Larger Structures. Acta Crystallogr. A**46** (1990) 467-473.
6. Sheldrick, G. M.: SHELXL-97. Program for the Refinement of Crystal Structures. University of Göttingen, Germany 1997.