Crystal structure of 3-methoxy-4-hydroxybenzylidene-malononitrile, $C_{11}H_8N_2O_2$

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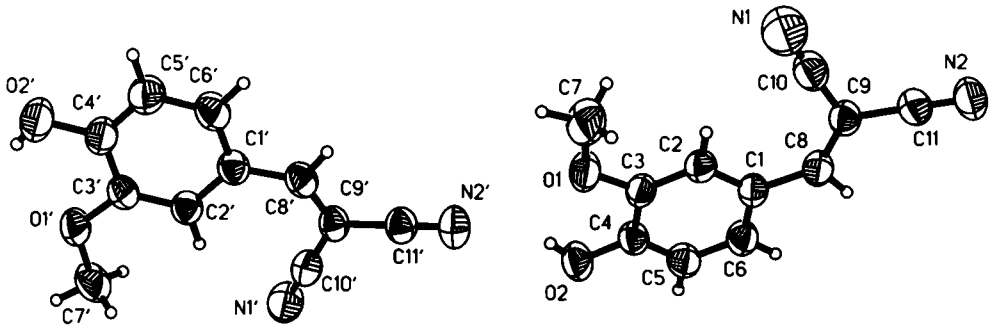

Abstract

$C_{11}H_8N_2O_2$, monoclinic, $P12_1/n1$ (No. 14), $a = 4.836(1)$ Å, $b = 13.031(1)$ Å, $c = 32.408(6)$ Å, $\beta = 92.82(2)°$, $V = 2039.6$ Å^3, $Z = 8$, $R_{gt}(F) = 0.051$, $wR_{ref}(F^2) = 0.140$, $T = 291$ K.

Source of material

The title compound was prepared by the following reaction: 1.52 g vaniline was dissolved in 7.5 ml 95% ethanol and 0.66 g malonodinitrile was dissolved in 5 ml 95% ethanol. Both solutions were mixed by stirring and 2-3 drops of piperidine were added as a catalyst. The mixture was stirred for 10 minutes. Yellow crystals were obtained from the resulting red solution by slow evaporation at room temperature. The product was isolated and recrystallized from ethanol (mp 407 K – 408 K). The crystals were obtained by multiple recrystallization from anhydrous ethanol.

Experimental details

For the oxygen atoms and the methyl groups, hydrogen atoms were placed in calculated positions with U_{iso} constrained to be $1.5 \times U_{eq}$ of the carrier atom, and for the remaining hydrogen atoms $1.2 \times U_{eq}$ of the corresponding carrier atom.

Discussion

In the course of our investigations on organic nonlinear optical (NLO) materials we now determined the crystal structure of the title compound. The asymmetric unit of the crystal contains two formula units $C_{11}H_8N_2O_2$.The molecules are nearly planar and coplanar with distances and angles in the normal range and are linked via a O—H···N hydrogen bonds (d(O2—H2···N2′) = 2.849(3) Å, ∠O–H···N = 149°).

Table 1. Data collection and handling.

Crystal:	yellow block, size 0.25 × 0.30 × 0.50 mm
Wavelength:	Mo K_α radiation (0.71073 Å)
μ:	0.92 cm^{-1}
Diffractometer, scan mode:	Nonius MACH3, 2θ/ω
$2\theta_{max}$:	50.16°
$N(hkl)_{measured}$, $N(hkl)_{unique}$:	7995, 3634
Criterion for I_{obs}, $N(hkl)_{gt}$:	$I_{obs} > 2\ \sigma(I_{obs})$, 2607
$N(param)_{refined}$:	275
Programs:	CORINC [1], SHELXS-97 [2], SHELXTL-Plus [3], SHELXL-97 [4], PARST95 [5]

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

Atom	Site	x	y	z	U_{iso}
H(2)	4e	0.2836	0.2136	0.1365	0.099
H(2A)	4e	–0.3026	0.1921	0.0308	0.059
H(5)	4e	–0.0753	0.4304	0.1332	0.069
H(6)	4e	–0.4278	0.4651	0.0846	0.067
H(7A)	4e	–0.1473	0.0289	0.0578	0.128
H(7B)	4e	0.1652	–0.0036	0.0598	0.128
H(7C)	4e	0.0572	0.0778	0.0273	0.128
H(8)	4e	–0.7088	0.4239	0.0292	0.063
H(2′)	4e	1.9389	–0.3939	0.2088	0.116
H(2′A)	4e	1.4092	–0.1281	0.2264	0.057
H(5′)	4e	1.5094	–0.3781	0.1267	0.089
H(6′)	4e	1.1815	–0.2524	0.1164	0.079
H(7′A)	4e	1.8049	–0.1239	0.2678	0.110
H(7′B)	4e	1.9125	–0.2057	0.3002	0.110
H(7′C)	4e	1.5942	–0.1946	0.2896	0.110
H(8′)	4e	0.9410	–0.1118	0.1366	0.062

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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)	4*e*	0.0999(4)	0.1316(2)	0.08350(6)	0.058(1)	0.058(1)	0.073(1)	0.017(1)	–0.012(1)	–0.008(1)
O(2)	4*e*	0.2152(4)	0.2698(2)	0.14105(6)	0.060(1)	0.072(1)	0.064(1)	0.010(1)	–0.019(1)	–0.006(1)
N(1)	4*e*	–0.5497(7)	0.1507(2)	–0.0441(1)	0.115(3)	0.075(2)	0.100(2)	0.017(2)	–0.018(2)	–0.023(2)
N(2)	4*e*	–1.1303(6)	0.4003(2)	–0.05385(8)	0.064(2)	0.078(2)	0.069(2)	0.011(1)	–0.018(1)	–0.001(1)
C(1)	4*e*	–0.4017(5)	0.3336(2)	0.05217(8)	0.043(2)	0.051(2)	0.050(2)	0.004(1)	–0.001(1)	0.001(1)
C(2)	4*e*	–0.2584(5)	0.2400(2)	0.05136(8)	0.046(2)	0.051(2)	0.050(2)	0.003(1)	–0.000(1)	–0.005(1)
C(3)	4*e*	–0.0538(5)	0.2192(2)	0.08082(8)	0.041(1)	0.051(2)	0.053(2)	0.006(1)	0.003(1)	0.003(1)
C(4)	4*e*	0.0151(5)	0.2904(2)	0.11178(8)	0.043(2)	0.057(2)	0.050(2)	0.001(1)	–0.003(1)	0.003(1)
C(5)	4*e*	–0.1226(6)	0.3824(2)	0.11284(9)	0.061(2)	0.056(2)	0.056(2)	0.004(1)	–0.006(1)	–0.009(1)
C(6)	4*e*	–0.3320(6)	0.4034(2)	0.08346(9)	0.056(2)	0.052(2)	0.058(2)	0.008(1)	–0.003(1)	–0.003(1)
C(7)	4*e*	0.0389(8)	0.0523(3)	0.0548(1)	0.101(3)	0.059(2)	0.093(3)	0.025(2)	–0.013(2)	–0.015(2)
C(8)	4*e*	–0.6206(6)	0.3629(2)	0.02285(9)	0.047(2)	0.054(2)	0.056(2)	0.007(1)	–0.000(1)	0.002(1)
C(9)	4*e*	–0.7206(5)	0.3178(2)	–0.01223(9)	0.042(1)	0.056(2)	0.053(2)	0.001(1)	0.000(1)	0.002(1)
C(10)	4*e*	–0.6217(6)	0.2248(2)	–0.0294(1)	0.061(2)	0.063(2)	0.060(2)	0.002(2)	–0.010(2)	–0.002(2)
C(11)	4*e*	–0.9488(6)	0.3639(2)	–0.03556(9)	0.053(2)	0.060(2)	0.054(2)	–0.001(1)	–0.005(1)	–0.001(1)
O(1′)	4*e*	1.7940(4)	–0.2649(2)	0.24662(6)	0.060(1)	0.069(1)	0.057(1)	0.018(1)	–0.015(1)	–0.001(1)
O(2′)	4*e*	1.8365(5)	–0.4067(2)	0.18859(7)	0.081(2)	0.067(1)	0.081(2)	0.027(1)	–0.015(1)	–0.010(1)
N(1′)	4*e*	1.2497(6)	0.0577(2)	0.24513(9)	0.071(2)	0.068(2)	0.078(2)	0.007(1)	–0.012(2)	–0.016(2)
N(2′)	4*e*	0.5893(5)	0.1018(2)	0.15120(9)	0.055(2)	0.072(2)	0.081(2)	0.012(1)	–0.008(1)	0.010(1)
C(1′)	4*e*	1.2593(5)	–0.1755(2)	0.16954(8)	0.045(2)	0.053(2)	0.050(2)	0.000(1)	–0.005(1)	–0.001(1)
C(2′)	4*e*	1.4285(5)	–0.1783(2)	0.20631(8)	0.045(1)	0.049(1)	0.049(2)	0.001(1)	–0.003(1)	–0.001(1)
C(3′)	4*e*	1.6203(5)	–0.2545(2)	0.21259(8)	0.046(2)	0.053(2)	0.046(2)	–0.001(1)	–0.004(1)	0.003(1)
C(4′)	4*e*	1.6520(6)	–0.3304(2)	0.18274(9)	0.056(2)	0.053(2)	0.062(2)	0.009(1)	–0.002(1)	–0.001(1)
C(5′)	4*e*	1.4882(7)	–0.3282(3)	0.1468(1)	0.086(2)	0.070(2)	0.064(2)	0.022(2)	–0.016(2)	–0.020(2)
C(6′)	4*e*	1.2932(7)	–0.2523(2)	0.14056(9)	0.068(2)	0.071(2)	0.056(2)	0.009(2)	–0.019(2)	–0.010(2)
C(7′)	4*e*	1.7749(7)	–0.1913(3)	0.27866(9)	0.085(2)	0.078(2)	0.054(2)	0.007(2)	–0.023(2)	–0.004(2)
C(8′)	4*e*	1.0522(6)	–0.0987(2)	0.16025(8)	0.045(2)	0.060(2)	0.050(2)	–0.002(1)	–0.007(1)	0.002(1)
C(9′)	4*e*	0.9911(5)	–0.0110(2)	0.17981(8)	0.038(1)	0.053(2)	0.055(2)	–0.001(1)	–0.003(1)	0.008(1)
C(10′)	4*e*	1.1348(6)	0.0272(2)	0.21623(9)	0.045(2)	0.051(2)	0.065(2)	0.003(1)	0.000(1)	0.001(1)
C(11′)	4*e*	0.7673(6)	0.0520(2)	0.16365(9)	0.043(2)	0.058(2)	0.060(2)	–0.001(1)	0.001(1)	0.008(1)

References

1. Dräger, M.; Gattow, G.: CORINC. Acta Chem. Scand. **25** (1971) 761-762; revised by Wiehl, L.; Schollmeyer, D.: Universität Mainz, Germany 1994.
2. Sheldrick, G. M.: Phase Annealing in SHELX-90: Direct Methods for Larger Structures. Acta Crystallogr. A**46** (1990) 467–473.
3. Sheldrick, G. M.: SHELXTL-Plus. Release 4.1 Siemens Analytical X-ray Instruments Inc., Madison, Wisconsin, USA 1991.
4. Sheldrick, G. M.: SHELXL-97. Program for the Refinement of Crystal Structures, University of Göttingen, Germany 1997.
5. Nardelli, M.: PARST95 – an update to PARST: A system of Fortran routines for calculating molecular structure parameters from the results of crystal structure analyses. J. Appl. Crystallogr. **28** (1995) 659.