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Crystal structure of 2,2′-[(1,4-butylene)dioxybis(nitrilomethylidyne)]-dinaphthol, $C_{26}H_{24}N_2O_4$

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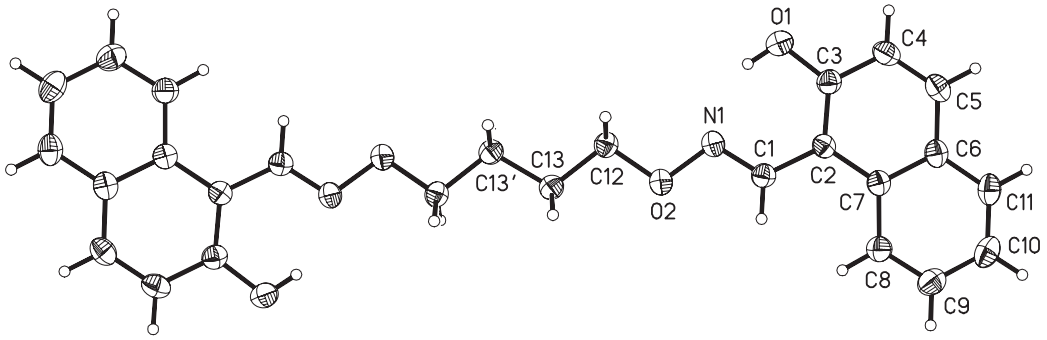

Abstract

$C_{26}H_{24}N_2O_4$, monoclinic, $P12_1/c1$ (no. 14), a = 5.611(2) Å, b = 11.723(2) Å, c = 16.636(3) Å, β = 95.432(2)°, V = 1089.4 Å^3, Z = 2, $R_{gt}(F)$ = 0.048, $wR_{ref}(F^2)$ = 0.153, T = 298 K.

Source of material

To an ethanol solution (10 ml) of 2-hydroxy-1-naphthaldehyde (349.5 mg, 2.01 mmol) was added an ethanol solution (3 ml) of 1,4-bis(aminooxy)butane (120.12 mg, 1.0 mmol). After the solution had been stirred at 55 °C for 4 h, the mixture was filtered, washed successively with ethanol and ethanol/hexane (1:4), respectively. The isolated compound was dried under reduced pressure and purified with recrystallization from ethanol to yield 192.8 mg of pale-brown crystalline solid (yield 45 %, m.p. 142-143 °C). Pale-brown single crystals were obtained by slow evaporation from an acetone solution at room temperature.
Elemental analysis – found: C, 72.85 %; H, 5.42 %; N, 6.73 %; calc. for $C_{26}H_{24}N_2O_4$: C, 72.88 %; H, 5.65 %; N, 6.54 %.

Discussion

Recently, a great deal of attention has focused on a series of *N*,*N*′-bis(salicylidene)ethylenediamine (salen) and its derivatives [1,2], because these compounds easily form metallosalen complexes and some of them have excellent catalytic activities for epoxidation, aziridination, etc. [3,4]. In addition, they are also used as models of reaction centers of metalloenzymes [5,6], and nonlinear optical materials [7]. Moreover, in biological and artificial systems allosteric regulation is effective in controlling molecular functions, such as molecular recognition and biological activity [8]. To tune or improve such functions, chemical modifications of a basic salen skeleton is very interesting and important [9-12]. So the introduction of some functional groups or substitution of some parts with appropriate ones are effective and inevitable. Some of such compounds have two salicylidene units, but the linkage was limited to an alkylene (–CH=N–C–X–C–N=CH–) group due to the ease of preparation. If an *O*-alkyloxime moiety (–CH=N–O–$(CH_2)_n$–O–N=CH–) is used instead of (–CH=N–C–*X*–C–N=CH–) group, the large electronegativity of oxygen atoms is expected to affect strongly the electronic properties of N_2O_2 coordination sphere, which can lead to different and novel properties and structures of the resulted complexes. We have recently reported some *O*-alkyloxime derivatives of salen, such as 4,4′-dibromo-2,2′-[ethylenedioxybis(nitrilomethylidyne)]-diphenol [13] and the crystal structure of 4,4′-dibromo-2,2′-[(1,3-propylene)dioxybis(nitrilomethylidyne)]diphenol [14], which show high stability under the conditions where the imine derivatives suffer metathesis reaction of the C=N bonds.
The crystal structure of the title compound is built up by only the $C_{26}H_{24}N_2O_4$ molecules, in which all bond lengths are in normal ranges. The X-ray diffraction analysis revealed all-conformation of the (–CH=N–O–$(CH_2)_4$–O–N=CH–) bridge, which resulted in a planar structure with two nitrilomethylidyne units apart from each other. The IR spectrum of the compound shows a C=N stretching band at 1624 cm^{-1}, but no C=O band around 1637 cm^{-1} which is observed for 2-hydroxy-1-naphthaldehyde. The title compound is sufficiently stable to resist scrambling of the C=N bonds. This may be ascribed to lower reactivity of the oxime C=N bonds toward nucleophiles.

Table 1. Data collection and handling.

Crystal:	pale-brown block, size 0.50 × 0.53 × 0.65 mm
Wavelength:	Mo K_α radiation (0.71073 Å)
μ:	0.89 cm^{-1}
Diffractometer, scan mode:	Bruker SMART CCD, φ/ω
$2\theta_{max}$:	50°
$N(hkl)_{measured}$, $N(hkl)_{unique}$:	5499, 1927
Criterion for I_{obs}, $N(hkl)_{gt}$:	$I_{obs} > 2\,\sigma(I_{obs})$, 1211
$N(param)_{refined}$:	145
Programs:	SHELXS-97 [15], SHELXL-97 [16]

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

Atom	Site	*x*	*y*	*z*	U_{iso}
H(1)	4*e*	1.1972	0.8157	1.5468	0.100
H(1A)	4*e*	1.2359	0.6307	1.7001	0.056
H(4)	4*e*	1.6633	0.9853	1.5794	0.069
H(5)	4*e*	1.9510	0.9738	1.6845	0.069
H(8)	4*e*	1.4947	0.6255	1.7968	0.067
H(9)	4*e*	1.7918	0.6162	1.8991	0.077
H(10)	4*e*	2.1167	0.7398	1.9036	0.079
H(11)	4*e*	2.1265	0.8795	1.8088	0.071
H(12A)	4*e*	0.8461	0.5936	1.4829	0.058
H(12B)	4*e*	0.6742	0.6705	1.5292	0.058
H(13A)	4*e*	0.5180	0.5057	1.5852	0.057
H(13B)	4*e*	0.6851	0.4303	1.5365	0.057

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*e*	1.0950(3)	0.6848(1)	1.5982(1)	0.049(1)	0.047(1)	0.051(1)	–0.0038(8)	0.0035(8)	–0.0048(8)
O(1)	4*e*	1.2990(3)	0.8659(1)	1.54699(9)	0.078(1)	0.064(1)	0.0561(9)	–0.0067(8)	–0.0068(8)	0.0108(7)
O(2)	4*e*	0.9297(3)	0.5962(1)	1.60133(8)	0.056(1)	0.0577(9)	0.0540(9)	–0.0124(7)	–0.0030(7)	0.0014(7)
C(1)	4*e*	1.2485(4)	0.6849(2)	1.6599(1)	0.051(1)	0.046(1)	0.043(1)	0.001(1)	0.006(1)	–0.0018(9)
C(2)	4*e*	1.4421(4)	0.7674(2)	1.6691(1)	0.047(1)	0.039(1)	0.042(1)	0.0009(9)	0.0080(9)	–0.0054(8)
C(3)	4*e*	1.4608(4)	0.8526(2)	1.6122(1)	0.057(1)	0.046(1)	0.043(1)	0.002(1)	0.004(1)	–0.0031(9)
C(4)	4*e*	1.6534(4)	0.9293(2)	1.6185(1)	0.071(2)	0.045(1)	0.057(1)	–0.005(1)	0.014(1)	0.002(1)
C(5)	4*e*	1.8249(4)	0.9220(2)	1.6811(1)	0.061(2)	0.048(1)	0.065(2)	–0.011(1)	0.014(1)	–0.008(1)
C(6)	4*e*	1.8172(4)	0.8377(2)	1.7416(1)	0.049(1)	0.047(1)	0.049(1)	0.001(1)	0.010(1)	–0.0142(9)
C(7)	4*e*	1.6214(4)	0.7603(2)	1.7361(1)	0.047(1)	0.041(1)	0.043(1)	0.0039(9)	0.0087(9)	–0.0087(8)
C(8)	4*e*	1.6207(4)	0.6772(2)	1.7980(1)	0.064(2)	0.052(1)	0.052(1)	–0.002(1)	0.002(1)	–0.000(1)
C(9)	4*e*	1.7990(4)	0.6711(2)	1.8591(1)	0.076(2)	0.064(2)	0.051(1)	0.006(1)	0.001(1)	0.000(1)
C(10)	4*e*	1.9930(5)	0.7462(2)	1.8625(1)	0.068(2)	0.075(2)	0.053(1)	0.009(1)	–0.007(1)	–0.015(1)
C(11)	4*e*	1.9994(4)	0.8283(2)	1.8056(1)	0.054(1)	0.065(2)	0.058(1)	–0.005(1)	0.004(1)	–0.019(1)
C(12)	4*e*	0.7625(3)	0.5993(2)	1.5312(1)	0.045(1)	0.050(1)	0.050(1)	0.0021(9)	0.002(1)	–0.0014(9)
C(13)	4*e*	0.5948(3)	0.5010(2)	1.5354(1)	0.045(1)	0.049(1)	0.049(1)	0.0042(9)	0.0043(9)	0.0001(9)

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