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Synthesis and Supramolecular Structure of 5,5'-Dihydroxy-2,2'-[1,1'-(propane-1,3-diyldioxydinitrilo)dimethylidyne]diphenol

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The compound, 5,5'-dihydroxy-2,2'-[1,1'-(propane-1,3-diyldioxydinitrilo)dimethylidyne]diphenol with the molecular formula $C_{17}H_{18}N_2O_6$, assumes a *cis* conguration in which two phenoloxime moieties adopts an extended form. The dihedral angle between the two benzene rings of the molecule is 1.83(3)º. There are two fairly strong intramolecular O3-H3···N1 and O5-H5···N2 hydrogen bonds. Each compound molecule links three other neighboring molecules through two pairs of intermolecular O6-H6···O3 and O4-H4···O6 hydrogen-bonding interactions into an infinite wave-like 2D-layer supramolecular structure parallel to the *ab* crystallographic plane.

Key Words: Bisoxime compound, Synthesis, Supramolecular structure.

INTRODUCTION

Salen and its derivatives are very important compounds as versatile ligands, they can accommodate one, or more metal centers to form complexes with interesting properties and applications[1]. These complexes have been found to possess effective activities including catalysis[2], medical imaging[3], inorganic biochemistry[4], thin films[5] and optical materials[6]. Herein, we report on the synthesis and crystal structure of Salen-type bisoxime compound, 5,5'-dihydroxy-2,2'-[1,1'-(propane-1,3-diyldioxydinitrilo)dimethylidyne]diphenol.

EXPERIMENTAL

2,4-Dihydroxybenzaldehyde was purchased and used without further purification. 1,3-*Bis*(aminooxy)propane was synthesized according to an analogous method reported earlier[7]. The other reagents and solvents were analytical grade reagents from Tianjin Chemical Reagent Factory. C, H and N analyses were carried out with a GmbH VariuoEL V3.00 automatic elemental analyzer. X-Ray single crystal structure was determined on a Bruker Smart 1000 CCD area detector. Melting points were measured by the use of a microscopic melting point apparatus made in Beijing Taike Instrument Limited Company and the thermometer was uncorrected.

General procedure: 5,5'-Dihydroxy-2,2'-[1,1'-(propane-1,3-diyldioxydinitrilo)dimethylidyne]diphenol was synthesized according to an analogous method reported earlier[8]. To an ethanol solution (10 mL) of 2,4-dihydroxybenzaldehyde (284.9 mg, 2.06 mmol) was added an ethanol solution (5 mL) of 1,3-*bis*(aminooxy)propane (111.68 mg, 0.95 mmol). The reaction mixture was stirred at 328-333 K for 6 h. The formed precipitate was separated by filtration and washed successively with ethanol and *n*-hexane. The product was dried under vacuum to yield 195.3 mg of the title compound. Yield 59.4 %. m.p. 462.5-463.5 K. Anal. calcd. (%) for $C_{17}H_{18}N_2O_6$: C, 58.96; H, 5.24; N, 8.09. Found (%): C, 59.12; H, 5.33; N, 7.93.

A solution of the title compound (5.8 mg, 0.015 mmol) in acetone (2 mL) was added slowly to deionized water (2 mL), the mixture was allowed to stand at room temperature for about one week. When the acetone was partially evaporated, several colorless needle-like single crystals suitable for X-ray crystallographic analysis were obtained.

X-Ray structure determination: The single crystal of the title compound, with approximate dimensions of 0.38 mm × 0.11 mm × 0.07 mm was placed on a Bruker Smart 1000 diffractmeter equipped with Apex CCD area detector. The diffraction data were collected using a graphite monochromated MoK_α radition (λ = 0.71073 Å) at 293(2) K. The structure was solved by using the program SHELXS-97 and Fourier difference techniques and refined by full-matrix least-squares method on F^2 using SHELXL-97. Details of the data collection and refinements of the title compound are given in Table-1. The non-hydrogen atoms were refined anisotropically. Hydrogen atoms were added theoretically. CCDC: 878707.

RESULTS AND DISCUSSION

X-Ray crystallographic analysis revealed the crystal structure of the title compound (Fig. 1). Selected bond distances

TABLE- 1
CRYSTAL DATA AND STRUCTURE REFINEMENT FOR THE TITLE COMPOUND

Empirical formula	$C_{17}H_{18}N_2O_6$
Formula weight	346.33
Temperature (K)	293(2)
Wavelength (Å)	0.71073
Crystal system	Monoclinic
Space group	P 2_1/c
Cell dimensions, (Å, deg)	a = 11.2799(10), b = 10.6845(7), c = 14.1488(14), β = 106.080(1)
Volume (Å^3)	1638.5(2)
Z	4
Density (calculated) (mg/m^3)	1.404
Absorption coefficient (mm^{-1})	0.108
$F_{(000)}$	728.0
Index ranges	-13 ≤ h ≤ 12, -10 ≤ k ≤12, -16 ≤1 ≤ 15
Reflections collected	5451/2897 [$R_{(int)}$ = 0.0286]
Independent reflections	633
Data/restraints/parameters	2897/0/226
Goodness of fit indicator	0.993
R [I > 2σ(I)]	R_1 = 0.0528, wR_2 = 0.1094
Largest diff. peak and hole (e Å^{-3})	0.144 and -0.197

and angles are listed in Table-2. The single crystal structure of the title compound is built up by only the $C_{17}H_{18}N_2O_6$ molecule. The title compound is a typical Salen-type bisoxime derivative with normal geometric parameters. The molecule adopts a *cis* conguration in which two phenoloxime moieties adopts an extended form, where the oximes and phenolic alcohols lie in *cis* positions relative to the C2 atom in the N-O-CH_2-CH_2-CH_2-O-N linkage, which isn't similar to what is observed in our previously reported Salen-type bisoxime compounds[9-12]. The dihedral angle formed by the two benzene rings in each molecule is *ca*. 1.83(3)º.

In the crystal structure, two intramolecular O3-H3···N1 and O5-H5···N2 hydrogen bonds (Table-3) involving the hydroxyl groups and oxime N atoms generate S(6) ring motifs in each molecule[13-16]. Moreover, each compound molecule links three other neighboring molecules through two pairs of intermolecular O6-H6···O3 and O4-H4···O6 hydrogen-bonding interactions into an infinite wave-like 2D-layer supramolecular structure parallel to the *ab* crystallographic plane (Fig. 2).

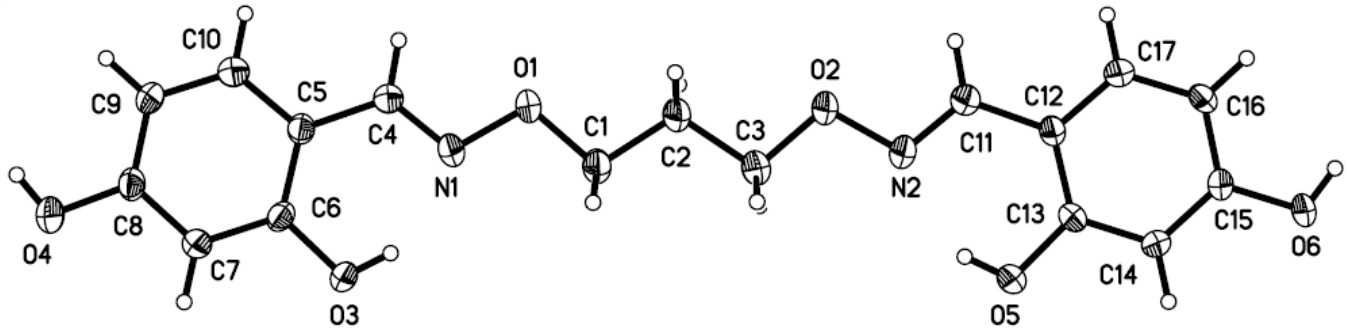

Fig. 1. Molecule structure of the title compound with atom numbering scheme. Displacement ellipsoids for non-H atoms are drawn at the 30 % probability level

TABLE- 2
SELECTED BOND DISTANCES (Å) AND ANGLES (º) FOR THE TITLE COMPOUND

Bond	Lengths	Bond	Lengths	Bond	Lengths
N(1)-C(4)	1.227(3)	O(6)-C(15)	1.379(3)	C(9)-C(10)	1.375(3)
N(1)-O(1)	1.399(2)	C(1)-C(2)	1.502(3)	C(11)-C(12)	1.454(3)
N(2)-C(11)	1.276(3)	C(2)-C(3)	1.509(3)	C(12)-C(17)	1.391(3)
N(2)-O(2)	1.403(2)	C(4)-C(5)	1.453(3)	C(12)-C(13)	1.394(3)
O(1)-C(1)	1.431(3)	C(5)-C(10)	1.391(3)	C(13)-C(14)	1.386(3)
O(2)-C(3)	1.427(3)	C(5)-C(6)	1.402(3)	C(14)-C(15)	1.380(3)
O(3)-C(6)	1.371(3)	C(6)-C(7)	1.379(3)	C(15)-C(16)	1.373(3)
O(4)-C(8)	1.373(3)	C(7)-C(8)	1.383(3)	C(16)-C(16)	1.379(3)
O(5)-C(13)	1.360(3)	C(8)-C(9)	1.377(3)	–	–
Bond	**Angles**	**Bond**	**Angles**	**Bond**	**Angles**
C(4)-N(1)-O(1)	111.1(2)	O(3)-C(6)-C(7)	118.0(2)	C(17)-C(12)-C(11)	119.9(2)
C(11)-N(2)-O(2)	111.1(2)	O(3)-C(6)-C(5)	120.8(2)	C(13)-C(12)-C(11)	122.6(2)
N(1)-O(1)-C(1)	109.16(19)	C(7)-C(6)-C(5)	121.3(2)	O(5)-C(13)-C(14)	117.2(2)
N(2)-O(2)-C(3)	109.85(19)	C(6)-C(7)-C(8)	119.5(2)	O(5)-C(13)-C(12)	121.9(2)
O(1)-C(1)-C(2)	105.9(2)	O(4)-C(8)-C(9)	122.1(2)	C(14)-C(13)-C(12)	120.8(2)
C(1)-C(2)-C(3)	113.2(2)	O(4)-C(8)-C(7)	117.3(2)	C(15)-C(14)-C(13)	119.7(2)
O(2)-C(3)-C(2)	105.3(2)	C(9)-C(8)-C(7)	120.6(2)	C(16)-C(15)-O(6)	117.6(2)
N(1)-C(4)-C(5)	121.6(3)	C(10)-C(9)-C(8)	119.4(2)	C(16)-C(15)-C(14)	120.8(2)
C(10)-C(5)-C(6)	117.3(2)	C(9)-C(10)-C(5)	122.0(3)	O(6)-C(15)-C(14)	121.5(2)
C(10)-C(5)-C(4)	120.5(2)	N(2)-C(11)-C(12)	121.2(2)	C(15)-C(16)-C(17)	118.9(2)
C(6)-C(5)-C(4)	122.3(2)	C(17)-C(12)-C(13)	117.5(2)	C(16)-C(17)-C(12)	121.1(2)

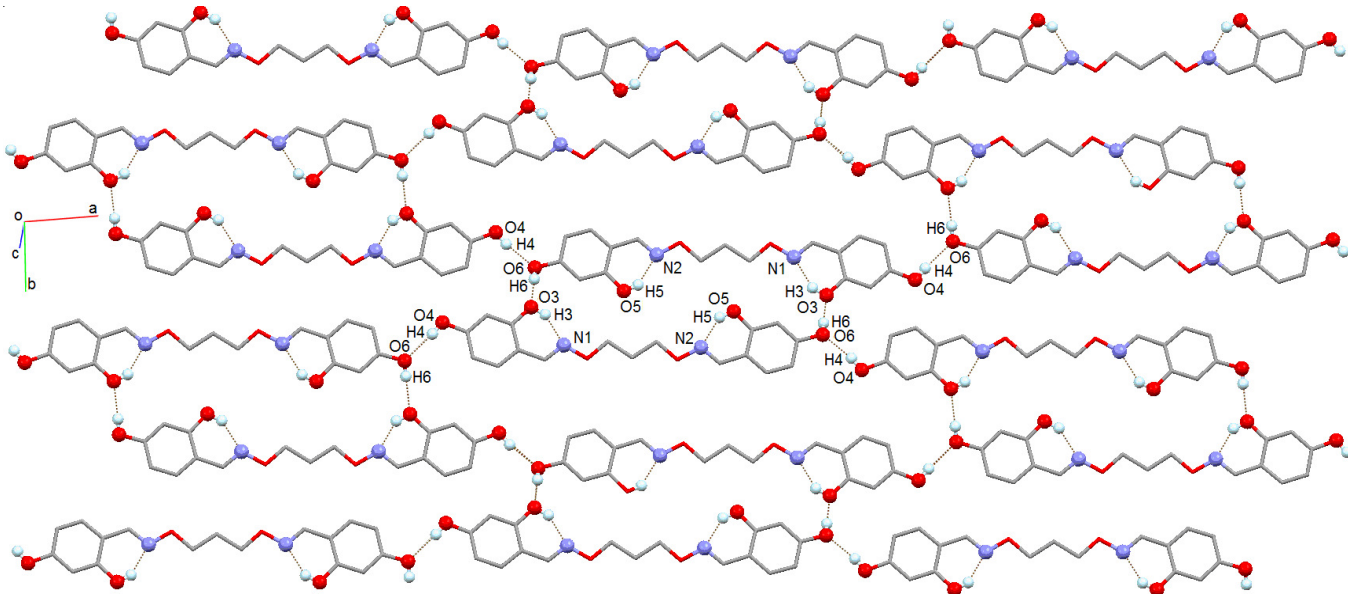

Fig. 2. Part of 2D wave-like supramolecular structure of the title compounds parallel to the ab crystallographic plane. Hydrogen atoms, except those forming hydrogen bonds, are omitted for clarity

TABLE- 3
HYDROGEN BONDS [Å, °] FOR THE TITLE COMPOUND

D-H···A	d(D-H)	d(H···A)	∠DHA	d(D···A)
O3-H3···N1	0.82	1.88	147	2.605(3)
O5-H5…N2	0.82	1.91	146	2.628(3)
O4-H4···O6	0.82	1.99	167	2.796(2)
O6-H6···O3	0.82	1.99	178	2.812(2)

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