Asian Journal of Chemistry; Vol. 25, No. 11 (2013), 6467-6468

ASIAN JOURNAL OF CHEMISTRY

http://dx.doi.org/10.14233/ajchem.2013.14313

NOTE

Synthesis and Supramolecular Structure of 4,4',6,6'-Tetrachloro-2,2'-[(decane-1,10-diyldioxy)*bis*(nitrilomethylidyne)]diphenol

WEN-KUI DONG*, XIANG LI, JUN-JIANG WU, YANG ZHANG and XIAO-LU TANG

School of Chemical and Biological Engineering, Lanzhou Jiaotong University, Lanzhou 730070, P.R. China

*Corresponding author: E-mail: dongwk@126.com

(*Received*: 16 July 2012; *Accepted*: 14 May 2013) AJC-13500

A new salen-type bisoxime compound, 4,4',6,6'-tetrachloro-2,2'-[(decane-1,10-diyldioxy)*bis*(nitrilomethylidyne)]diphenol, has been synthesized and characterized structurally. In each molecule, there is a *trans* configuration with respect to the methylidene unit. The two phenyl rings in each molecule are paralle to each other, with a perpendicular interplanar spacing of *ca.* 6.583(2) Å. In the crystal structure, each molecule links other two molecules into finite 2D supramolecular structure by two pairs of intermolecular C10-H10···Cl2 hydrogen bonds.

Key Words: Salen-type bisoxime, Synthesis, Supramolecular structure.

Salen-type compound and its derivatives have played a significant role in the development of the coordination chemistry as they are one of the most important mixed-donor ligands in the modern coordination chemistry and these compounds have caused growing concern because of their strong coordination capability and important properties, such as diverse biological activities[1-3], synthetic and catalytic activities[4,5], magnetic properties[6] and supramolecular architectures[7]. In order to extend our work on supramolecular interactions of salen-type bisoxime compound, we reported the synthesis and crystal structure of the title compound.

3,5-Dichloro-2-hydroxybenzaldehyde was purchased from Alfa Aesar and used without further purification. 1,10-*Bis*-(aminooxy)decane was synthesized according to an analogous method reported earlier[8]. 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.

4,4',6,6'-Tetrachloro-2,2'-[(decane-1,10-diyldioxy)*bis*-(nitrilomethylidyne)]diphenol was synthesized according to an analogous method reported earlier[9,10]. To an ethanol solution (5 mL) of 3,5-dichloro-2-hydroxybenzaldehyde (38.2 mg, 0.2 mmol) was added an ethanol solution (5 mL) of 1,10-*bis*-(aminooxy)decane (20.4 mg, 0.1 mmol). The reaction mixture was stirred at 328 K for 4 h. The formed precipitate was separated by filtrationand washed successively with ethanol and *n*-hexane. The product was dried under vacuum to yield 32.9 mg of the title compound. Yield, 59.8 %. m.p. 364-365 K. anal. calcd. for $C_{24}H_{28}C_{l4}N_2O_4$: C, 52.38; H, 5.13; N, 5.09; Found: C, 52.71; H, 5.20; N, 4.97.

Colourless block-shaped single crystals suitable for X-ray diffraction studies were obtained after two weeks by slow evaporation from acetone/acetonitrile (1:4) solution of the title compound.

X-Ray structure determination: The single crystal of the title compound, with approximate dimensions of 0.43 × 0.22 × 0.09 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 298(2) K. The structure was solved by using the program SHELXS-97 and Fourier difference techniquesand 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: 712163.

X-ray crystallographic analysis revealed the crystal structure of the title compound. And the structure is shown in Fig. 1. Selected bond distances and angles are listed in Table-2. The single crystal structure of the title compound is only built up by the $C_{24}H_{28}N_2O_4Cl_4$ molecule. In each molecule, there is a *trans* configuration with respect to the methylidene unit. The two phenyl rings in each molecule are parallel to each other, with C1-O1-N1-C7 torsion angles of -179.2 (8)º and a perpendicular

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

Bond	Lengths	Bond	Lengths	Bond	Lengths
N(1)-C(6)	1.272(12)	C(1)-C(2)	1.498(14)	C(7)-C(12)	1.395(14)
N(1)-O(1)	1.405(10)	C(2)-C(3)	1.471(13)	C(7)-C(8)	1.430(13)
O(1)-C(1)	1.433(11)	C(3)-C(4)	1.513(13)	C(8)-C(9)	1.402(14)
O(2)-C(8)	1.368(11)	C(4)-C(5)	1.495(14)	C(9)-C(10)	1.359(14)
C(11)-C(9)	1.731(10)	C(5)-C(5)	1.495(19)	C(10)-C11)	1.401(12)
C(12)-C(11)	1.736(9)	C(6)-C(7)	1.449(14)	C(11)-C(12)	1.338(13)
Bond	Angles	Bond	Angles	Bond	Angles
C(6)-N(1)-O(1)	111.3(8)	C(12)-C(7)-C(8)	116.5(9)	C(8)-C(9)-Cl(1)	119.1(8)
N(1)-O(1)-C(1)	109.7(7)	C(12)-C(7)-C(6)	120.9(9)	C(9)-C(10)-C(11)	119.5(9)
O(1)-C(1)-C(2)	107.3(8)	C(8)-C(7)-C(6)	122.6(9)	C(12)-C(11)-C(10)	120.2(9)
C(3)-C(2)-C(1)	116.2(8)	O(2)-C(8)-C(9)	119.6(8)	C(12)-C(11)-Cl(2)	121.2(7)
C(2)-C(3)-C(4)	114.6(8)	O(2)-C(8)-C(7)	120.9(9)	C(10)-C(11)-Cl(2)	118.6(7)
C(5)-C(4)-C(3)	116.1(8)	C(9)-C(8)-C(7)	119.5(9)	C(11)-C(12)-C(7)	123.2(9)
C(4)-C(5)-C(5)	117.2(11)	C(10)-C(9)-C(8)	121.0(9)		
N(1)-C(6)-C(7)	120.1(9)	C(10)-C(9)-Cl(1)	119.9(8)		

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

Empirical formula	$C_{24}H_{28}N_2O_4Cl_4$
Formula weight	550.28
Temperature (K)	298(2)
Wavelength (Å)	0.71073
Crystal system	Triclinic
Space group	P -1
Cell dimensions, (Å, deg)	a = 4.5804(6), b = 7.9141(11), c = 17.9692(18), α = 84.2770(10), β = 86.437(2), γ = 80.8340(10)
Volume, Å^3	639.15(14)
Z	1
Density (calculated), mg/m^3	1.430
Absorption coefficient, mm^{-1}	0.497
F(000)	286
Index ranges	-5 ≤ h ≤ 5, -9 ≤ k ≤ 7, -16 ≤ l ≤ 21
Reflections collected	2889/2079 [*R*(int) = 0.0529]
Independent reflections	930
Data/restraints/parameters	2079/0/155
Goodness of fit indicator	1.041
R [I > 2σ(I)]	R_1 = 0.0529, wR_2 = 0.1214
Largest diff. peak and hole, e·Å^{-3}	0.803 and -0.583

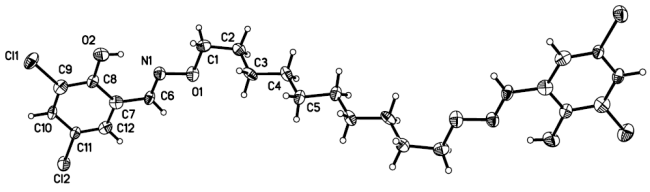

Fig. 1. Molecule structure of the title compound with atom numbering scheme. Displacement ellipsoids for non-H atoms are drawn at the 30 % probability level

interplanar spacing of *ca*. 6.583(2) Å. There are two intramolecular O2-H2···N1 hydrogen bonds between the hydroxyl groups and the oxime N atoms in each molecule (Table-3). In the crystal structure, each molecule links other two mole-cules into finite 2D supramolecular structure by two pairs of intermolecular C10-H10···Cl2 hydrogen bonds (Fig. 2).

TABLE-3
HYDROGEN BOND (Å, °) FOR THE TITLE COMPOUND

D–H···A	d(D–H)	d(H···A)	∠DHA	d(D···A)
O2-H2···N1	0.82	1.89	146	2.609(3)
C10-H10···Cl2	0.93	2.88	166	3.794(3)

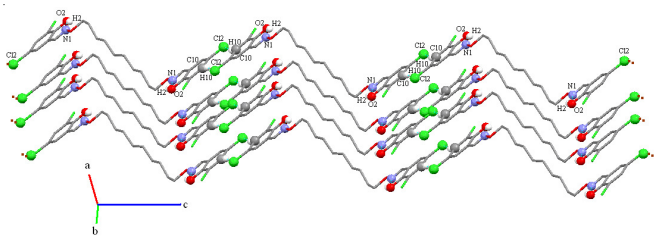

Fig. 2. A perspective view of the intramolecular and intermolecular hydrogen-bond interactions

ACKNOWLEDGEMENTS

This work was supported by the Fundamental Research Funds for the Gansu Province Universities (212086) and the Science and Technology support funds of Gansu Province (212249), which are gratefully acknowledged.

REFERENCES

1. S. Koizumi, M. Nihei, M. Nakano and H. Oshio, *Inorg. Chem.*, **44**, 1208 (2005).
2. C. Boskovic, R. Bircher, P.L.W. Tregenna-Piggott, H.U. Güdel, C. Paulsen, W. Wernsdorfer, A.L. Barra, E. Khatsko, A. Neels and H. Stoeckli-Evans,, *J. Am. Chem. Soc.*, **125**, 14046 (2003).
3. H. Oshio, M. Nihei, S. Koizumi, T. Shiga, H. Nohiri, M. Nakano, N. Shirakawa and M. Akatsu, *J. Am. Chem. Soc.*, **127**, 4568 (2005).
4. S.A. Mohand, A. Levina and J. Muzart, *J. Chem. Res. (S)*, **25**, 2051 (1995).
5. J.E. Campbell, H. Zhou and T.S. Nguyen, *Org. Lett.*, **3**, 2391 (2001).
6. J. Nishijo, C. Okabe, O. Oishi and N. Nishi, *Carbon*, **44**, 2943 (2006).
7. S. Akine, T. Taniguchi, W.K. Dong, S. Masubuchi and T. Nabeshima, *J. Org. Chem.*, **70**, 1704 (2005).
8. W.K. Dong, G. Wang, S.S. Gong, J.F. Tong, Y.X. Sun and X.H. Gao, *Transition Met. Chem.*, **37**, 271 (2012).
9. W.K. Dong, J.Y. Shi, J.K. Zhong, Y.Q. Tian and J.G. Duan, *Chinese J. Inorg. Chem.*, **24**, 10 (2008).
10. W.K. Dong, J.Y. Shi, J.K. Zhong, Y.X. Sun and J.G. Duan, *Struct. Chem.*, **19**, 95 (2008).