

Synthesis and Supramolecular Structure of 1-(4-{{(E)-3,5-Dibromo-2-hydroxybenzylidene}amino}phenyl)ethanone O-benzoyloxime

LI ZHAO*, HONG-XIA GUO, YIN-XIA SUN, WEI-SHENG MENG, CHENG-JUAN YANG and XIN-YIANG ZHANG

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

*Corresponding author: E-mail: zhaoli_72@163.com

(Received: 17 September 2012;

Accepted: 10 May 2013)

AJC-13473

The compound, 1-(4-{{(E)-3,5-dibromo-2-hydroxybenzylidene}amino}phenyl)ethanone O-benzoyloxime, has been synthesized by ((4-amino}phenyl)ethanone O-benzoyloxime and 3,5-dibromosalicylaldehyde in ethanol solution and characterized by X-ray crystallography. There is one fairly strong intramolecular O1-H1...N2 hydrogen bond. In the crystal structure, each compound is linked to two other molecules into an infinite 1D supramolecular chain *via* Br...O interactions [3.354(4) Å].

Key Words: Oxime-type compound, Synthesis, Supramolecular structure.

INTRODUCTION

Oxime-type compounds are a traditional class of chelating ligands widely used in coordination and analytical chemistry¹⁻³. They can accommodate one or more metal centers to form complexes with interesting properties and applications⁴⁻⁶. In the last few years, a large number of oxime-type compounds and their complexes are reported^{7,8}. In present report, the synthesis and crystal structure of 1-(4-{{(E)-3,5-dibromo-2-hydroxybenzylidene}amino}-phenyl)ethanone O-benzoyloxime are reported.

EXPERIMENTAL

4-Aminoacetophenone, O-benzylhydroxylamine, 3,5-dibromosalicylaldehyde were purchased and used without further purification. 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: 1-(4-{{(E)-3,5-Dibromo-2-hydroxybenzylidene}amino}phenyl)-ethanone O-benzoyloxime was synthesized by O-benzylhydroxylamine, 4-aminoacetophenone and 3,5-dibromosalicylaldehyde⁹. To an ethanolic solution (6 mL) of O-benzylhydroxylamine (340.9 mg, 3 mmol) was added an ethanol solution (12 mL) of 4-aminoaceto-

Empirical formula	C ₂₂ H ₁₈ Br ₂ N ₂ O ₂
Formula weight	502.20
Temperature (K)	298(2)
Wavelength (Å)	0.71073
Crystal system	Monoclinic
Space group	P 2 ₁ /c
Cell dimensions, (Å, deg)	a = 17.8560(15), b = 6.5063(8), c = 19.1289(17), α = 90, β = 106.080(1), γ = 90
Volume (Å ³)	2018.8(4)
Z	4
Density (calculated) (mg/m ³)	1.652
Absorption coefficient (mm ⁻¹)	4.036
F ₍₀₀₀₎	1000
Index ranges	-18 ≤ h ≤ 21, -7 ≤ k ≤ 7, -22 ≤ l ≤ 17
Reflections collected	10145/3558 [R _(int) = 0.0630]
Independent reflections	831
Data/restraints/parameters	3558/0/254
Goodness of fit indicator	0.953
R [I > 2σ(I)]	R ₁ = 0.0366, wR ₂ = 0.0488
Largest diff. peak and hole (e Å ⁻³)	0.439 and -0.354

phenone (349.5 mg, 3 mmol) and 3 drops of acetic acid. The reaction of mixture solution was stirred at 338 K for 24 h. The solvent was evaporated under *vacuo*. After cooling to room temperature, the formed precipitate was filtered and washed successively with ethanol and ethanol-water (1:4), respectively. Resulting in 640.6 mg of ((4-amino}phenyl) ethanone

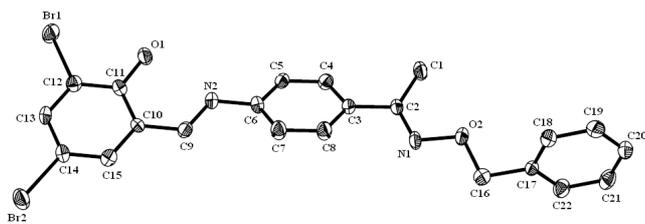


Fig. 1. ORTEP drawing of the title compound with the atom numbering. Displacement ellipsoids for non-H atoms are drawn at the 30 % probability level

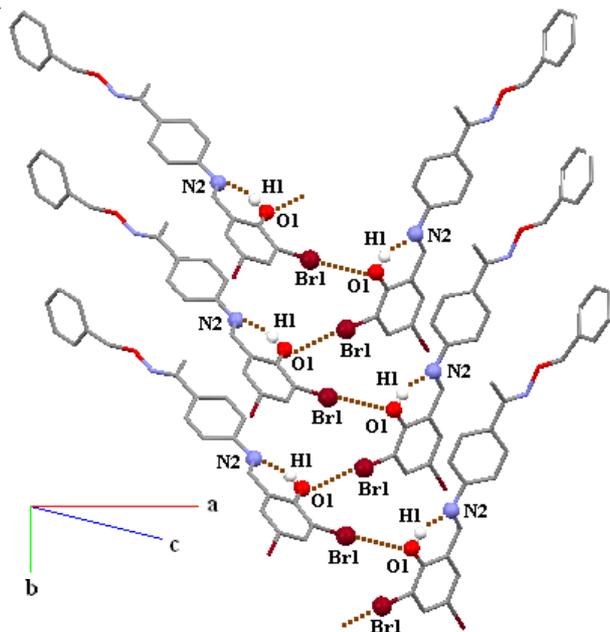


Fig. 2. View of the 1D chain motif of the title compound units along the c axis (hydrogen atoms, except those forming hydrogen bonds, are omitted for clarity)

O-benzyloxime as crystalline solid. Yield, 92.8 %, m.p. 348–350 K. Anal. calcd. (%) for $C_{15}H_{16}N_2O$: C, 74.97; H, 6.71; N, 11.66. Found (%): C, 74.68; H, 6.80; N, 11.52.

To an ethanol solution (4 mL) of (4-amino)phenyl ethanone O-benzyloxime (213.2 mg, 0.89 mmol) was added an ethanolic solution (2 mL) of 3,5-dibromosalicylaldehyde (249.3 mg, 0.89 mmol). The reaction mixture was stirred at 333 K for 18 h. After cooling to room temperature, the formed precipitate was filtered and washed successively with ethanol and ethanol-hexane (1:4), respectively. The product was dried *in vacuo* and purified by recrystallization from ethanol to yield 273.3 mg of solid. Yield, 59.1 %. m.p. 441–442 K. Anal. calcd. (%) for $C_{22}H_{18}N_2O_2Br_2$ (%): C, 52.62; H, 3.61; N, 5.58. Found (%): C, 52.49; H, 3.57; N, 5.53.

X-Ray structure determination: The single crystal of the title compound, with approximate dimensions of 0.37 mm × 0.17 mm × 0.09 mm was placed on a Bruker Smart 1000 diffractometer equipped with Apex CCD area detector. The diffraction data were collected using a graphite monochromated MoK_{α} radiation ($\lambda = 0.71073 \text{ \AA}$) at 298(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: 901313.

RESULTS AND DISCUSSION

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 built up

TABLE-2
SELECTED BOND DISTANCES (\AA) AND ANGLES ($^{\circ}$) FOR THE TITLE COMPOUND

Bond	Lengths	Bond	Lengths	Bond	Lengths
Br(1)-C(12)	1.892(4)	C(3)-C(8)	1.396(5)	C(12)-C(13)	1.375(5)
Br(2)-C(14)	1.884(4)	C(3)-C(4)	1.396(4)	C(13)-C(14)	1.403(4)
N(1)-C(2)	1.276(4)	C(4)-C(5)	1.376(5)	C(14)-C(15)	1.376(5)
N(1)-O(2)	1.428(4)	C(5)-C(6)	1.376(4)	C(16)-C(17)	1.510(5)
N(2)-C(9)	1.280(4)	C(6)-C(7)	1.408(5)	C(17)-C(22)	1.378(5)
N(2)-C(6)	1.417(4)	C(7)-C(8)	1.376(5)	C(17)-C(18)	1.380(5)
O(1)-C(11)	1.350(4)	C(9)-C(10)	1.450(5)	C(18)-C(19)	1.372(5)
O(2)-C(16)	1.435(4)	C(10)-C(15)	1.388(4)	C(19)-C(20)	1.369(5)
C(1)-C(2)	1.488(5)	C(10)-C(11)	1.416(4)	C(20)-C(21)	1.378(5)
C(2)-C(3)	1.487(5)	C(11)-C(12)	1.396(5)	C(21)-C(22)	1.393(5)
Bond	Angles	Bond	Angles	Bond	Angles
C(2)-N(1)-O(2)	110.5(3)	C(8)-C(7)-C(6)	119.5(4)	C(15)-C(14)-Br(2)	121.4(3)
C(9)-N(2)-C(6)	121.9(3)	C(7)-C(8)-C(3)	122.4(3)	C(13)-C(14)-Br(2)	119.1(3)
N(1)-O(2)-C(16)	108.4(3)	N(2)-C(9)-C(10)	122.9(4)	C(14)-C(15)-C(10)	121.6(4)
N(1)-C(2)-C(3)	115.5(4)	C(15)-C(10)-C(11)	118.9(4)	O(2)-C(16)-C(17)	107.4(3)
N(1)-C(2)-C(1)	124.5(4)	C(15)-C(10)-C(9)	120.4(3)	C(22)-C(17)-C(18)	119.1(4)
C(3)-C(2)-C(1)	120.0(3)	C(11)-C(10)-C(9)	120.7(3)	C(22)-C(17)-C(16)	120.3(4)
C(8)-C(3)-C(4)	117.1(4)	O(1)-C(11)-C(12)	119.2(4)	C(18)-C(17)-C(16)	120.5(4)
C(8)-C(3)-C(2)	121.7(3)	O(1)-C(11)-C(10)	121.8(3)	C(19)-C(18)-C(17)	120.9(4)
C(4)-C(3)-C(2)	121.1(4)	C(12)-C(11)-C(10)	119.0(4)	C(20)-C(19)-C(18)	120.6(5)
C(5)-C(4)-C(3)	120.6(4)	C(13)-C(12)-C(11)	121.2(4)	C(19)-C(20)-C(21)	119.0(4)
C(4)-C(5)-C(6)	122.1(3)	C(13)-C(12)-Br(1)	119.6(3)	C(20)-C(21)-C(22)	120.9(4)
C(5)-C(6)-C(7)	118.2(4)	C(11)-C(12)-Br(1)	119.2(3)	C(17)-C(22)-C(21)	119.5(4)
C(5)-C(6)-N(2)	118.0(3)	C(12)-C(13)-C(14)	119.8(4)	—	—
C(7)-C(6)-N(2)	123.6(4)	C(15)-C(14)-C(13)	119.5(4)	—	—

by only the $C_{22}H_{18}N_2O_2Br_2$ molecule, in which all bond lengths are in normal ranges. The title compound is a typical oxime-type derivative with normal geometric parameters. In the crystal structure, the intramolecular O1-H1...N2 hydrogen bonds (Table-3) involving the hydroxyl groups and oxime N atoms generate S (6) ring motifs in each molecule¹⁰⁻¹². Each compound is linked to two other molecules into an infinite 1D supramolecular chain *via* Br...O interactions [3.354(4) Å]¹³.

The 1D chain motif of title compound units along the c-axis is viewed in Fig. 2.

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

D-H...A	d(D-H)	d(H...A)	∠DHA	d(D...A)
O1-H1...N2	0.82	1.88	147	2.612(3)

ACKNOWLEDGEMENTS

This work was supported by the Foundation of Preparative Research of Jin-Chuan Corporation (No. 209125-1104) is gratefully acknowledged.

REFERENCES

1. T. Katsuki, *Coord. Chem. Rev.*, **140**, 189 (1995).
2. W.K. Dong, Y.X. Sun, C.Y. Zhao, X.Y. Dong and L. Xu, *Polyhedron*, **29**, 2087 (2010).
3. W.K. Dong, X.N. He, H.B. Yan, Z.W. Lv, X. Chen, C.Y. Zhao and X.L. Tang, *Polyhedron*, **28**, 1419 (2009).
4. J.P. Costes, F. Dahan and A. Dupuis, *Inorg. Chem.*, **39**, 165 (2000).
5. W.K. Dong, Y.X. Sun, G.H. Liu, L. Li, X.Y. Dong and X.H. Gao, *Z. Anorg. Allg. Chem.*, **638**, 1370 (2012).
6. M.F. Summers, L.G. Marzilli, P.N. Bresciani and L. Randaccio, *J. Am. Chem. Soc.*, **106**, 4478 (1984).
7. W.K. Dong, Y.X. Sun, S.J. Xing, Y. Wang and X.H. Gao, *Z. Naturforsch.*, **67b**, 197 (2012).
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. L. Zhao and S. Weng, *Acta Crystal.*, **E66**, o2473 (2010).
10. W.K. Dong, J.G. Duan, Y.H. Guan, J.Y. Shi and C.Y. Zhao, *Inorg. Chim. Acta*, **362**, 1129 (2009).
11. W.K. Dong, Y.X. Sun, Y.P. Zhang, L. Li, X.N. He and X.L. Tang, *Inorg. Chim. Acta*, **362**, 117 (2009).
12. H.L. Wu, X.C. Huang, J.K. Yuan, F. Kou, F. Jia, B. Liu and Y. Bai, *Z. Naturforsch.*, **66b**, 1049 (2011).
13. Y. Fan, W. You, H.-F. Qian, J.-L. Liu and W. Huang, *Acta Cryst.*, **E64**, o799 (2008).