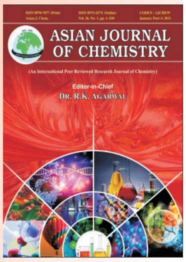
Asian Journal of Chemistry; Vol. 26, No. 16 (2014), 5113-5115

ASIAN JOURNAL OF CHEMISTRY

http://dx.doi.org/10.14233/ajchem.2014.16410



Synthesis and Crystal Structures of 1-(3-{[(*E*)-3,5-Dibromo-2-hydroxybenzylidene]amino}phenyl)ethanone *O*-methyloxime

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Received: 23 September 2013; *Accepted*: 21 February 2014; *Published online*: 28 July 2014; AJC-15642

A Schiff-base mono-oxime compound 1-(3-{[(*E*)-3,5-dibromo-2-hydroxybenzylidene]-amino}phenyl)ethanone *O*-methyloxime with the molecular formula $C_{16}H_{14}N_2O_2Br_2$, has been synthesized and its structure was characterized by X-ray single-crystallography. Within the molecule, the two aromatic rings formed a dihedral angle of 26.55°. In the crystal structure, a strong intramoleclar O-H···N hydrogen bond establish the molecular conformation. An infinite 1D supramolecular chain structure was formed by intermolecular C-H···π hydrogen bonding and π-π stacking interactions.

Keywords: Mono-oxime compound, Synthesis, Crystal Structure.

INTRODUCTION

Much attention has been focused on oxime-based ligands in recent years due to their high stability against imine metathesis reactions[1,2]. The design of new Schiff-base compound has received long-lasting research interest not only because of their appealing structural and topological novelty but also due to their potential wide application in the fields of biochemistry, synthesis and optical materials and conducting polymers. Schiff-base compounds and their derivatives are very important as versatile ligands, properties of interest in materials science, such as their nonlinear optical properties[3,4] and intriguing magnetic properties[5] have been investigated energetically. Previous workers also have been given considerable attention as potential models for biologically important enzymes[6-8]. As an extension of our work, a new Schiff-base oxime-type compound, 1-(3-{[(*E*)-3,5-dibromo-2-hydroxybenzylidene]amino}phenyl)-ethanone *O*-methyloxime, was synthesized and the single crystal structure was also determined.

EXPERIMENTAL

1-(4-Aminophenyl)ethanone, *O*-methylhydroxylamine hydrochloride and 3,5-dibromo-2-hydroxybenzaldehyde were purchased from Aldrich 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. Elemental analysis for the compound was detected by an IRIS ER/S WP-1 ICP atomic emission spectrometer. IR spectra were recorded on a VERTEX70 FT-IR spectrophotometer using KBr pellets. UV-vis absorption spectra were recorded on a Shimadzu UV-2550 spectrometer. 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-(3-Aminophenyl)ethanone *O*-methyloxime and [3-(3,5-dibromo-2-hydroxybenzylideneamino)-phenyl]ethanone *O*-methyloxime were synthesized according to an analogous methods reported earlier[9-11].

Preparation of 1-(3-aminophenyl)ethanone *O*-methyloxime: To an ethanolic solution (6 mL) of 1-(3-aminophenyl)-ethanone (273.6 mg, 2 mmol) was added an ethanol solution (5 mL) of *O*-methylhydroxylamine hydrochloride (169.8 mg, 2 mmol). The mixture solution was stirred at 328 K for 13 h. When cooled to room temperature, the precipitate was filtered and washed successively with ethanol and hexane, respectively. The product was dried under vacuum and purified with recrystallization from ethanol to obtain 289.3 mg of 1-(3-aminophenyl)ethanone *O*-methyl oxime. Yield, 88.2 %, m.p. 448-450 K. Anal. calcd. for C_8H_9NO: C, 65.83; H, 7.37; N, 17.06. Found: C, 65.92; H, 7.34; N, 17.12.

Preparation of 1-(3-{[(*E*)-3,5-dibromo-2-hydroxy benzylidene]amino}phenyl) ethanone *O*-methyloxime: To an ethanol solution (4 mL) of 1-(3-aminophenyl)ethanone *O*-methyl oxime (165.59 mg, 1 mmol) was added an ethanol solution (4 mL) of 3,5-dibromo-2-hydroxybenzaldehyde

(280.94 mg, 1 mmol). After the solution had been stirred at 328 K for 8 h, The formed precipitate was separated by filtration and washed successively with ethanol and ethanol/hexane (1:4), respectively. The product was dried under reduced pressure to obtain 218.02 mg orange-yellow title compound HL. Yield, 51.3 %. m.p. 427-428 K. Anal calcd. (%) for: $C_{16}H_{14}Br_2N_2O_2$: C, 45.10; H, 3.31; N, 6.57. Found (%): C, 45.02; H, 3.36; N, 6.60.

Yellow needle-shaped single crystals suitable for X-ray diffraction studies were obtained after two weeks by slow evaporation from an ethanol/dichloromethane/trichloromethane (2:1:1) solution of the title compound.

X-Ray structure determination: A crystal of dimensions 0.38 mm × 0.09 mm × 0.05 mm was used to determine the crystal structures by X-ray diffraction technique on Bruker SMART CCD area-detector diffractometer using MoK_α radiation (λ = 0.71073 Å, T = 293 (2) K) graphite monochromation radiation. All calculations were performed using the SHELXL-97[12] crystal graphic software package. All the non-hydrogen atoms were refined anisotropically. The hydrogen atoms were located by difference synthesis and refined isotropically. Crystal data and structure refinement for the title compound in Table-1. The non-hydrogen atoms were refined anisotropically. Hydrogen atoms were added theoretically.

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

Empirical formula	$C_{16}H_{14}Br_2N_2O_2$
Crystal colour	Yellow, needle-shaped
Formula weight	426.11
Temperature	293(2) K
Wavelength	0.71073 Å
Crystal system	Monoclinic
Space group	P2(1)/c
Cell dimensions	a = 17.0138 (15) Å, b = 13.3065 (14) Å, c = 7.2209 (6) Å β = 95.7650 (10)
Volume	1626.5 (3) Å^3
Z	4
Density (calculated)	1.740 mg/m^3
Absorption coefficient	4.992 mm^{-1}
F(000)	840
Index ranges	$-20 \leq h \leq 20$, $-15 \leq k \leq 15$, $-8 \leq 1 \leq 8$
Reflections collected	2871
Independent reflections	2871 [R_{int}= 0.0000]
Data/restraints/parameters	2871/0/203
Goodness of fit indicator	1.021
Final R indices [I > 3σ(I)]	R_1 = 0.0718, wR_2 = 0.0819
Largest diff. peak and hole	0.575 and -0.503 e. Å

RESULTS AND DISCUSSION

The crystal structure of the title compound is only built up by the $C_{16}H_{14}N_2O_2Br_2$ molecule, the molecular structure is shown in Fig. 1 and selected bond lengths and angles are listed in Table-2. All bond lengths and angles are in normal ranges. The molecule crystallizes in the monoclinic system, space group $P2_1/c$, with four crystallographically independent molecules in the unit cell. The two benzene rings form a dihedral angle of 26.55°.

In the crystal structure, a strong intramoleclar O-H···N hydrogen bond establish the molecular conformation. And an infinite 1D supramolecular chain structure was formed by intermolecular C-H···π hydrogen bonding and π-π stacking interactions. The weak C-H···π (C_3-C_8) hydrogen bonds and the intermolecular π-π stacking interactions between neighbouring aromatic rings (C10-C15) with centroid-to-centroid distances of 3.619 (6) Å (Fig. 2) establish the molecular conformation. In addition, the moleculars are further linked by the weak intermolecular C4-H4···π hydrogen bonds and the intermolecular π-π stacking interactions to form the infinite one-dimensional ribbon structure along c axis.

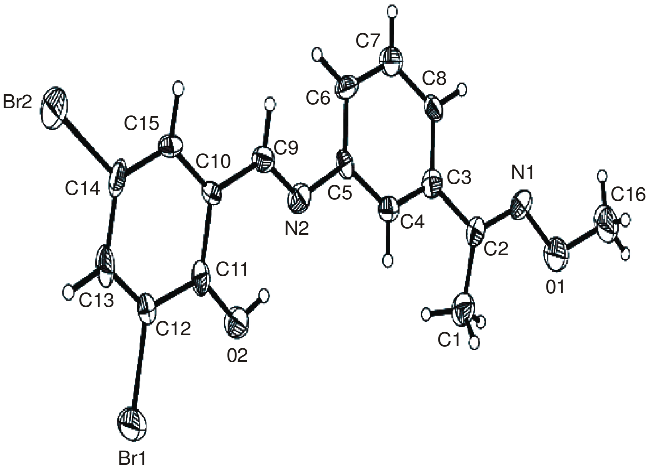

Fig. 1. Molecular structure of the title compound with atom numbering scheme

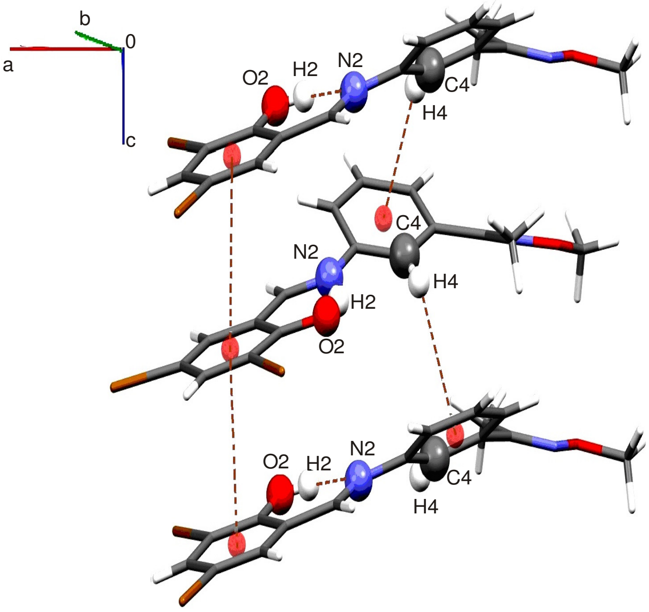

Fig. 2. Part of intramolecular O-H···N and intermolecular C-H···π interactions and π-π stacking interactions

ACKNOWLEDGEMENTS

The authors thank the science and technology support fund of Gansu province (212251) for financial support of this work.

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

Atom	Distance	Atom	Angle
N1-C2	1.232(4)	C2-N1-O1	110.2(3)
N1-O1	1.391(4)	N1-O1-C1	106.2(3)
O1-C1	1.475(5)	N1-C2-C3	118.5(4)
C2-C3	1.453(5)	C4-C3-C8	120.3(3)
C3-C4	1.347(4)	C4-C3-C2	119.1(3)
C3-C8	1.390(4)	C8-C3-C2	120.6(3)
C4-C5	1.404(5)	C11-C12-C7	122.2(4)
C5-C6	1.322(4)	C3-C4-C5	120.3(3)
C6-C7	1.375(5)	C6-C5-C4	119.9(4)
C7-C12	1.379(5)	C5-C6-C7	122.1(4)
C7-C8	1.416(5)	C6-C7-C12	122.0(4)
C8-C9	1.397(4)	C6-C7-C8	118.5(3)
C9-C10	1.340(4)	C12-C7-C8	119.4(4)
C10-C11	1.405(6)	C3-C8-C9	124.8(3)
C11-C12	1.338(5)	C3-C8-C7	118.8(3)
–	–	C9-C8-C7	116.4(3)
–	–	C10-C9-C8	123.3(4)
–	–	C9-C10-C11	119.0(4)
–	–	C12-C11-C10	119.6(4)

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