European Journal of Chemistry 8 (1) (2017) 15-17

European Journal of Chemistry

Journal webpage: www.eurjchem.com

Synthesis and X-ray crystallography of *N,N'*-di(2-hydroxybenzylidene)hydrazine

Sohail Saeed [1,2,*], Naghmana Rashid [1] and Shaaban Kamel Mohamed [3]

[1] *Department of Chemistry, Research Complex, Allama Iqbal Open University, Islamabad-44000, Pakistan*
[2] *National Engineering and Scientific Commission, Islamabad-44000, Pakistan*
[3] *Division of Chemistry and The Environmental Science, School of Science and The Environment, Manchester Metropolitan University, M15GD, England*

** Corresponding author at: Department of Chemistry, Research Complex, Allama Iqbal Open University, Islamabad-44000, Pakistan. Tel.: +92.51.90133246. Fax: +92.51.90133685. E-mail address: sohail262001@yahoo.com (S. Saeed).*

ARTICLE INFORMATION

DOI: 10.5155/eurjchem.8.1.15-17.1521

Received: 18 December 2016
Received in revised form: 07 January 2017
Accepted: 08 January 2017
Published online: 31 March 2017
Printed: 31 March 2017

KEYWORDS

Synthesis
Hydrazine
Schiff base
Crystal lattice
Hydroxybenzylidene
X-ray crystallography

ABSTRACT

The biologically active *N,N'*-di(2-hydroxybenzylidene)hydrazine has been synthesized and specifically characterized by X-ray crystallography. There are two molecules of *N,N'*-di(2-hydroxybenzylidene)hydrazine, $C_{14}H_{12}N_2O_2$, in the unit cell. The *N,N'*-di(2-hydroxy benzylidene)hydrazine molecule is planar, with maximum deviation from the mean plane being less than 0.028(2) Å. Intramolecular O-H···N hydrogen bonding interactions were observed in the crystal lattice which connected the molecules into chain running along *b*-axis.

Cite this: *Eur. J. Chem.* **2017**, *8(1)*, 15-17

1. Introduction

Schiff base compounds are important class of materials due to their flexibility, structural similarities with natural bio-active substances and also due to presence of imine (-N=CH-), which imports in elucidating the mechanism of transformation and racemization reaction in biological system [1]. Schiff bases-bimolecular condensation products of hydrazine with aldehydes- represent valuable intermediates in organic synthesis with various applications [2]. Schiff bases resulted from aromatic aldehydes *ortho*-substituted with a hydroxyl group have initially arouse the researches interest due to the several donor atoms in their structures which give them an advantage to form a water soluble transition metal complexes [3]. This advantage makes them have a potential application in water treatment [4]. They could also act as valuable ligands whose biological activity has been shown to increase on complexation [5,6]. Further to our interest in synthesis of diverse biologically active molecules, we have synthesized *N,N'*-di(2-hydroxybenzylidene)hydrazine and characterized by X-ray crystallography.

2. Experimental

2.1. Materials and physical measurements

Analytical grade chemicals were purchased from Sigma-Aldrich. Single crystal X-ray diffraction data for the title compound was collected using graphite-monochromated MoKα radiation (λ = 0.71073 Å) on a Bruker SMART 1000 CCD diffractometer.

2.2. X-ray structure determination

The structures were solved using direct methods and refined using least-square methods on F-squared [7,8]. All non-hydrogen atoms were refined anisotropically. Hydrogen atoms were included in calculated positions, assigned isotropic thermal parameters, and allowed to ride on their parent carbon atoms.The crystal-to-detector distance was 50.00 mm. Indexing was performed from 60 images that were exposed for 10 s for a preliminary unit cell determination. Of which, 63 out of total of 74 reflections were successfully indexed.

ISSN 2153-2249 (Print) / ISSN 2153-2257 (Online)
http://dx.doi.org/10.5155/eurjchem.8.1.15-17.1521

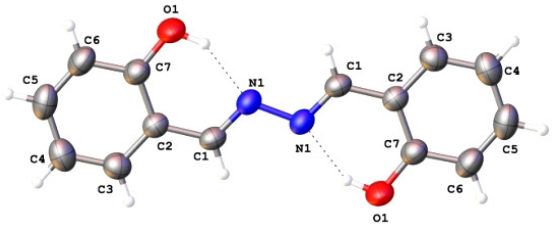

Figure 1. X-ray structure of *N,N′*-di(2-hydroxybenzylidene)hydrazine.

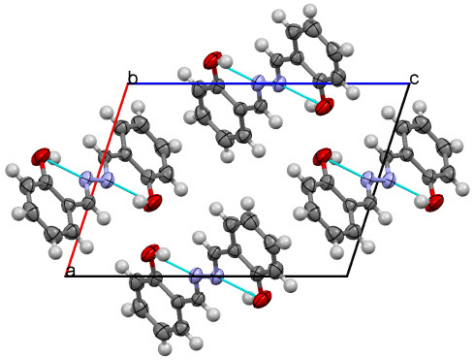

Figure 2. Packing diagram of *N,N′*-di(2-hydroxybenzylidene)hydrazine viewed along the *c*-axis.

The data were collected at a temperature of 23(1) °C to a maximum 2θ value of 50.0°. A total of 1421 oscillation images were collected in 4 runs. A sweep of data was done using ω scans from 330.0 to 148.2° in -0.3° step, at χ= 54.7° and Ø= 0.0°. The exposure rate was 50.0 [sec./°]. The detector swing angle was -30.00°. A second sweep was performed using ω scans from 330.0 to 199.5° in -0.3° step, at χ = 54.7° and Ø = 90.0°. The detector swing angle was -30.00°. A third sweep was performed using ω scans from 330.0 to 261.0° in -0.3° step, at χ= 54.7° and Ø = 180.0°. The detector swing angle was -30.00°. A last sweep was performed using ω scans from 330.0 to 285.0° in -0.3° step, at χ = 54.7° and Ø = 270.0°. The detector swing angle was -30.00°.

2.3. Preparation of N,N'-di(2-hydroxybenzylidene) hydrazine

The compound has been synthesized un-intentionally and isolated as a major product from the reaction of 0.01 mol 1,6-dibromo hexane with 0.01 mole hydrazine in presence ofa catalytic amount of potassium carbonate (10 mg) in methanol for 10 minutes at 338 K followed by addition of 0.01 molsalicylaldehyde. The reaction mixture was heated for further 30 minutes then poured on crushed ice (50 g). A pink cake was filtered off, washed with cold ethanol, dried and recrystallized from ethanol. The resulting shiny yellow crystals were pure enough in 83% yield and were suitable for X-ray diffraction. M.p.: 499-503 K. Suitable single crystals forX-ray diffraction were grown from a dilute ethanolic solution by slow evaporation method.

3. Results and discussion

The molecular structure and unit cell diagrams of *N,N′*-di(2-hydroxybenzylidene)hydrazine are shown in Figure 1 and 2, respectively. Crystallographic data, selected bond lengths and angles, atomic displacement parameters and hydrogen-bond geometry are listed in Table 1-4, respectively.

There are four molecules of *N,N′*-di(2-hydroxybenzylide ne)hydrazine, in the unit cell (Figure 2). The crystal of *N,N′*-di (2-hydroxybenzylidene)hydrazine, $C_{14}H_{12}N_2O_2$, a light-yellow plate, having approximate dimensions of 0.06 × 0.20 × 0.36 mm was mounted on glass fiber. Cell constants and an orientation matrix for data collection corresponded to a primitive monoclinic cell with dimensions: a = 8.5332(11), b = 6.3185(8), c = 11.8338(15) Å, V = 607.10(13) Å^3 and β = 107.917(2)° for Z = 2, the calculated density is 1.314 g/cm^3. Based on a statistical analysis of intensity distribution, and the successful solution and refinement of the structure, the space group was determined to be: $P2_1/n$ (#14). The data were collected at a temperature of 24(1) °C to a maximum 2θ value of 50.0°. Of the 3711 reflections that were collected, 1071 reflections were unique. (R_{int} = 0.0129); equivalent reflections were merged.

Table 1. Crystallographic data of the compound.

Empirical formula	$C_{14}H_{12}N_2O_2$
Formula weight	240.26
Temperature/K	297(2)
Crystal system	Monoclinic
Space group	$P2_1/n$
Hall symbol	-P 2yn
a/Å	8.5332(11)
b/Å	6.3185(8)
c/Å	11.8338(15)
β/°	107.917(2)
Volume/Å^3	607.10(13)
Z	2
ρ_{calc}g/cm^3	1.314
μ/mm^{-1}	0.090
F(000)	252.0
Crystal size/mm^3	0.36 × 0.20 × 0.06
Radiation	MoKα (λ = 0.71073)
2Θ range for data collection/°	5.2 to 50.04
Index ranges	$-10 \le h \le 9$, $-5 \le k \le 7$, $-14 \le l \le 14$
Reflections collected	3211
Absorption correction	Multi-scan
T_{min}, T_{max}	0.968, 0.995
F(000)	252
Independent reflections	1071 [R_{int} = 0.0129]
Data/restraints/parameters	1071/0/86
Goodness-of-fit on F^2	1.008
Final R indexes [I≥2σ (I)]	R_1 = 0.0343, wR_2 = 0.0994
Final R indexes [all data]	R_1 = 0.0389, wR_2 = 0.1061
Largest diff. peak/hole / e Å^{-3}	0.10/-0.18

Table 2. Selected bond lengths (Å) and angles (°).

Bond Lengths (Å)			
O1—C7	1.3500 (15)	O1—H1O	0.88 (2)
O1—H1O	0.88 (2)	N1—C1	1.2804 (15)
N1—C1	1.2804 (15)	C3—C4	1.3762 (19)
N1—N1[i]	1.3966 (18)	C4—C5	1.386 (2)
C1—C2	1.4474 (16)	C5—C6	1.3671 (19)
C2—C3	1.3957 (16)	C6—C7	1.3878 (18)
C2—C7	1.4068 (16)	C3—C4	1.3762 (19)
O1—C7	1.3500 (15)	C4—C5	1.386 (2)
Bond Angles (°)			
C7—O1—H1O	107.5 (13)	C6—C5—C4	120.59 (12)
C1—N1—N1[i]	113.83 (11)	C5—C6—C7	120.70 (12)
N1—C1—C2	121.48 (10)	O1—C7—C6	118.46 (11)
C3—C2—C7	118.14 (11)	O1—C7—C2	121.79 (11)
C3—C2—C1	119.99 (10)	C6—C7—C2	119.74 (12)
C7—C2—C1	121.86 (11)	O1—C7—C6	118.46 (11)
C4—C3—C2	121.59 (12)	O1—C7—C2	121.79 (11)
C7—O1—H1O	107.5 (13)	C6—C7—C2	119.74 (12)
C4—C3—C2	121.59 (12)		

[1]Symmetry code: (i) 1-*x*,-*y*,-*z*.

Table 3. Anisotropic displacement parameters (Å²) for the title compound.

Atom	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
O1	0.0614 (6)	0.0835 (8)	0.0507 (6)	0.0177 (5)	-0.0083 (4)	-0.0223 (5)
N1	0.0497 (6)	0.0424 (5)	0.0354 (5)	-0.0017 (4)	0.0093 (4)	-0.0039 (4)
C1	0.0430 (6)	0.0451 (7)	0.0338 (6)	-0.0049 (5)	0.0064 (4)	0.0001 (4)
C2	0.0436 (6)	0.0427 (6)	0.0377 (6)	-0.0045 (5)	0.0134 (5)	0.0009 (5)
C3	0.0495 (7)	0.0513 (8)	0.0487 (7)	0.0018 (5)	0.0118 (5)	0.0041 (5)
C4	0.0636 (8)	0.0509 (8)	0.0703 (9)	0.0093 (6)	0.0291 (7)	0.0056 (6)
C5	0.0765 (9)	0.0477 (8)	0.0662 (9)	-0.0052 (6)	0.0390 (7)	-0.0102 (6)
C6	0.0633 (8)	0.0614 (8)	0.0482 (7)	-0.0088 (6)	0.0182 (6)	-0.0156 (6)
C7	0.0475 (7)	0.0540 (7)	0.0402 (6)	-0.0022 (5)	0.0121 (5)	-0.0049 (5)

Table 4. Hydrogen-bond geometry (Å, °).

D*—H···*A	***D*—H**	**H···*A***	***D*···*A***	***D*—H···*A***
O1—H1*O*···N1 [i]	0.88 (2)	1.83 (2)	2.6134 (13)	148.7 (19)

Symmetry code: (i) -*x*+1, -*y*, -*z*.

4. Conclusion

N,*N*'-Di(2-hydroxybenzylidene)hydrazine was synthesized successfully and its structure has been determined by X-ray single crystallography. Due to presence of imine (-N=CH-) group in the compound it may be helpful in elucidating the mechanism of transformation and racemization reaction in biological systems. In future, we will study its biological activities.

Acknowledgments

Dr. Sohail Saeed is thankful to the National Engineering and Scientific Commission (NESCOM), Islamabad, Pakistan for providing the lab space and necessary analytical facilities.

Supplementary data

Crystallographic data for the structure reported in this article have been deposited with Cambridge Crystallographic Data Center, CCDC 932806. Copies of this information may be obtained free of charge from the Director, CCDC, 12 Union Road, Cambridge, CBZ IEZ, UK. Facsimile (44) 01223 336 033, E-mail: deposit@ccdc.cam.ac.uk or http://www.ccdc.com.ac.uk/deposit.

References

[1]. Rajavel, P.; Senthil, M. S; Anitha, C. *E-Journal Chem*. **2008**, *5(3)*, 620-626.
[2]. Ugras, H. I.; Basaran, I.; Kilic. T.; Cakir, U. *J. Heterocyclic Chem*. **2006**, *43*, 1679-1684.
[3]. Wadher, S. J.; Puranik, M. P.; Karande, N. A.; Yeole, P. G. *Int. J. Pharm. Tech. Res.* **2009**, *1*, 22-33.
[4]. Elena, E. F.; Torres, L.; Antonia, M.; Mendialoey, M. A.; Sevela, M. T. *Polyhedron* **2000**, *19*, 441-451.
[5]. Mohamed, G. G.; Omar, M. M.; Hindy, A. M. *Turk. J. Chem.* **2006**, *30*, 361-382.
[6]. Siddiqui, H. L.; Iqbal, A.; Ahmad, S.; Weaver, G. W. *Molecules* **2006**, *11*, 206-211.
[7]. Oxford Diffraction, CrysAlis PRO and CrysAlis RED. Oxford Diffraction Ltd, Abingdon, Oxfordshire, England, 2010.
[8]. Sheldrick, G. M. *Acta Cryst. A* **2008**, *64*, 112-122.