

# Synthesis of mono and bis-substituted compounds 1-(2-hydroxybenzylidene)carbonohydrazide and 1,5-bis(2-hydroxybenzaldehyde)carbohydrazone : Study spectroscopy and X-ray diffraction

## Abstract

The unsymmetric monocarbonohydrazide Schiff base ( $H_5L^1$ ) (1) and the symmetric dicarbonohydrazone ( $H_4L^2$ ) (2) were prepared from a monocondensation reaction between carbonohydrazide and 2-hydroxybenzaldehyde, respectively, in 1/1 and 1/2 ratio. The compounds were characterized by elemental analysis,  $^1H$  and  $^{13}C$  NMR and FTIR spectroscopy. The NMR and FTIR data analysis revealed that compounds (1) and (2) are in the amide form in the solid state as well as in DMSO solution. The molecular structure of each Schiff base compound has been elucidated by X-ray diffraction analysis. Compound  $H_5L^1$  (1) crystallizes in the monoclinic system in  $P2_1/c$  space group with cell parameters:  $a = 17.0790(2)$  Å,  $b = 11.07410(10)$  Å,  $c = 10.56110(10)$  Å,  $\beta = 106.5730(10)^\circ$ ,  $V = 1914.49(3)$  Å<sup>3</sup>,  $Z = 4$ ,  $R_1 = 0.0432$ ,  $wR_2 = 0.1275$ . The compound  $H_4L^2$  (2) crystallizes in the orthorhombic system in  $C222_1$  space group with cell parameters;  $a = 4.5494(1)$  Å,  $b = 12.2213(2)$  Å,  $c = 27.4001(6)$  Å,  $V = 1523.43(5)$  Å<sup>3</sup>,  $Z = 4$ ,  $R_1 = 0.039$ ,  $wR_2 = 0.125$ . Both compounds exhibit inter and intramolecular hydrogen bonds which consolidate the structures into three-dimensional networks.

*Keywords:* carbonohydrazide, 2-hydroxybenzaldehyde, NMR, FTIR, crystal, and XRD

## 1. INTRODUCTION

Carbonohydrazide is a symmetrical two-armed molecule bearing two highly reactive hydrazide units [1-3]. They are widely used to synthesize organic compounds with applications in various fields [4-6]. With ketones, less reactive than aldehydes, carbonohydrazides can perform monocondensation or di-condensation reactions to yield monocarbonohydrazide or dicarbonohydrazone Schiff bases [7-8]. However, the monocondensation of carbonohydrazide with aldehyde provides a mixture of dicarbonohydrazone and monocarbonohydrazide compounds due to the high reactivity of aldehydes. Thus, many chemists are looking for an easy, reproducible synthetic method to isolate monocarbonohydrazide derivatives of aldehydes in high yields. The only result observed in the literature is the compound 1-(2'-hydroxy-3'-methoxybenzylidene)carbonohydrazide [9] whose structure is confirmed by infrared spectroscopic techniques, NMR, and elemental analyses. Many of these carbonohydrazide derivatives are used as precursors in the preparation of heterocyclic compounds with interesting biological properties [10-13]. Some of them have made it possible to develop drugs with a broad spectrum of activity such as antimicrobial [14], anticonvulsant [15], antidepressant [16], antioxidant [17-19], analgesic [20], antifungal [21], antiplatelet [22], antituberculosis [23], anti-HIV [24], inflammatory [25], antidiabetic [26], anticancer [17], and antimalarial [28]. Due to the presence of several donor atoms in these derivatives and the possibility for these molecules to appear in different conformations, they can be used in coordination chemistry. Coordination compounds with various structures and properties are reported in the literature [29-32]. It has been reported in the literature that dicarbonohydrazone with their *cis-enol* conformations or *cis-enol* and *trans* association lead to square or rectangular grid polynuclear complexes, respectively, with interesting physical properties [33-34]. In these coordination compounds, the dicarbonohydrazone act as  $\mu_2-O$  [35-36] and/or  $\mu_2-N-N$  bridged pentadentate or hexadentate ligand [37-40]. Depending on their tridentate or tetradentate behavior, monocarbonohydrazide yield, generally, mononuclear or dinuclear complexes with transition metal ions [41]. Recently, derivative complexes with remarkable magnetic [42, 43], luminescent [44] or catalytic [45] properties have been synthesized and studied. In this article, we report the synthesis and characterization of one monosubstituted ( $H_5L^1$ ) and one di-substituted ( $H_4L^2$ ) Schiff bases derived from carbonohydrazide.

## 2. EXPERIMENTAL PART

## 2.1 Materials and instruments

2-Hydroxybenzaldehyde, carbonohydrazide, and solvent from Sigma-Aldrich were analytical grade and were used directly without further purification. Elemental analyzes of C, H and N were recorded on a VxRio EL instrument. Infrared spectra were obtained on a Perkin Elmer Spectrum Two FTIR spectrometer in the 4000-400  $\text{cm}^{-1}$  region. NMR spectra were recorded in DMSO- $d_6$  solution with a Bruker 250 MHz spectrometer at room temperature using TMS as internal reference. UV-Vis spectra were recorded in a DMF solution with a concentration of  $10^{-3}$  M at 25 °C on a Perkin Elmer Lambda 365 UV-Vis spectrophotometer.

## 2.2 Synthesis of the 1-(2-hydroxybenzylidene)carbonohydrazide ligand (1)

3 g (33.3 mmol) of carbonohydrazide are dissolved in a 250 mL two-necked flask containing 30 mL of methanol and 10 mL of distilled water. A methanol solution containing 4.0702 g (33.3 mmol) of 2-hydroxybenzaldehyde is added dropwise using a separatory funnel. After the total addition of the solution, the mixture is maintained at reflux for six hours. A clear yellow solution is obtained which gives a precipitate after cooling at 4°C overnight. This precipitate is filtered off and washed with cold methanol (2 x 10 mL) and then with diethyl ether (2 x 10 mL) before being dried under vacuum. The purity of the product is checked by thin layer chromatography (TLC) using gel plates of silica, the eluent being a methanol/dichloromethane mixture with a ratio of 9/1 (V/V). M.p. = 187°C. Yield : 86.3%. Anal. [Found (calculated)] ( $\text{C}_8\text{H}_{10}\text{N}_4\text{O}_2$ ): % C, 49.43 (49.48); %H, 5.14 (5.19); %N, 28.83 (28.63). FTIR ( $\nu$ ,  $\text{cm}^{-1}$ ): 3352 (N-H); 3280 (O-H); 3233 (N-H); 3092 ( $\text{C}_{\text{sp}^2}$ -H); 1676 (C=O); 1632 (C=N); 1606 ( $\square(\text{NH}_2)$ ); 1544 ( $\text{C}_{\text{Ar}} = \text{C}_{\text{Ar}}$ ); 1489 ( $\text{C}_{\text{Ar}} = \text{C}_{\text{Ar}}$ ); 1460 ( $\text{C}_{\text{Ar}} = \text{C}_{\text{Ar}}$ ); 1272 (C-O); 1068 (N-N).  $^1\text{H}$  NMR (dms- $d_6$ ,  $\delta(\text{ppm})$ ): 4.14 (s, 2H,  $\text{NH}_2$ ); 6.85-7.92 (m, 4H, H-Ph); 8.21 (s, 1H,  $-(\text{C}=\text{O})-\text{NH}-\text{NH}_2$ ); 8.44 (s, 2H,  $\text{N}=\text{C}-\text{H}$ ); 10.41 (s, 1H,  $-\text{O}-\text{H}_{\text{phenol}}$ ); 10.87 (s, 1H,  $-(\text{C}=\text{O})-\text{NH}-(\text{C}=\text{N})-$ ).  $^{13}\text{C}$  NMR (dms- $d_6$ ,  $\delta(\text{ppm})$ ): 156.13 ( $\text{C}=\text{N}$ ); 151.99 ( $\text{C}_{\text{Ar}}-\text{OH}$ ); 140.1 ( $\text{C}=\text{N}$ ); 130.61 ( $\text{C}_{\text{Ar}}$ ); 129.91 ( $\text{C}_{\text{Ar}}$ ); 127.86 ( $\text{C}_{\text{ispo}}$ ); 119.13 ( $\text{C}_{\text{Ar}}$ ); 115.97 ( $\text{C}_{\text{Ar}}$ ). UV-vis (Solution, DMF,  $\lambda$  (nm)): 240, 285, 322.

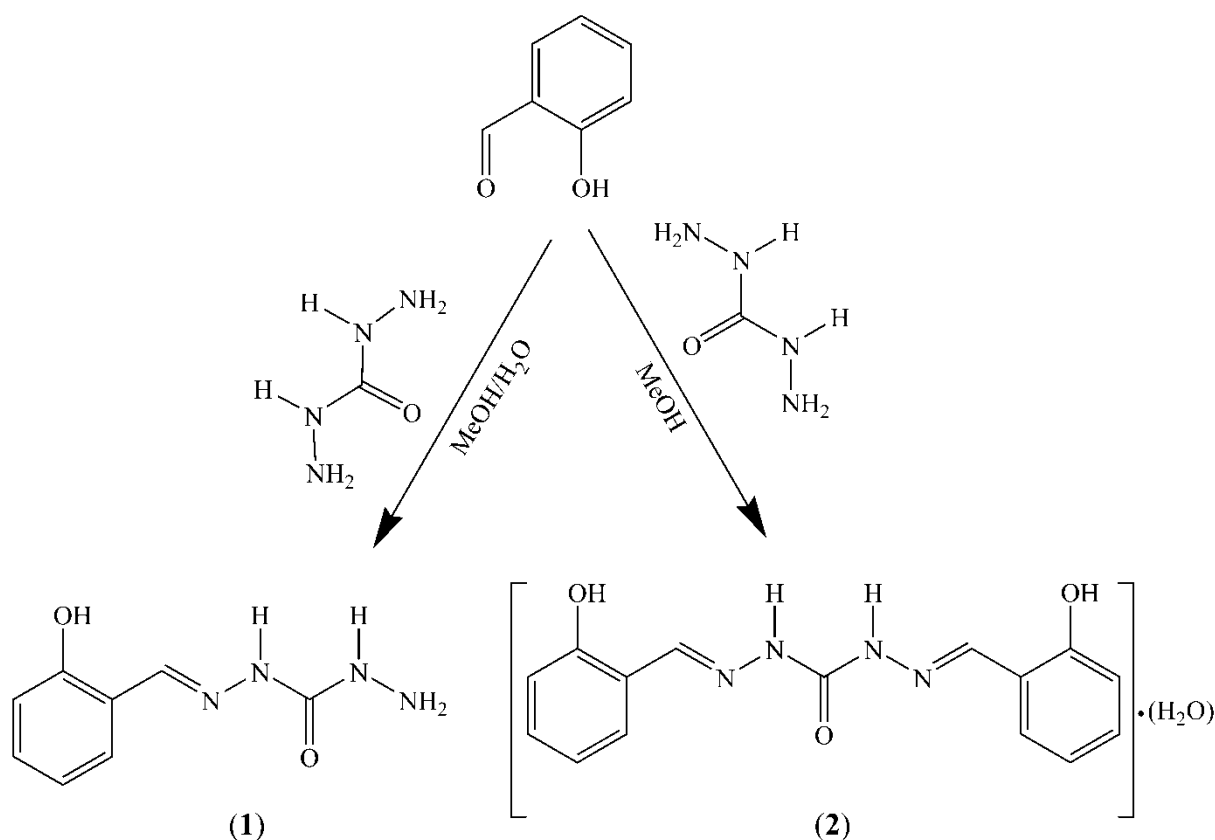
## 2.3 Synthesis of the ligand 1,5-Bis(2-hydroxybenzaldehyde)carbohydrazone monohydrate (2)

1 g (11.1 mmol) of carbonohydrazide was suspended in 10 mL of methanol. 2.710 g (22.2 mmol) of 2-hydroxybenzaldehyde previously dissolved in 10 mL of methanol were added. After adding a few drops of glacial acetic acid, the mixture was heated under reflux for two hours. The white precipitate formed, was recovered by filtration, and washed with 5 mL of methanol before being dried in the open air. M.p. = 218°C. Yield : 93.4%. Anal. [Found (calculated)] ( $\text{C}_{15}\text{H}_{16}\text{N}_4\text{O}_4$ ): %C, 56.93 (56.96); %H, 5.08 (5.10); %N, 17.68 (17.71). FTIR (KBr,  $\text{cm}^{-1}$ ): 3260 (O-H); 3184 (N-H); 3042 (C-H); 1686 (C=O); 1620 (C=N); 1576 ( $\text{C}_{\text{Ar}} = \text{C}_{\text{Ar}}$ ); 1489 ( $\text{C}_{\text{Ar}} = \text{C}_{\text{Ar}}$ ); 1464 ( $\text{C}_{\text{Ar}} = \text{C}_{\text{Ar}}$ ); 1270 (C-O); 1073 (N-N).  $^1\text{H}$  NMR (dms- $d_6$ ,  $\delta(\text{ppm})$ ): 6.90-7.27 (m, 8H, H-Ph); 7.71 (s, 2H, NH); 8.45 (s, 2H,  $\text{N}=\text{C}-\text{H}$ ); 10.89 (s, 4H,  $-\text{O}-\text{H}_{\text{phenol}}$ ).  $^{13}\text{C}$ -NMR (dms- $d_6$ ,  $\delta(\text{ppm})$ ): 156.57 (C=O); 151.44 ( $\text{C}_{\text{Ar}}-\text{OH}$ ); 146.1 (C=N); 130.39 ( $\text{C}_{\text{Ar}}$ ); 128.05 ( $\text{C}_{\text{Ar}}$ ); 119.90 ( $\text{C}_{\text{Ar}}$ ); 118.85 ( $\text{C}_{\text{ispo}}$ ); 116.13 ( $\text{C}_{\text{Ar}}$ ). UV-vis (Solution, DMF,  $\lambda$  (nm)): 257, 290, 316.

## 3. RESULTS AND DISCUSSION

### 3.1 General study

The compounds  $\text{H}_5\text{L}^1$  (**1**) was obtained after the condensation reaction between carbonohydrazide and 2-hydroxybenzaldehyde in 1/1 in MeOH/ $\text{H}_2\text{O}$  (1/1) solution while the ligand  $\text{H}_4\text{L}^2$  (**2**) was obtained after the condensation reaction between carbonohydrazide and 2-hydroxybenzaldehyde in 1/2 in MeOH solution (Figure 1). The two compounds **1** and **2** were characterized by elemental analyses, FTIR,  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectroscopies. The data of the elementary analyzes of the two compounds **1** and **2** confirm their formations with the chemical formulas  $\text{C}_8\text{H}_{10}\text{N}_4\text{O}_2$  (**1**) and  $\text{C}_{15}\text{H}_{14}\text{N}_4\text{O}_3 \cdot \text{H}_2\text{O}$  (**2**), respectively. The FTIR spectrum of the compound **1** shows bands at 3280  $\text{cm}^{-1}$  and 3233  $\text{cm}^{-1}$  attributable respectively to the phenolic  $\nu(\text{OH})$  and the hydrazide  $\nu(\text{NH})$  vibrations [46-47]. The bands pointed at 3352  $\text{cm}^{-1}$  and 1606  $\text{cm}^{-1}$  are, respectively, characteristic of the  $\nu(\text{NH}_2)$  and  $\square(\text{NH}_2)$  vibrations of the  $-\text{NH}_2$  group. Their appearance proves the monocondensation reaction [9]. The  $\nu(\text{C}=\text{O})$  carbonyl vibration band appears at 1676  $\text{cm}^{-1}$  while the  $\nu(\text{C}=\text{N})$  band of the imine function is pointed at 1632  $\text{cm}^{-1}$  [48]. Between 1544 and 1460  $\text{cm}^{-1}$ , bands corresponding to the  $\nu(\text{C}=\text{C})$  vibrations of the aromatic rings are pointed. A band at 1272  $\text{cm}^{-1}$  corresponding to the vibration of the  $\text{C}_{\text{Ar}}-\text{O}$ -bond is also observed on the FTIR spectrum.



**Figure 1. Synthesis diagram of compounds H<sub>5</sub>L<sup>1</sup> (1) and (2)**

The vibration band of the N–N hydrazine bond appears at  $1068\text{ cm}^{-1}$ . The absence of the  $\nu(\text{NH}_2)$  and  $\nu(\text{NH})$  vibration bands of the  $\text{NH}_2$  group on the IR spectrum of the compound **2** expected respectively at  $\text{ca. } 3300\text{ cm}^{-1}$  and  $1600\text{ cm}^{-1}$  confirms the 1/2 ratio condensation reaction between carbonohydrazide and 2-hydroxybenzaldehyde. The  $\nu(\text{NH})$  vibration bands of the hydrazide function,  $\nu(\text{C}=\text{O})$  carbonyl and  $\nu(\text{C}=\text{N})$  imine of the ligand **2** are located respectively at  $3184\text{ cm}^{-1}$ ,  $1686\text{ cm}^{-1}$  and  $1620\text{ cm}^{-1}$ . The band attributable to  $\nu(\text{C}_{\text{Ar}}-\text{O}-)$  vibration is observed at  $1270\text{ cm}^{-1}$  and the band due to  $-\text{N}-\text{N}-$  bond of the hydrazine moiety is pointed at  $1073\text{ cm}^{-1}$ . In the  $^1\text{H}$  NMR spectrum of compound **1**, the presence of the signal due to  $-\text{NH}_2$  at  $4.14\text{ ppm}$  is indicative of the monocondensation reaction. The signal at  $10.41\text{ ppm}$  is due to the single phenolic proton. In addition, the two  $-\text{NH}$  protons are observed at  $7.44\text{ ppm}$  and  $10.87\text{ ppm}$ . No signal due to an alcoholic proton is observed. These observations indicate that the compound remained in its keto form in DMSO solution. The signal of the azomethine proton is pointed at  $8.44\text{ ppm}$  and those corresponding to the protons of the aromatic rings are pointed between  $6.85\text{--}7.92\text{ ppm}$ . Indeed, when the  $^{13}\text{C}$  NMR spectrum of compound **1** is examined, the signals of the carbon atoms of the  $\text{C}=\text{O}$ ,  $\text{C}_{\text{Ar}}-\text{O}-$  and  $\text{C}=\text{N}$  functions appear respectively at  $156.13\text{ ppm}$ ,  $151.99\text{ ppm}$  and  $140.1\text{ ppm}$ . The signals between  $115.97\text{--}130.61\text{ ppm}$  correspond to the carbon atoms of the aromatic ring. No signal attributable to a carbon atom carrying an alcohol function  $-\text{N}=\text{C}(\text{OH})-\text{N}-$  was observed. This confirms that only the ketone form is present in solution of **1**. As in the case of compound **1**, compound **2** in solution may exhibit *Z/E* isomerism around the two imine bonds and the presence of the  $-\text{HN}-\text{C}(=\text{O})$  motif can induce keto tautomerism. Several conformers can coexist in solution.  $^1\text{H}$  and  $^{13}\text{C}$  NMR of compound **2** at room temperature give information on tautomerism. In the  $^1\text{H}$  spectrum, the singlet signal at  $10.89\text{ ppm}$  is due to the  $-\text{OH}$  proton of the phenolic function while the signal at  $7.71\text{ ppm}$  represents two  $-\text{NH}$  protons of the  $\text{N}-\text{HN}-\text{C}(=\text{O})$  moiety. The absence of signal attributable to the  $-\text{NH}_2$  group expected around  $4.14\text{ ppm}$  is indicative of the success of the 1/2 ratio condensation reaction. In the  $^1\text{H}$  NMR spectrum, no signal attributable to the enol proton  $-\text{HN}-\text{C}(\text{OH})=\text{N}-$  is observed. This observation demonstrates clearly that compound **2** is only present in its keto form in solution. The  $^1\text{H}$  NMR spectrum also contains a singlet signal at  $8.45\text{ ppm}$  representing the azomethine proton and a multiplet representing eight aromatic protons in the range  $6.90\text{--}7.27\text{ ppm}$ . On the  $^{13}\text{C}$  NMR spectrum, the single signal observed at  $156.57\text{ ppm}$  was attributed to  $\text{C}=\text{O}$  and two signals at  $151.44\text{ ppm}$  and  $146.1\text{ ppm}$  are, respectively, attributed to the carbon atoms  $\text{C}_{\text{Ar}}-\text{OH}$  and  $\text{C}=\text{N}$ . Aromatic carbon atom signals are located between  $116.13\text{--}$

130.39 ppm. No signal attributable to the enol carbon atom  $\text{-HN-C(-OH)=N-}$  is observed in the  $^{13}\text{C}$  NMR spectrum of compound **2** confirming that compound **2** is only present in its keto form in solution. The UV-visible spectra of the two compounds were recorded in DMSO solution. In the UV-vis spectra of the two compounds **1** and **2**, the characteristic transition bands of the aromatic ring  $\pi \rightarrow \pi^*$  and  $n \rightarrow \pi^*$  of the chromophore  $\text{C=N}$  are observed in the range 240-290 nm and 316-322 nm, respectively.

**Table 1. Crystallographic data and refinement parameters for compounds 1 and 2**

	1	2
Chemical formula	$2(\text{C}_8\text{H}_{10}\text{N}_4\text{O}_2)$	$\text{C}_{15}\text{H}_{14}\text{N}_4\text{O}_3 \cdot \text{H}_2\text{O}$
M (g/mol)	388.38	318.33
Temperature (K)	295	298
Crystal system	monoclinic	Orthorhombic
Space group	$P2_1/c$	$C222_1$
$a$ (Å)	17.0790(2)	4.5494 (1)
$b$ (Å)	11.07410(10)	12.2213 (2)
$c$ (Å)	10.56110(10)	27.4001 (6)
$\alpha$ (°)	90	90
$\beta$ (°)	106.5730(10)	90
$\gamma$ (°)	90	90
$V$ (Å <sup>3</sup> )	1914.49(3)	1523.43 (5)
Z	4	4
Radiation type	Cu K $\alpha$	Mo K $\alpha$
$\rho_{\text{calc}}$ (g/cm <sup>3</sup> )	1.347	1.388
$\mu$ (mm <sup>-1</sup> )	0.867	0.10
Crystal size (mm)	0.32 × 0.26 × 0.08	0.30 × 0.28 × 0.08
$F(000)$	816.0	672
$\theta_{\text{max}}$ ; $\theta_{\text{min}}$ (°)	71.0 ; 4,8	32.2 ; 3.3
$h, k, l$	$-20 \leq h \leq 20, -13 \leq k \leq 13, -12 \leq l \leq 12$	$-6 \leq h \leq 6, -18 \leq k \leq 18, -40 \leq l \leq 41$
No. of measured reflections	33758	48700
Independent reflections	3684	2627
Observed reflections [ $I > 2\sigma(I)$ ]	3439	2086
$R_{\text{int}}$	0.0292	0.047
$R_1, wR_2$ ( $I \geq 2\sigma(I)$ )	$R_1 = 0.0432, wR_2 = 0.1275$	$R_1 = 0.039, wR_2 = 0.125$

Data/parameters/restraints	3684/257/0	2627/107/0
GOF	1.112	1.08
$\Delta\rho_{\max}, \Delta\rho_{\min}$ ( $e \text{ \AA}^{-3}$ )	0.16/-0.15	0.14, -0.29

### 3.2 Description of the crystal structure of the compound 1

The monocarbonohydrazide compound **1** crystallizes in the monoclinic system with the space group  $P2_1/c$ . Selected bond lengths and bond angles are listed in Table 2. Figure 1 shows the ORTEP view of the compound **1**. The asymmetric unit of the crystal structure of compound **1** consists of two molecules of the ligand. In the structure each molecule adopts an *E* configuration with respect to the C=N bond of the azomethine functions [C7=N1 or C15=N5]. In each molecule, the oxygen atom of the carbonyl function and the nitrogen atom of the azomethine are in *syn* conformation with respect to the corresponding C–N bond [O2 and N1/C8–N2; O4 and N5/C16–N6] with torsion angle values of  $-3.7(4)^\circ$  [N1–N2–C8–O2] and  $4.04(4)^\circ$  [N5–N6–C16–O4]. The carbonyl oxygen and the amino nitrogen atom in each molecule are also in *cis* conformation with respect to the C–N bond [O2 and N4/C8–N3 and O4 and N8/C16–N7] with torsion angle values of  $3.9(4)^\circ$  [N4–N3–C8–O2] and  $-2.9(4)^\circ$  [N8–N7–C16–O4]. The nitrogen atom of the azomethine group C=N is in *trans* position with the hydrazine nitrogen atom (NH–NH<sub>2</sub>) within each molecule with respect to the C–N bond [N1 and N3/C8–N2 and N5 and N7/C16–N6] with torsion angle values of  $177.2(2)^\circ$  [N1–N2–C8–N3] and  $-175.7(2)^\circ$  [N5–N6–C16–N7]. The bond lengths C8–O2 and C16–O4 of 1.197 (3) Å characteristic of a double bond character, show that the two molecules of compound **1** are in the keto form in the solid state [49]. This tautomeric form is confirmed by bond distances N2–C8 [1.359 (3) Å], N1–N2 [1.360 (3) Å], N6–C16 [1.355 (3) Å], N5–N6 [1.363 (3) Å] which indicate that they are single bonds [50] and by the bonds C7–N1 [1.287 (3) Å] and C15–N5 [1.292 (3) Å] which are double bonds [51]. In each molecule of compound (**1**), intramolecular hydrogen bonds of the type O–H...N [O1–H1...N1 and O3–H3...N5] which are established between the phenolic oxygen atoms as donor and the azomethine nitrogen atoms as acceptor resulting in S(6) rings. The different molecules in the crystal are connected by intermolecular hydrogen bonds of type N–H...O [N2–H2...O1<sup>i</sup>:  $i = -x, y+1/2, -z+3/2$ ; N8–H8A...O4<sup>ii</sup>, N4–H4B...O2<sup>ii</sup>:  $ii = x, -y+1/2, z-1/2$ ; and N6–H6...O3<sup>iii</sup>:  $iii = -x+1, y+1/2, -z+1/2$ ; N8–H8B...O2<sup>v</sup>:  $v = x+1, -y+1/2, z-1/2$ ]; N–H...N [N8–H8A...N5<sup>ii</sup>] and C–H...N [C11–H11...N7<sup>iv</sup>:  $iv = -x+1, y-1/2, -z+1/2$ ] (Table 3). All of these intramolecular and intermolecular hydrogen bonds ensure the stability of the structure and its development in a three-dimensional network (Figure 2).

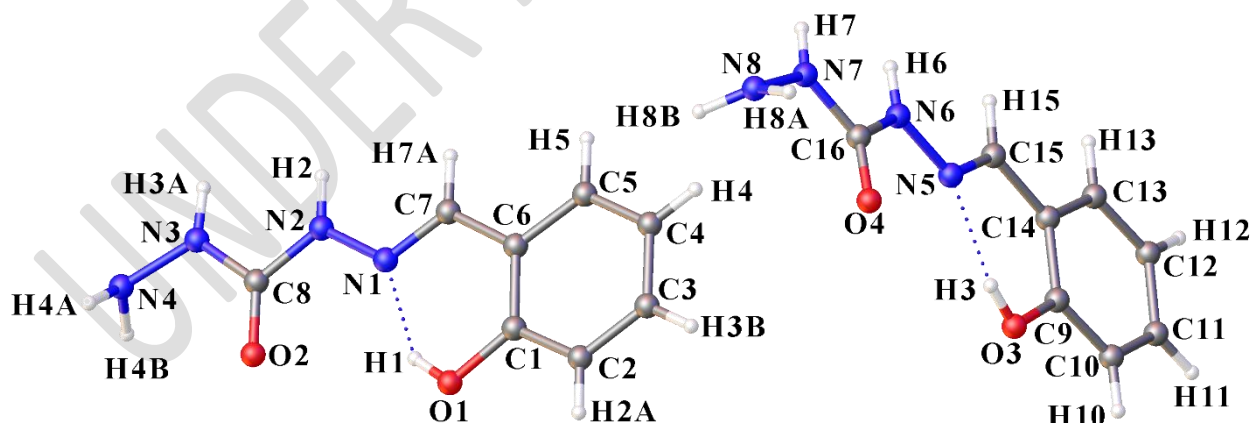


Figure 2. ORTEP view of compound 1

Table 2. Selected bond distances [Å] and angles [deg] for compound 1

O3—C9	1.361 (3)	O1—C1	1.363 (3)
O4—C16	1.197 (3)	O2—C8	1.195 (3)
N7—C16	1.339 (3)	N3—C8	1.338 (3)
N7—N8	1.441 (4)	N3—N4	1.442 (4)
N5—C15	1.292 (3)	N1—C7	1.287 (3)
N5—N6	1.363 (3)	N1—N2	1.360 (3)
N6—C16	1.355 (3)	N2—C8	1.359 (3)
C15—N5—N6—C16	-175.7 (2)	N5—N6—C16—N7	-175.7 (2)
O3—C9—C10—C11	179.8 (2)	N4—N3—C8—O2	3.9 (4)
N8—N7—C16—O4	-2.9 (4)	N4—N3—C8—N2	-177.1 (2)
N5—N6—C16—O4	4.0 (4)	N1—N2—C8—O2	-3.7 (4)

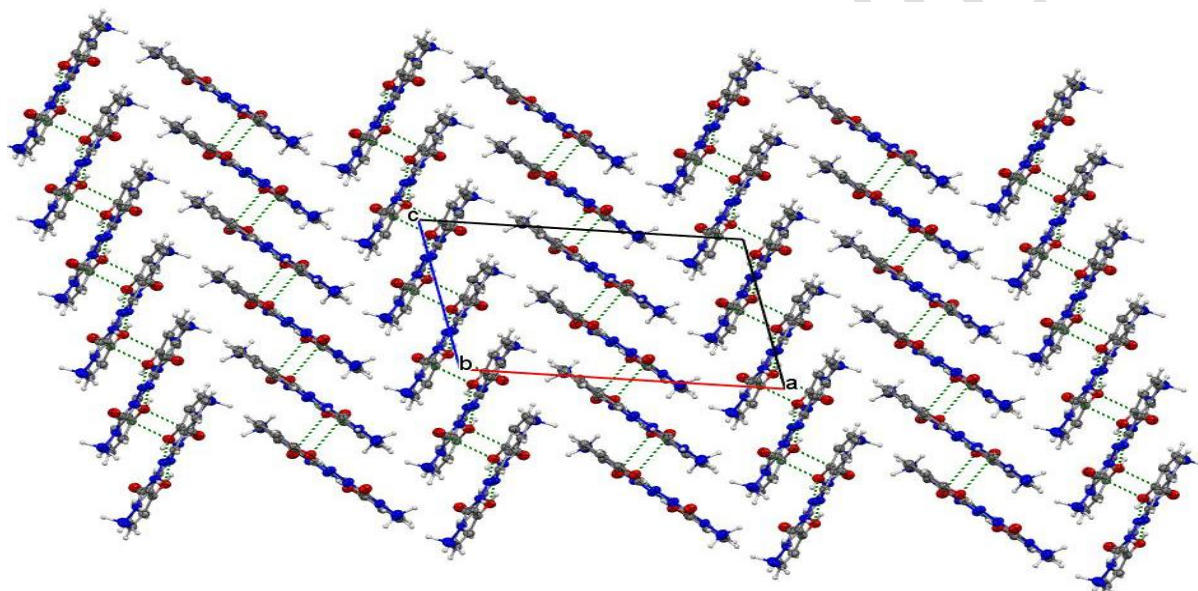


Figure 3: View of the Packing diagram of compound 1 along the b axis

Table 3. Hydrogen-bond geometry (Å, °) for compound 1

<i>D</i> —H... <i>A</i>	<i>D</i> —H	H... <i>A</i>	<i>D</i> ... <i>A</i>	<i>D</i> —H... <i>A</i>
O1—H1...N1	0.82	1.88	2.592 (3)	144
N2—H2...O1 <sup>i</sup>	0.86	2.17	3.021 (3)	168
N4—H4B...O2 <sup>ii</sup>	1.15 (5)	2.54 (5)	3.656 (4)	163 (3)
O3—H3...N5	0.82	1.85	2.573 (3)	146
N6—H6...O3 <sup>iii</sup>	0.86	2.07	2.925 (3)	176
C11—H11...N7 <sup>iv</sup>	0.93	2.55	3.435 (4)	160
N8—H8A...O4 <sup>v</sup>	1.27 (5)	2.58 (5)	3.507 (4)	128 (3)
N8—H8A...N5 <sup>vi</sup>	1.27 (5)	2.68 (5)	3.923 (4)	166 (3)

N8–H8B···O2<sup>v</sup> 1.14 (5) 2.53 (5) 3.587 (4) 154 (3)

Symmetry codes: (i)  $-x, y+1/2, -z+3/2$ ; (ii)  $x, -y+1/2, z-1/2$ ; (iii)  $-x+1, y+1/2, -z+1/2$ ; (iv)  $-x+1, y-1/2, -z+1/2$ ; (v)  $x+1, -y+1/2, z-1/2$ .

### 3.3 Description of the crystal structure of the compound 2

The compound **2** crystallizes in an orthorhombic system with C222<sub>1</sub> space group. The crystal structure is shown in Figure 4. The selected bond lengths and bond angles are listed in Table 4. The asymmetric unit contains one symmetrical carbohydrazone ligand molecule and one water molecule linked by a hydrogen bond of the type O<sub>water</sub>–H···O<sub>carbonyl</sub> [O3–H3A···O2]. The crystal structure reveals that the Schiff base compound **2** adopts the keto form, as shown by the bond length C1–O1 [1.234 (2) Å] which has a double bond character [52]. Moreover, the bond lengths N1–N2 [1.3686 (17) Å] and C2–N2 [1.281 (2) Å] have single bond and double bond character, respectively, (Table 4) [53, 54]. The oxygen atom O1 and the azomethine nitrogen atom N2 adopt a *cis* configuration with respect to the C1–O1 bond with a torsion angle 0.28° [N2–N1–C1–O1]. The mean plane of the carbonohydrazone moiety C=N–N–C(O)–N–N=C is twisted with respect to each mean plane of the phenyl rings with an angle value of 3.41(1)°. The value of the dihedral angle between the mean planes of the phenyl rings is 4.63(1)°. The torsion angle C2–N2–N1–C1 and the torsion angle defined by the related symmetry atoms (177.67(2)°) show that the central part of the molecule is almost linear. The molecule adopts an *E,E* configuration with respect to the bond C2–N2 and the bond defined by the related symmetry atoms. The crystal structure of compound **2** is stabilized by intramolecular O<sub>phenol</sub>–H···N<sub>carbohydrazone</sub> [O2–H2···N2] hydrogen bond resulting in S(6) rings and O<sub>water</sub>–H···O<sub>carbonyl</sub> [O3–H3A···O1]. The different molecules in the crystal are connected by intermolecular hydrogen bonds of type O<sub>water</sub>–H···N<sub>hydrazinyl</sub> [O3–H3B···N1<sup>i</sup>:  $i = x-1/2, y-1/2, z$ ]; O<sub>water</sub>–H···O<sub>phenolic</sub> [O3–H3B···O2<sup>ii</sup>:  $ii = x-1, y, z$ ] and N<sub>hydrazinyl</sub>–H···O<sub>water</sub> [N–H···O<sup>iii</sup>:  $iii = x+1/2, y+1/2, z$ ] (Table 5, Figure 5).

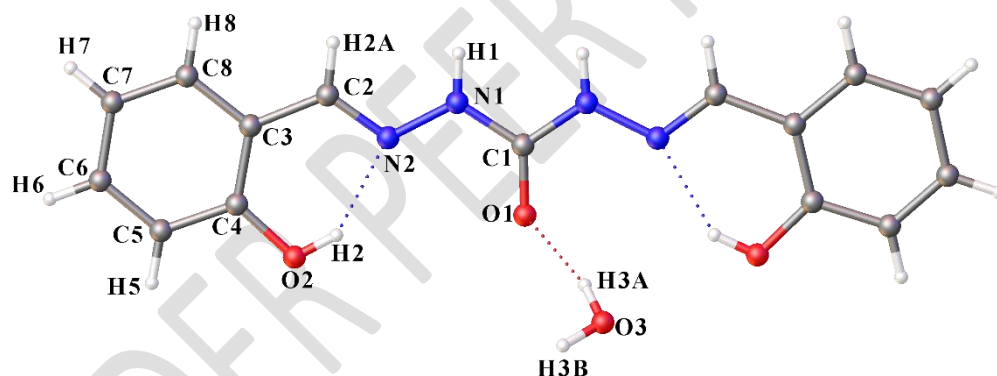


Figure 4. ORTEP view of compound **2**

Table 4. Selected bond distances [Å] and angles [deg] for compound **2**

2			
O1–C1	1.234 (2)	C3–C4	1.401 (2)
C3–C8	1.394 (2)	O2–C4	1.352 (2)
N2–C2	1.281 (2)	C6–C7	1.385 (3)
N1–C1	1.3549 (16)	C6–C5	1.363 (3)
C4–C5	1.395 (2)	N1–N2	1.3686 (17)
C8–C7	1.378 (3)	C2–C3	1.453 (2)
C1–N1–N2	119.27 (13)	C8–C3–C4	118.12 (15)

C2–N2–N1	116.34 (13)	O2–C4–C3	122.60 (15)
O1–C1–N1 <sup>i</sup>	123.62 (8)	O2–C4–C5	117.70 (17)
O1–C1–N1	123.62 (8)	C5–C4–C3	119.70 (18)
N1 <sup>i</sup> –C1–N1	112.77 (17)	C7–C8–C3	121.67 (17)
N2–C2–C3	121.86 (14)	C5–C6–C7	120.49 (18)
C4–C3–C2	122.68 (16)	C8–C7–C6	119.22 (19)
C8–C3–C2	119.19 (14)	C6–C5–C4	120.78 (19)

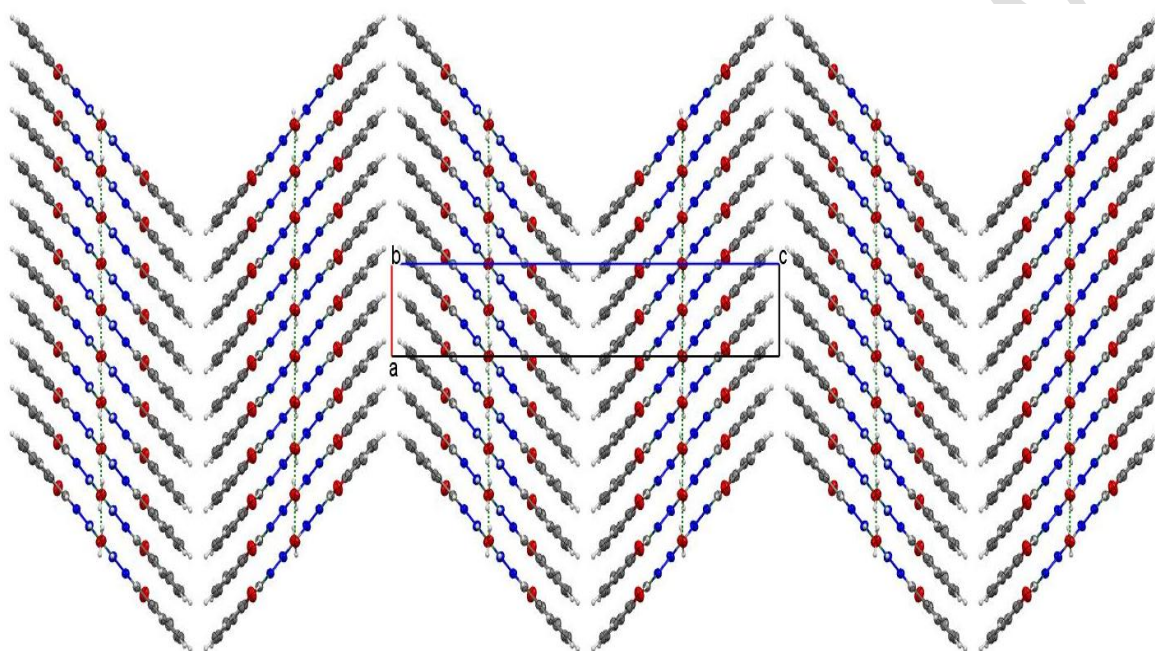


Fig. 5. View of the Packing diagram of compound 2 along the b axis

Table 5. Hydrogen-bond geometry (Å, °) for compound 2

<i>D</i> –H... <i>A</i>	<i>D</i> –H	H... <i>A</i>	<i>D</i> ... <i>A</i>	<i>D</i> –H... <i>A</i>
O3–H3A...O1	0.85	1.90	2.7385 (11)	168
O3–H3B...N1 <sup>i</sup>	0.85	2.39	2.8358 (19)	113
O3–H3B...O2 <sup>ii</sup>	0.85	2.53	3.1716 (16)	133
N1–H1...O3 <sup>iii</sup>	0.86	2.05	2.8358 (19)	152
O2–H2...N2	0.82	1.95	2.6695 (17)	145

Symmetry codes: (i)  $x-1/2, y-1/2, z$ ; (ii)  $x-1, y, z$ ; (iii)  $x+1/2, y+1/2, z$ .

#### 4. CONCLUSION

Monosubstituted 1-(2-hydroxybenzylidene)carbohydrazide **1** and disubstituted carbohydrazide derivatives 1,5-bis(2-hydroxybenzaldehyde)carbohydrazone **2** were prepared by 1/1 ratio

condensation and 1/2 ratio condensation reactions, respectively, between carbonohydrazide and 2-hydroxybenzaldehyde. The structures of the two compounds **1** and **2** were confirmed by elementary analyzes and FT-IR, <sup>1</sup>H and <sup>13</sup>C NMR spectroscopic techniques. The molecular structures of two the molecules **1** and **2** have been determined by of X-ray diffraction.

## 5. SUPPORTING INFORMATION

CCDC-2254010 and 2254011 contains the supplementary crystallographic data for this paper. These data can be obtained free of charge via <https://www.ccdc.cam.ac.uk/structures/>, or by e-mailing [data\\_request@ccdc.cam.ac.uk](mailto:data_request@ccdc.cam.ac.uk), or by contacting The Cambridge Crystallographic Data Centre, 12 Union Road, Cambridge CB2 1EZ, UK; fax: +44(0)1223-336033.

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