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Synthesis, spectroscopic studies and crystal structure of (*E*)-2-(2,4-dihydroxybenzylidene)thiosemicarbazone and (*E*)-2-[(1*H*-indol-3-yl)methylene]thiosemicarbazone

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ABSTRACT

Thiosemicarbazone Schiff bases (**1** and **2**) derived from 2,4-dihydroxybenzaldehyde, indoline-3-carbaldehyde and thiosemicarbazone have been synthesized and their structures were elucidated by elemental analysis, FT-IR, ^{1}H NMR, ^{13}C NMR and UV–visible spectroscopic techniques. The structures of compounds **1** and **2** have also been examined cyrstallographically. The title compounds **1** and **2** crystallize in the monoclinic space group $C2/c$ and triclinic space group $P\bar{1}$, with unit cell parameters: $a = 21.421(1)$ and $7.233(1)$, $b = 4.131(1)$ and $11.166(1)$, $c = 24.942(2)$ and $13.648(1)$ Å, $V = 1856.1(2)$ and $1019.5(1)$ Å^3, $D_x = 1.512$ and 1.422 g cm^{-3} and $Z = 8$ and 4, respectively.

1. Introduction

Thiosemicarbazones and their metal complexes are a broad class of biologically active compounds [1,2]. Due to this biological activity, there is considerable interest in metal complexes of heterocyclic thiosemicarbazones [3]. Spectral and structural investigations of a series of biologically active heterocyclic base adducts of copper(II) complexes of salicylaldehyde and 5-bromosalicylaldehyde thiosemicarbazones [4–6] have been studied. The thiosemicarbazone ligand usually coordinates with the metal through the imine nitrogen and sulfur atom. The ligands feature more than two covalent sites, the number of which depends on the aldehyde and on the tautomeric equilibrium of the thiosemicarbazone, although the most common way to coordinate is through the thiolate form [7]. Thiosemicarbazones are an important group of multidentate ligands with potential binding sites available for a wide variety of metal ions [8–10]. Tautomerism in Schiff bases with OH group in *ortho* position to the imino group both in solution and in solid state were investigated using spectroscopy and X-ray crystallography techniques [11–19]. Schiff bases with OH group in *ortho* position to the imino group are of interest mainly due to the existence of either O—H...N or O...H—N type of hydrogen bond and tautomerism between enol-imine and keto-amine form. In these compounds, short hydrogen bonds between the OH group in *ortho* position to the imino group and the imine nitrogen is due to the stereochemistry.

Although a series of thiosemicarbazone Schiff base complexes have been investigated crystallographically, there are only a few reports about free Schiff base ligands [20–35]. We report here the syntheses and characterization of two thiosemicarbazone Schiff base ligands derived from 2,4-dihydroxybenzaldehyde and indoline-3-carbaldehyde (Scheme 1). The structural analysis and tautomerism studies were carried out utilizing FT-IR, ^{1}H NMR, ^{13}C NMR, UV–visible spectroscopic and X-ray crystallographic techniques.

2. Experimental procedures

2.1. Reagents and techniques

The ^{1}H and ^{13}C NMR spectra were recorded on a Bruker DPX FT-NMR spectrometer operating at 400 and 101.6 MHz. Infrared absorption spectra were obtained from a Perkin-Elmer BX II spectrometer in KBr discs and were reported in cm^{-1} units. The UV–visible spectra were measured using a SHIMADZU 1208 series spectrometer. Carbon, nitrogen and hydrogen analyses were performed on a LECO CHNS-932 analyzer. Melting points were

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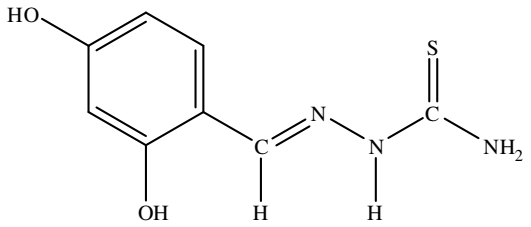
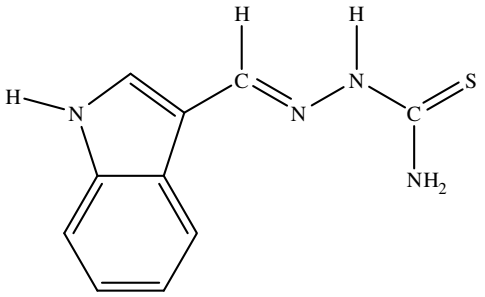


(*E*)-2-(2,4-dihydroxybenzylidene)thiosemicarbazone (**1**)



(*E*)-2-[(1*H*-indol-3-yl)methylene]thiosemicarbazone (**2**)

Scheme 1. Chemical formula of the title compounds.

determined on an Electro Thermal IA 9100 apparatus using a capillary tube. Thiosemicarbazide, indoline-3-carbaldehyde, 2,4-dihydroxybenzaldehyde, THF, DMSO were purchased from Merck (Germany).

2.2. Synthetic procedures

2.2.1. (E)-2-(2,4-dihydroxybenzylidene)thiosemicarbazone (*1*)

Thiosemicarbazide (0.91 g; 1.0×10^{-2} mol) was added to a dry THF (100 mL) solution of 2,4-dihydroxybenzaldehyde (1.38 g; 1.0×10^{-2} mol). The mixture was stirred and heated for 2 h. Compound **1** was obtained from the evaporation of THF. It was crystallized from chloroform/*n*-heptane as a yellow crystals, m.p. 191 °C, 1.84 g (87%) yield. Found: C, 45.39; H, 4.26; N, 19.90. Calc. for $C_8H_9N_3O_2S$; C, 45.49; H, 4.29; N, 19.89%. IR (KBr, cm^{-1}); *v*N—H; 3477–3339–3175 s, *v*Ar—H; 3055 w, *v*C=N; 1632 s, *v*C=C; 1556 s, *v*C—N; 1466 s, *v*C—O; 1317 s, *v*C=S; 1239. ^{1}H NMR (DMSO); δ ppm, 11.20 (s, 1H, Ar-O*H*); 9.78 (s, 2H, Ar-O*H* and N—*H*); 8.25 (s, 1H, Ar-C*H*= N—); 7.96 (s, 2H, —NH_2—); 7.76; 7.76–6.25 (m, 3H, Ar-*H*).

2.2.2. (E)-2-[(1H-indol-3-yl)methylene]thiosemicarbazone (*2*)

Thiosemicarbazide (0.91 g; 1.0×10^{-2} mol) was added to a dry THF (100 mL) solution of indoline-3-carbaldehyde (1.47 g; 1.0×10^{-2} mol). The mixture was stirred and heated for 2 h. Compound **2** was obtained from the evaporation of THF. It was crystallized from chloroform/*n*-heptane as a yellow crystals, m.p. 92 °C, 0.65 g (67%) yield. Found: C, 54.50; H, 5.49; N, 25.43. Calc. for $C_{10}H_{10}N_4S$; C, 55.05; H, 4.59; N, 25.68%. IR (KBr, cm^{-1}); *v*N—H; 3448–3310–3224 s, *v*Ar-H; 3042 m, *v*C—H; 2974–2922–2878 m, *v*C=N; 1611 s, *v*C=C; 1576 s, *v*C—N; 1441 s, *v*C=S; 1251. ^{1}H NMR (DMSO); δ ppm, 11.57 (s, 1H, N—*H*); 11.17 (s, 1H, N—*H*); 8.22 (d, 1H, —C*H*=N—); 7.43 (s, 2H, NH_2); 8.24–7.02 (m, 5H, Ar-H + C = CH).

2.3. Crystallography

The data collection for both compounds was performed on a STOE IPDS-2 diffractometer employing graphite-monochromatized Mo Kα radiation (λ = 0.71073 Å). Data collection, reduction and corrections for absorption and crystal decomposition for compound **1** and for compound **2** were achieved using X-AREA, X-RED software [36]. The structures were solved by SHELXS-97 and refined with SHELXL-97 [37,38]. The positions of the H atoms bonded to C atoms were calculated (C—H distance 0.96 Å), and refined using a riding model. The H atom displacement parameters were restricted to be $1.2U_{eq}$ of the parent atom. The crystal structures were solved by direct methods and refined by full-matrix least squares. The details of the X-ray data collection, structure solution and structure refinements are given in Table 1. Bond distances and angles are listed in Table 2. The molecular structure with the atom-numbering scheme is shown in Fig. 1 [39]. Crystallographic data (excluding structure factors) for the structure reported in this paper have been deposited with the Cambridge Crystallographic Data Centre as supplementary publication no. CCDC 691504 & 691505 [40].

Table 1
Crystal and experimental data

Compound	**1**	**2**
Formula	$C_8H_9N_3O_2S$	$C_{10}H_{10}N_4S$
Color/shape	Yellow/plate	Yellow/plate
Formula weight	211.24	218.28
Crystal system	Monoclinic	Triclinic
Space group	*C*2/*c*	$P\bar{1}$
Crystal dimension	0.25 × 0.30 × 0.35 mm^3	0.30 × 0.38 × 0.45 mm^3
Unit cell parameters	*a* = 21.421(1) Å *b* = 4.131(2) Å β = 122.6(1)° *c* = 24.942(2) Å	*a* = 7.232(1) Å α = 69.0(1)° *b* = 11.166(1) Å β = 85.3(1)° *c* = 13.648(1) Å γ = 82.4(1)°
V	1856.1(6) Å^3	1019.5(1) Å^3
Z	8	4
D_c (g cm^{-3})	1.512 g cm^{-3}	1.422 g cm^{-3}
μ (Mo Kα)	0.325 mm^{-1}	0.287 mm^{-1}
F (000)	880	456
$2\theta_{max}$	53.52°	55.14°
h, *k*, *l* range	$-26 \leqslant h \leqslant 26$ $-5 \leqslant k \leqslant 5$ $-31 \leqslant l \leqslant 30$	$-9 \leqslant h \leqslant 9$ $-14 \leqslant k \leqslant 14$ $-17 \leqslant l \leqslant 17$
No. of measured reflections	13406	19971
No. of independent reflections	1977	4699
No. of observed reflections	1662	3968
Goodness-of-fit on F^2	1.029	1.069
Measurement	STOE IPDS 2	STOE IPDS 2
Program system	STOE X-AREA	STOE X-AREA
Structure determination	SHELXS-97	SHELXS-97
Refinement method	Full-matrix least squares on F^2	Full-matrix least squares on F^2
R, R_w ($I > 2\sigma(I)$)	0.035, 0.086	0.032, 0.083
($\Delta\rho$)max, ($\Delta\rho$)min	0.201, −0.230 e Å^{-3}	0.267, −0.209 e Å^{-3}

3. Results and discussion

3.1. FT-IR, ^{1}H NMR, ^{13}C NMR and UV–visible spectroscopic studies

The FT-IR data of the compounds are given in synthetic procedures. Vibration bands with the wave numbers of 3477–3339–3175 and 3448–3310–3224 cm^{-1} (ν_{N-H}), 3055 and 3042 cm^{-1} (ν_{C-H}, Ar-H), 1632 and 1611 cm^{-1} ($\nu_{C=N}$), 1556 and 1576 cm^{-1} ($\nu_{C=C}$), 1239 and 1251 cm^{-1} ($\nu_{C=S}$) were observed for compounds **1** and **2**, respectively (Fig. 2). The stretching frequency observed at 2804, 2738 and 2811, 2733 cm^{-1} in **1** and **2** shows the presence of O—H...N and N—H...N intramolecular hydrogen bonds

Table 2
Bond lengths (Å), bond angles (°) and torsion angles (°) for compounds **1** and **2**

Compound ***1***			
S1—C8	1.698(2)	O2—C4	1.363(2)
N2—C8	1.327(2)	C1—C6	1.393(2)
N2—N1	1.389(2)	C1—C2	1.404(2)
N1—C7	1.281(2)	C1—C7	1.441(2)
C8—N3	1.317(2)	O1—C2	1.355(2)
C8—N2—N1	121.7(1)	N2—C8—S1	119.3(1)
C8—N2—H4	120.4(1)	O1—C2—C3	118.3(1)
N1—N2—H4	117.7(1)	O1—C2—C1	121.1(1)
C7—N1—N2	114.7(1)	N1—C7—C1	123.4(1)
N3—C8—N2	118.4(2)	N1—C7—H1	119.7(1)
N3—C8—S1	122.4(1)		
C8—N2—N1—C7	−163.8(2)	C7—C1—C2—O1	−5.2(3)
N1—N2—C8—N3	2.7(3)	N2—N1—C7—C1	−172.7(2)
N1—N2—C8—S1	−177.6(1)	C6—C1—C7—N1	−177.9(2)
C6—C1—C2—O1	179.4(2)	C2—C1—C7—N1	6.7(3)
Compound ***2***			
S1—C1	1.687(1)	S2—C11	1.693(1)
N3—C2	1.280(2)	N6—C11	1.333(2)
N3—N2	1.386(1)	N6—N7	1.383(1)
C3—C4	1.372(2)	N7—C12	1.278(2)
C3—C2	1.432(2)	N8—C14	1.357(2)
C3—C10	1.438(2)	N8—C15	1.370(2)
N2—C1	1.332(2)	C12—C13	1.437(2)
N1—C1	1.330(2)	C13—C14	1.368(2)
N4—C4	1.350(2)	C13—C20	1.439(2)
N4—C5	1.371(2)	C11—N5	1.323(2)
C2—N3—N2	114.5(1)	N4—C4—C3	110.2(1)
C4—C3—C2	123.8(1)	C11—N6—N7	122.1(1)
C4—C3—C10	106.3(1)	N7—N6—H6A	119.1(1)
C2—C3—C10	129.8(1)	C12—N7—N6	113.4(1)
C1—N2—N3	120.1(1)	C14—N8—C15	109.2(1)
N3—C2—C3	122.5(1)	N7—C12—C13	123.3(1)
N3—C2—H2	119.0(1)	N7—C12—H12	119.3(1)
C3—C2—H2	118.4(1)	N5—C11—N6	117.6(1)
C4—N4—C5	109.4(1)	N5—C11—S2	123.4(1)
N1—C1—N2	117.1(1)	N6—C1—S2	119.0(1)
N1—C1—S1	122.7(1)	N8—C14—C13	110.3(1)
N2—C1—S1	120.3(1)	N8—C14—H14	120.7(1)
C2—N3—N2—C1	173.2(1)	C2—C3—C10—C5	−177.2(1)
N2—N3—C2—C3	177.8(1)	C11—N6—N7—C12	−178.6(1)
C4—C3—C2—N3	178.3(1)	N6—N7—C12—C13	−178.5(1)
C10—C3—C2—N3	−4.9(2)	N7—C12—C13—C14	−179.7(1)
N3—N2—C1—N1	1.0(2)	N7—C12—C13—C20	1.5(2)
N3—N2—C1—S1	−178.2(1)	N7—N6—C11—N5	0.7(2)
C4—N4—C5—C6	−177.1(1)	N7—N6—C11—S2	−178.5(1)
C5—N4—C4—C3	−0.7(2)	C14—N8—C15—C20	0.9(1)
C2—C3—C4—N4	177.8(1)	C19—C20—C15—N8	−180.0(1)
C10—C3—C4—N4	0.3(2)	C15—N8—C14—C13	−1.3(2)

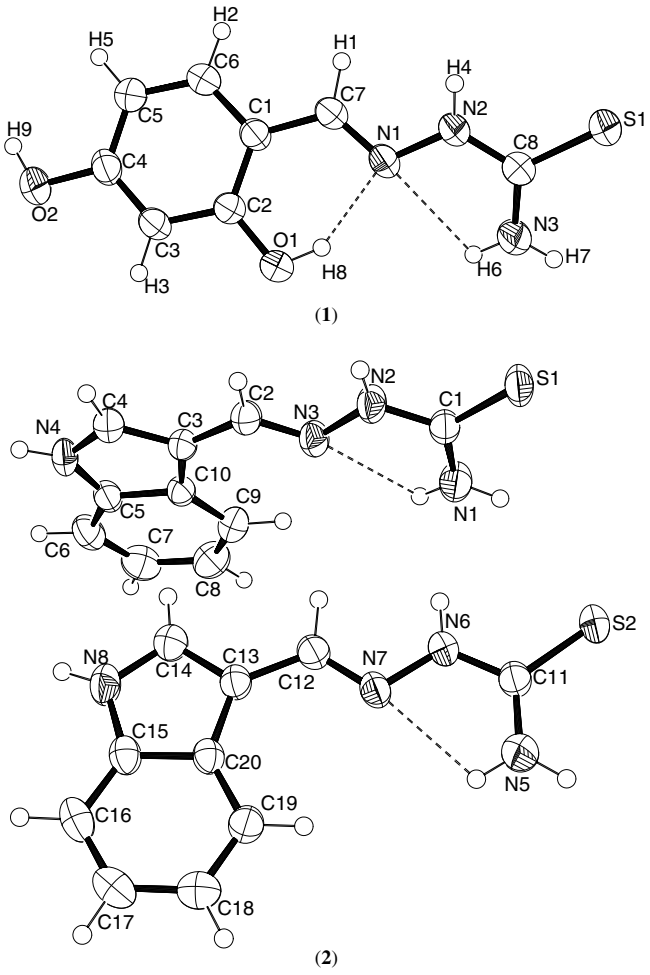


Fig. 1. The molecular structure of the compounds **1** and **2**.

[41,11]. The C=N bond which is accountable partially for the existence enol-imine form can also be inferred from the FT-IR spectra of compound **1**. The *ν*C—OH, *ν*C—NH and *ν*C—NH_2 vibrations band overlap with each other in compound **1**. The compound **1** with strong band at 1286 cm^{-1} possesses highest percentage of enol-imine tautomer due to the stabilization of phenolic C—O bond [42].

The ^{1}H NMR data for compound **1** show that the tautomeric equilibrium favours the enol-imine in DMSO. The OH protons are observed 11.20 and 9.78 ppm singlets for compound **1**. The azomethine protons are observed as singlets 8.25 ppm singlets and 8.34 ppm singlets for **1** and **2** (Fig. 3). The amine protons resonate at δ = 9.78, 7.96 ppm and δ = 11.57, 11.17, 7.43 ppm singlets, respectively, for compounds **1** and **2**. The phenyl protons of the compounds **1** and **2** gave a multiplets at δ = 7.76–6.25 ppm and δ = 8.24–7.02 ppm, respectively.

According to the ^{13}C NMR spectra compounds **1** and **2** have 8 and 10 signals (Fig. 4). ^{13}C NMR chemical shifts are given in Scheme 2 for compounds **1** and **2**.

The UV–visible studies of the compounds **1** and **2** were done in DMSO solvent (Fig. 5). The Schiff bases show absorption in the range greater than 400 nm in polar and nonpolar solvents [11–13]. It is point out that the new band belongs to the keto-amine form of the Schiff bases with OH group in ortho position to the imino group in polar and nonpolar solvents in both acidic and basic media [11–13,16–19]. The compound **1** showed no absorption above 400 nm in DMSO. So the enol-imine tautomer is dominant only in the DMSO solution for compound **1**. In conclusion, UV–visible, ^{1}H NMR and ^{13}C NMR results show that the compound **1** exist in the enol-imino form in DMSO solution.

3.2. Crystallographic study

Schiff base ligands consist of a variety of substituents with different electron-donating and electron-withdrawing groups, and therefore may have interesting electro-chemical properties. The Schiff bases compounds have been also under investigation during last years because of their potential applicability in optical communications and many of them have NLO behavior [43–45].

Conjugated organic molecules containing both donor and acceptor groups are of great interest for molecular electronic devices. Second-order NLO organic materials that contain stable molecules with large molecular hyperpolarizabilities in noncentrosymmetric packing are of great interest for device applications [46], but according to a statistical study, an overwhelming majority of achiral molecules crystallize centrosymmetrically.

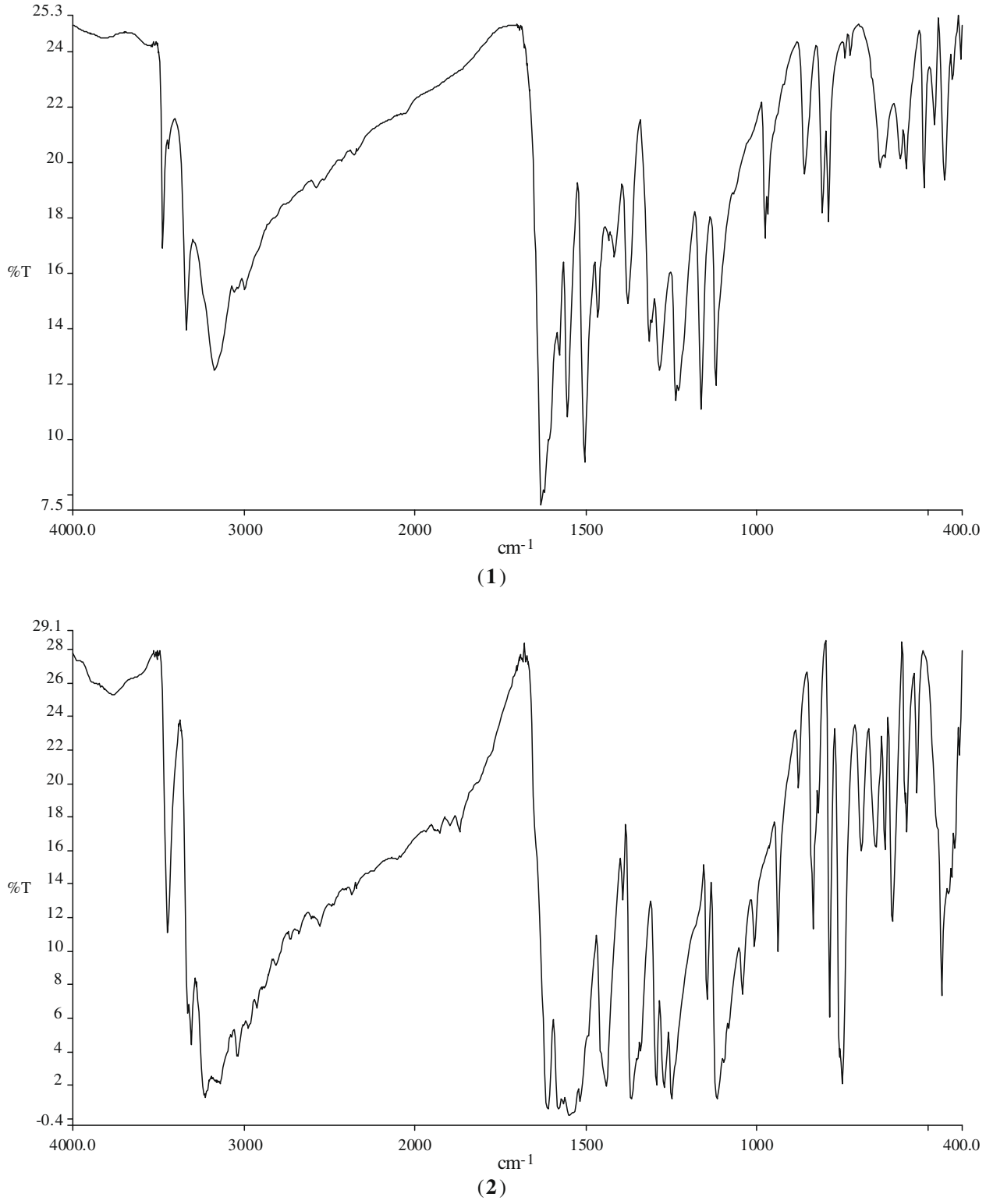


Fig. 2. FT-IR spectra of the compounds **1** and **2**.

The title molecules **1** and **2** are not planar. For **1**, the two Schiff base moieties (C1—C7, O1, O2) [planar with a maximum deviation of 0.072(1) Å for the C7 atom] and (N1—N3, C8, S1) [planar with a maximum deviation of 0.021(1) Å for the N2 atom] are inclined at angle of 27.5(1)°. The compound **2** has two independent molecules in asymmetric unit. The two planar phenyl rings bridged by the C=N imino moiety in the two cyrstallographically independent molecules, however, are inclined at angle of 11.3(1)° and 4.2(1)°, respectively.

The crystal structures are stabilized by intramolecular and intermolecular hydrogen bonding and their geometrical details are listed Table 3 [42,47,48]. Intramolecular hydrogen bonds occur between N3—H6...N1 [2.683(3) Å], O1—H8...N1 [2.683(2) Å] atoms for the molecule **1** and N1—H1B...N3 [2.633(2) Å], N5—H5B...N7

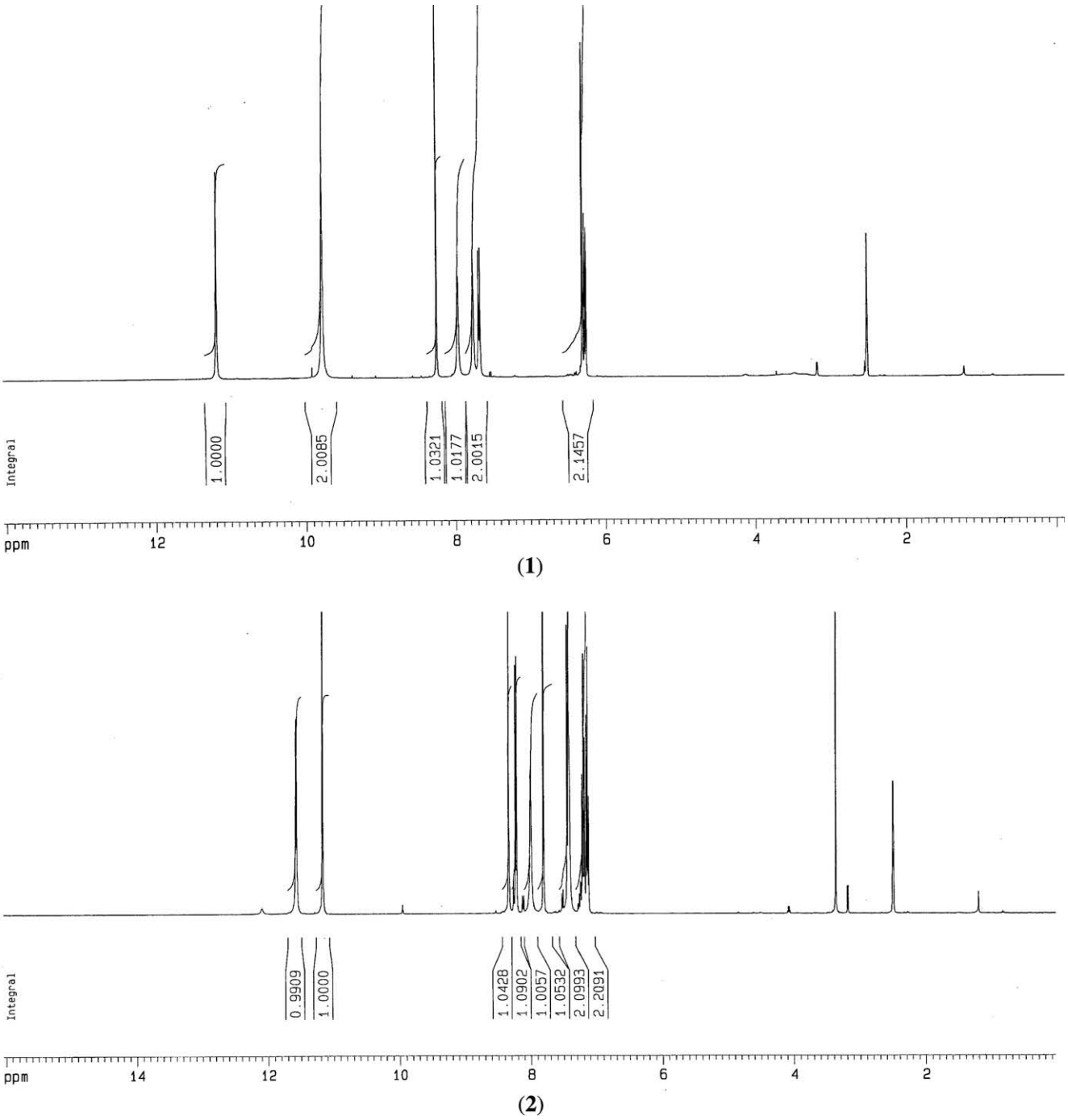


Fig. 3. ^{1}H NMR spectra of the compounds **1** and **2**.

[2.682(2) Å] atoms for the molecule **2** (Fig. 1). The sum of the Van der Waals radius of the O and N atoms (3.07 Å) is significantly longer than the intramolecular O...N hydrogen bond length [49]. There is also intermolecular hydrogen bond between N2...S1 [3.400(2) Å], N3...O1 [2.972(2) Å], and O2...S1 [3.290(2) Å] for the molecule **1** and N2 and S1 [3.401(1) Å], N4...S1 [3.355(1) Å], N8...S2 [3.399(2) Å], atoms of neighbouring molecules for the compound **2** (Fig. 6).

The thiosemicarbazone moiety in both compounds shows an *E* configuration about C1—N2 and C2—N3 or C11—N6 and C12—N7. The angles between the mean planes of the indoline and dihydroxybenzylidene ring and thiosemicarbazone moiety present a significant difference in the two compounds. The C=S bonds [1.699(2) and 1.687(1) Å] have a length intermediate between a single and double bond for **1** and **2**. The sum of the valence angles around atoms N2 and N3 and N6 and N5 or N2 and N1 indicate that these atoms are sp^2 hybridized. The N3—C8—N2 and N5—C11—N6 or N1—C1—N2 angles [118.4(2) and 117.6(1) or 117.1(1)°] being narrow than N3—C8—S1 and N5—C11—S2 or N1—C1—S1 [122.4(1) and 123.4(1) or 122.7(1)°], as observed in compounds **1** and **2**. This may be due to the intramolecular hydrogen bonding between the free NH_2 group and the imine nitrogen. The S1—C1—N2—N3 [−177.6(1)°] and N3—C2—C3—C4 [−178.1(2)°] torsion angles indicate a *trans* conformation with respect to the thiosemicarbazone moiety and the phenyl ring. The *trans* conformation adopted by the side chain is evident from the values of the C1—N2—N3-—C2

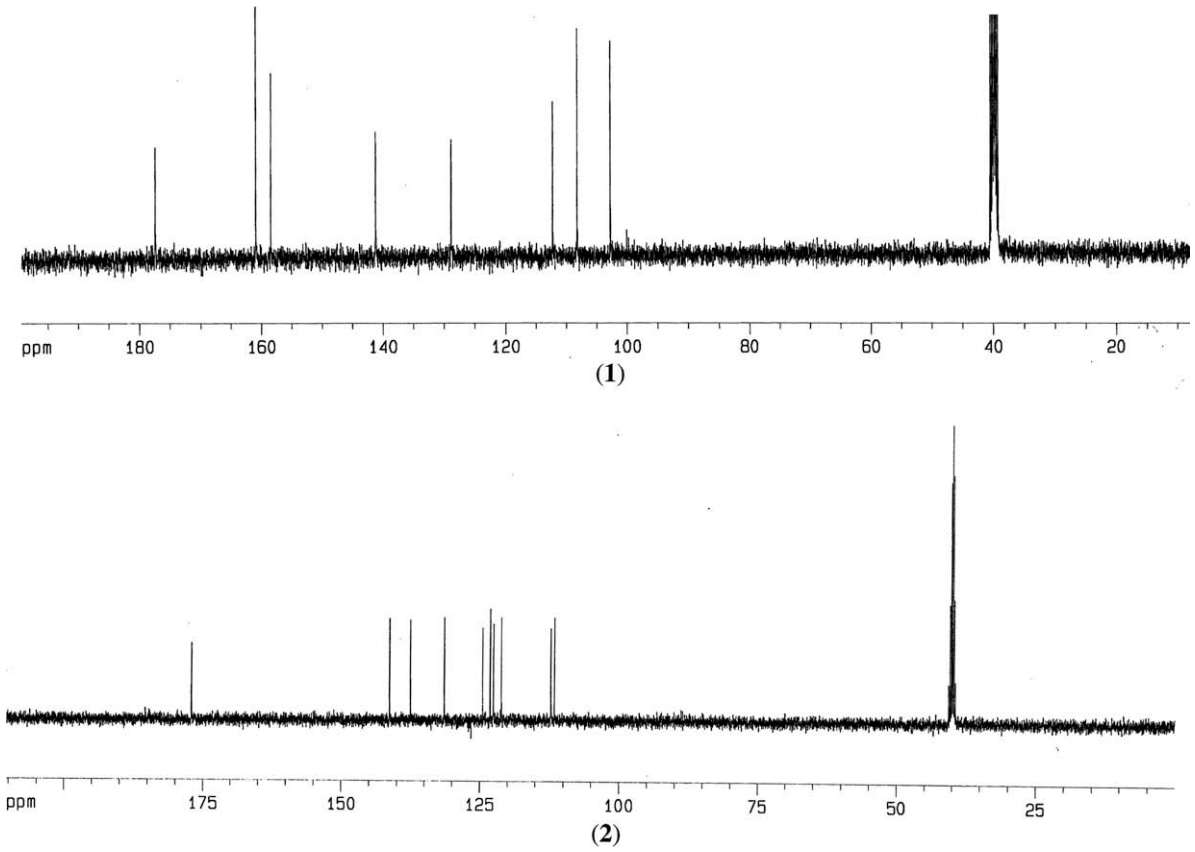
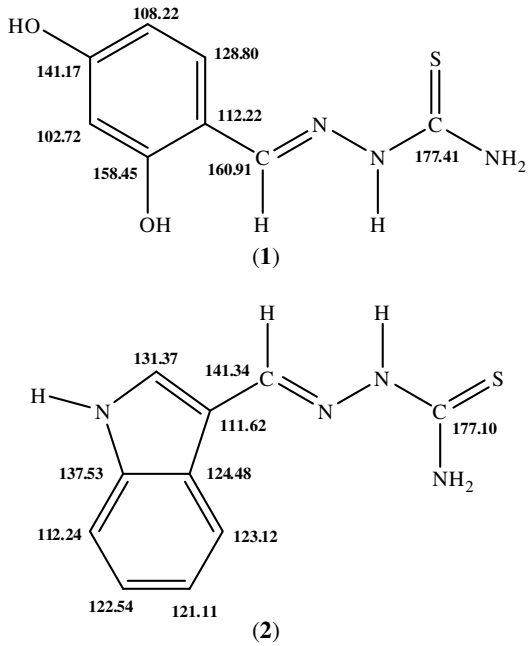


Fig. 4. ^{13}C NMR spectra of the compounds **1** and **2**.



Scheme 2. ^{13}C NMR Chemical shifts of the compounds in solution.

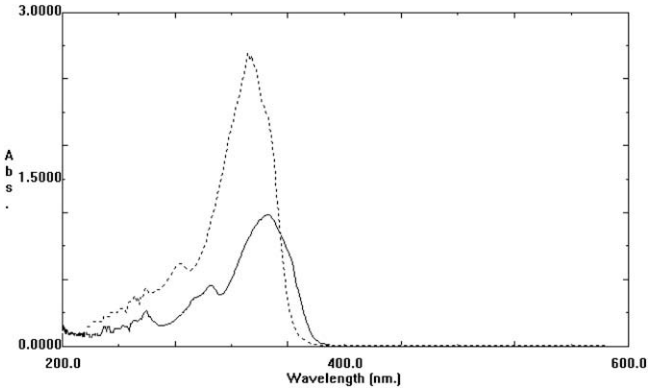


Fig. 5. UV–vis spectra of the compounds **1** and **2** in the DMSO, compound **1** (—), compound **2** (- - -).

[172.8(2)°], N2—N3—C2—C3 [−178.6(1)°], N3—C2—C3—C4 [−178.1(2)°] and C2—C3—C4—C5 [−178.4(2)°] torsion angles.

In compound **1** C—N group seems to have a strong electron-withdrawing character. Thus, the O1—C2 bond distance of 1.355(2) Å is also consistent with the C—O single bonding. The C=O bond distance indicates the presence of the keto form, with a partial double bond character of the CO group (>C=O ↔ C^{+}—O^{-}). X-ray structure determinations reveal that the enol-imine tautomer is favored over the keto-amine tautomer for the compound. This is evident from the observed O1—C2 bond distance of 1.355(2) Å, which is consistent with the O—C single bond; similarly the N1—C7 distance of 1.281(2) Å is also consistent with the N=C double bonding. From the X-ray diffraction experiment and spectroscopic studies, we were only able to detect the existence of the enol-imine tautomer for compound **1**.

In compound **2** the N2—C1 and N1—C1 bond distances are 1.332(2) and 1.330(2) Å while the N3—C2 distance is 1.280(2) Å. This is consistent with the N2—C1 and N1—C1 single bond; similarly is also consistent with the N3=C2 double bonding.

Table 3
Geometric details of intra- and inter-molecular hydrogen bonding for the title compounds

D—H...A (Å)	D—H	H...A (Å)	D...A (Å)	∠D—H...A (°)
For compound ***1***				
N3—H6...N1	0.83(3)	2.37(3)	2.683(3)	103(2)
O1—H8...N1	0.87(4)	1.90(3)	2.683(2)	149(3)
N2—H4...S1^{i}	0.87(2)	2.56(2)	3.400(2)	163(2)
N3—H6...O1^{ii}	0.83(3)	2.23(3)	2.972(2)	149(3)
O2—H9...S1^{iii}	0.87(3)	2.43(3)	3.290(2)	171(3)
For compound ***2***				
N1—H1B...N3	0.88(2)	2.31(2)	2.633(2)	101.9(2)
N5—H5B...N7	0.86(2)	2.32(2)	2.682(2)	105.9(2)
N2—H2A...S1^{iv}	0.85(2)	2.56(2)	3.401(1)	171.2(2)
N4—H4A...S1^{v}	0.83(2)	2.56(1)	3.355(1)	160.0(2)
N8—H8A...S2^{v}	0.84(2)	2.60(2)	3.399(1)	158.9(2)

Note: D, donor; A, acceptor. Symmetry transformation used to generate equivalent atoms: (i) ½ − *x*, −1/2 − *y*, 1 − *z*; (ii) −*x*, *y*, ½ − *z*; (iii) −1/2 + *x*, 3/2 + *y*, *z*; (iv) 1 − *x*, 1 − *y*, −*z*; (v) *x*, 1 + *y*, *z*.

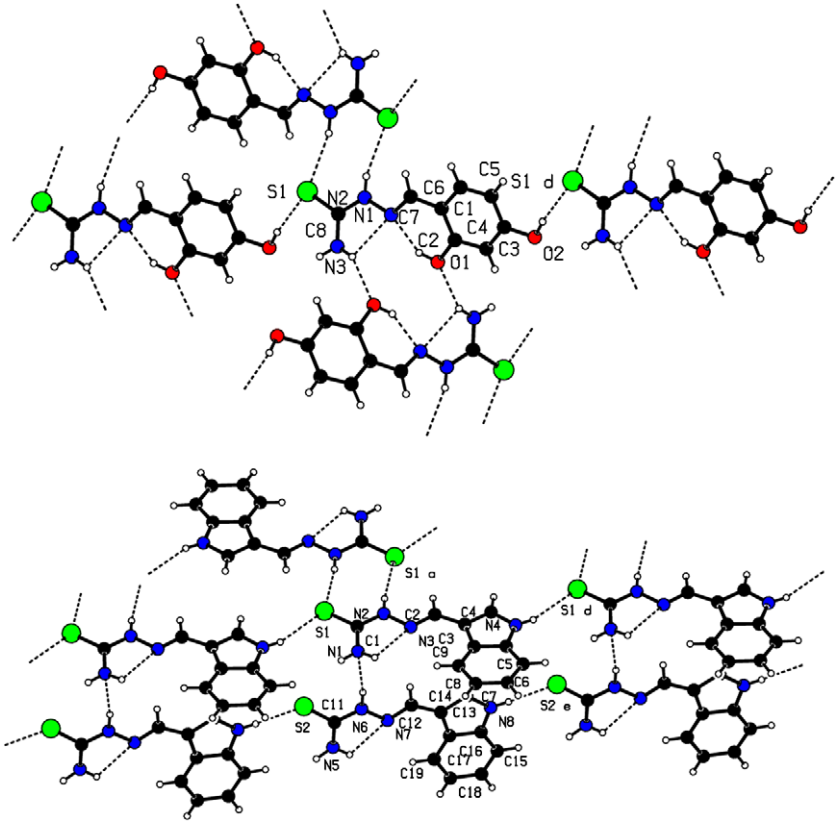


Fig. 6. In the crystal structure a perspective view of the molecules **1** and **2**, respectively. The intermolecular and intramolecular hydrogen bonds have been indicated by dashed lines.

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