

Synthesis of Schiff Bases Compounds from Oxamic Hydrazide: Spectroscopic Characterization, X-ray Diffraction Structure and Antioxidant Activity Study

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Abstract: The compounds (*E*)-2-amino-*N'*-(1-(2-hydroxyphenyl)ethylidene)-2-oxoacetohydrazide (I) and (*E*)-*N'*-(2-hydroxy-3-methoxybenzylidene)-2-amino-2-oxoacetohydrazide (II) were synthesized by the 1:1 ratio condensation reaction of oxamic hydrazide and 2-hydroxyacetophenone or *o*-vanillin respectively. The two compounds were characterized by physico-chemical analyses, elemental analysis, FTIR, ¹H and ¹³C NMR spectroscopies techniques. The structure of the compound (I) was determined by single-crystal X-ray diffraction study. The compound (I) (C₁₀H₁₁N₃O₃) crystallises in the triclinic space group *P*-1 with the following unit cell parameters: *a* = 7.0399 (5) Å, *b* = 8.6252 (8) Å, *c* = 9.5474 (9) Å, *a* = 81.730 (3)°, *b* = 72.738 (3)°, *g* = 67.450 (3)°, *V* = 510.99 (8) Å³, *Z* = 2, *T* = 173 (2) K, *m* = 0.11 mm⁻¹, *D*_{calc} = 1.438 g/cm³, *R*_{int} = 0.028, *R*_{sigma} = 0.073. The oxamic hydrazide moiety of the molecule is slightly twisted as reflected by the torsion angles values of 177.2 (2)° [N1-N2-C9-C10], -171.3 (3)° [N2-C9-C10-N3], -4.6 (4)° [O2-C9-N2-N1] and 8.4 (4)° [O3-C10-C9-N2]. The intramolecular hydrogen bond O1(phenol)-H1...N1(hydrazide) which close in *S* (6) ring stabilized the conformation. The intermolecular hydrogen bonds, C3-H3...O1ⁱ(phenol) (i: -x+1, -y, -z+1), N3(amide)-H3A...O3ⁱⁱ(amide) (ii: -x+1, -y+2, -z) and N3(amide)-H3B...O2ⁱⁱⁱ(hydrazide) (iii: -x+1, -y+1, -z) lead to the formation of sheets parallel to *ac* plane. Compounds (I) and (II) showed antioxidant activities less than 10% inhibition of DPPH.

Keywords: Oxamic Hydrazide, 2-hydroxyacetophenone, *O*-vanillin, Antioxidant, X-ray

1. Introduction

Potentially ditopic Schiff bases prepared from oxamic or thioxamic hydrazide have been widely reported in the literature by chemists and in particular those interested in coordination chemistry [1-6]. Oxamic hydrazide has two different arms. The hydrazide arm condenses more easily than the amide arm with carbonyl [7-10]. The use of these ligands has made it possible to prepare coordination compounds with atypical structures [11-13]. The derivatives of oxamic hydrazide have been used for the synthesis of

nucleoside [14, 15]. Oxamic hydrazide have been used as co-reactant in electroluminescence [16, 17]. A wide variety of heterocyclic molecules with good medicinal properties are obtained starting from these oxamic precursors [18]. They are used as antibacterial agent, [19] anticonvulsant, [8] anti-inflammatory [20], antituberculosis [18] and anticancer agents [4, 21]. Schiff's bases are synthesized from oxamic hydrazide and used for the preparation of complexes with transition metal and lanthanide ions [9, 22-25]. The properties of these complexes are evaluated in superoxide catalysis [2, 3] and magnetism [26]. It is in this perspective that we have studied these types of ligands and reported their

transition metal complexes [7, 27]. Continuing our work in this field, we obtained ligands (I) and (II). In the present study, we report the spectroscopic study of the two compounds and the structure of (I) obtained by X-ray diffraction.

2. Material and Methods

2.1. Materials and Physical Methods

Oxamic hydrazide, 2'-hydroxyacetophenone, o-vanillin, cyclohexanol and 1,1-diphenyl-2-picrylhydrazyl (DPPH.) were of analytical reagent grade and were obtained from Sigma-Aldrich Company. All used solvents were of UV spectroscopic quality. The elemental analyses of C, H and N were recorded on a VxRio EL Instrument. FT-IR spectra were recorded in the region of 4000–400 cm^{-1} using a Perkin Elmer Spectrum Two FT-IR spectrometer. The UV-Visible spectra were recorded on a Perkin Elmer Lambda UV-Vis spectrophotometer. The ^1H and ^{13}C NMR spectra were recorded in DMSO- d_6 on a Bruker 500 MHz spectrometer at room temperature using TMS as an internal reference.

2.2. Free Radical Scavenging Antioxidant Assay

Antioxidant capacities of compound (I) were measured according to Akhtar et al. [28] method with modifications. The methanol solution of 3.8 mL DPPH• was added to test compounds (200 μL) at different concentrations. The mixture was shaken vigorously and incubated in dark for 30 min at room temperature. After the incubation time, the absorbance of the solution was measured at 517 nm by using UV-vis spectrophotometer Perkin two. The DPPH• radical scavenger effect was calculated using the following equation:

$$\text{Scavenging activity (\% control)} = \frac{A_{\text{control}} - A_{\text{sample}}}{A_{\text{control}}} \times 100$$

where A_{control} is the absorbance of the control reaction and A_{sample} is the absorbance of the test compound. The tests were carried out in triplicate. Trolox was used as positive control.

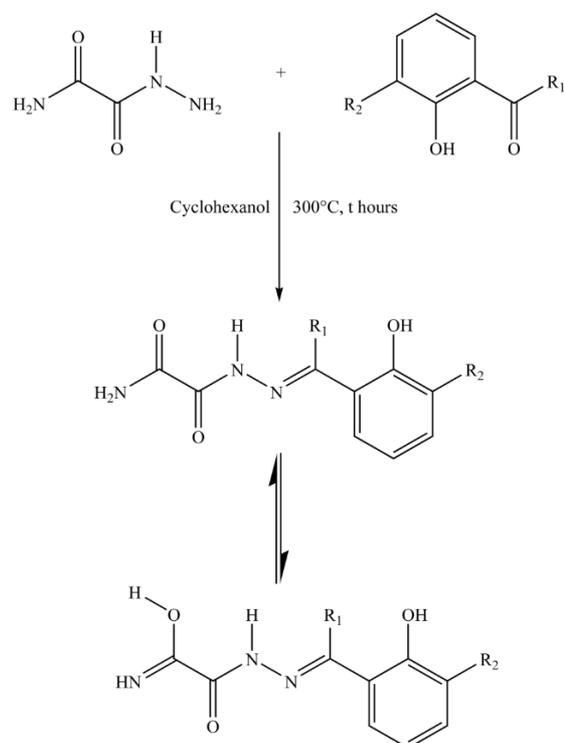
2.3. Synthesis of (E)-2-amino-N'-(1-(2-hydroxyphenyl)ethylidene)-2-oxoacetohydrazide (I)

Oxamic hydrazide (2.0307 g, 19.7 mmol) was suspended in 20 mL of cyclohexanol. 2'-hydroxyacetophenone (2.6821 g, 19.7 mmol) previously dissolved in 20 mL of cyclohexanol was added. The mixture was heated at reflux for 24 hours. On cooling, a white solid was collected by filtration, washed with 2 x 10 mL of ethanol, and dried in the open air. The precipitate was recrystallized in DMF and the solution was left at room temperature. Slow evaporation of the solvent gave colorless crystals after one week. Yield: 95%. Anal. Calc. for $[\text{C}_{10}\text{H}_{11}\text{N}_3\text{O}_3]$ (%): C, 54.29; H, 5.01; N, 19.00. Found: C, 54.27; H, 5.03; N, 19.03. FIT-IR (cm^{-1}): 3383(OH), 3293 (NH), 3222 (NH₂), 1702 (CONH), 1652 (CONH₂), 1606 (C=N), 1562 (C_{Ar}=C_{Ar}), 1466 (C_{Ar}=C_{Ar}), 1411 (C_{Ar}=C_{Ar}), 1250 (C-O_{phenol}). ^1H RMN δ : 2.424 (S, 3H, CH₃), 6.921–8.056 (m, 4H, H_{Ar}), 8.396 (Ar-OH), 9.940 (S,

1H, CONH), 11.428 (S, 1H, HNCOH), 12.925 (S, 1H, HNCOH). RMN ^{13}C (dmsO- d_6 , d (ppm)): 162.232 (C=O), 159.198 (HN-COH), 158.840 (C=N), 157.840 (C_{Ar}-OH), [132.344, 129.416, 119.607, 119.231, 117.812] (C_{Ar}), 14.677 (-CH₃),

2.4. Synthesis of (E)-N'-(2-hydroxy-3-methoxybenzylidene)-2-amino-2-oxoacetohydrazide (II)

Oxamic hydrazide (2.037 g, 19.7 mmol) was suspended in 20 mL of cyclohexanol. o-vanillin (2.9973 g, 19.7 mmol) previously dissolved in 20 mL of cyclohexanol was added. The mixture was heated at reflux for six hours. On cooling, a solid was collected by filtration, washed with 2 x 10 mL of ethanol and dried in the open air. The precipitate was recrystallized in DMF and the solution was left at room temperature. Slow evaporation of the solvent gave colorless precipitate after one week. Yield: 97%. Anal. Calc. for $[\text{C}_{10}\text{H}_{11}\text{N}_3\text{O}_4]$ (%): C, 50.63; H, 4.67; N, 17.71. Found: C, 50.60; H, 4.69; N, 17.75. FT-IR (cm^{-1}): 3375 (N-H), 3221 (O-H), 1704 (CONH), 1656 (C=O), 1603 (C=N), 1560 (C_{Ar}=C_{Ar}), 1464 (C_{Ar}=C_{Ar}), 1409 (C_{Ar}=C_{Ar}), 1252 (C-O_{phenol}), 1230 (C-O_{ether}), 1080 (N-N), 837 (C-H_{Ar}), 669 (C-H_{Ar}). RMN ^1H (dmsO- d_6 , d (ppm)): 3.80 (S, 3H, -OCH₃), [6.82–7.10] (m, 3H, H_{Ar}), 7.97 (S, 1H, CONH), 8.30 (S, 1H, Ar-OH), 8.75 (S, 1H, HC=N), 10.75 (S, 1H, HNCOH), 12.36 (S, 1H, HNCOH). RMN ^{13}C (dmsO- d_6 , d (ppm)): 161.51 (C=O), 156.54 (HN-COH), 150.46 (C=N), 147.87 (C_{Ar}-OH), 147.20 (C_{Ar}-OCH₃), [120.65, 119.03, 118.72, 113.96] (C_{Ar}), 55.76 (-OCH₃).



I: R₁ = -CH₃, R₂ = H and t = 24 hours; II: R₁ = H, R₂ = -OCH₃ and t = 6 hours

Figure 1. Synthetic scheme of (I) and (II).

2.5. Crystal Structure Determination

Crystals suitable for single-crystal X-ray diffraction, of the reported compound, was grown by slow evaporation of DMF solution of the compound. Details of the crystal structure solution and refinement are given in Table 1. Diffraction data were collected using a Bruker APEX-II CCD diffractometer with graphite monochromatized MoK α radiation ($\lambda = 0.71073 \text{ \AA}$). All data were corrected for Lorentz and polarization effects. The structure was solved and refined using the Bruker SHELXTL Software Package [29]. All the structures were refined on F^2 by a full-matrix least-squares procedure using anisotropic displacement parameters for all non-hydrogen atoms [30]. H atoms of the NH groups was located in the Fourier difference maps and refined without restraints. Other H atoms were geometrically optimized and refined as riding on their carriers with $U_{\text{iso}}(\text{H}) = 1.2U_{\text{eq}}(\text{C})(1.5 \text{ for } \text{CH}_3 \text{ group})$. Molecular graphics were generated using ORTEP-3 [31].

3. Result and Discussion

3.1. General Study

The synthesis of Schiff bases usually takes place in simple alcohols such as methanol, ethanol or propanol. In the synthesis of Schiff bases from oxamic hydrazide, the subject of our study, the use of these solvents leads to excessively long reaction times. In fact, for the condensation of oxamic hydrazide with carbonyl compounds, it is necessary to heat to temperature high enough to shorten the reaction time. Cyclohexanol which has a high boiling point (161.8°C) is suitable to prepare compounds (I) and (II) with short time reaction (Figure 1).

The results of elemental analysis agree with the expected formulas for the two compounds. The solid-state infrared spectrum of (I) reveals a broad band around 3383 cm^{-1} attributed to the OH stretching vibration and another band around 3293 cm^{-1} indicating the presence of NH. These two bands are present in the spectrum of compound (II) at 3375 cm^{-1} and 3221 cm^{-1} respectively [32, 33]. The band due to the C=N group formed after the condensation reaction between the oxamic hydrazide and the appropriate carbonyl is pointed at 1606 cm^{-1} for (I) and at 1603 cm^{-1} for (II). The stretching vibrations due to C=O of the oxamic unit were noted at 1702 cm^{-1} and 1652 cm^{-1} for (I) and at 1704 cm^{-1} and 1656 cm^{-1} for (II) [34, 35]. The shift of the second band towards the low frequencies is justified by the strong resonance of the oxalate group. The additional bands in the range [$1570\text{--}1405 \text{ cm}^{-1}$] are due to the aromatic groups.

The ^1H NMR spectra of the compounds, in DMSO- d_6 solution, are recorded. Compound (I) gives two signals characteristic of iminolisation. Indeed, the single signal designating the moiety [$-\text{C}(=\text{O})-\text{NH}_2$] does not appear on the spectrum. The two signals at 11.428 ppm and 12.925 ppm assigned respectively to $\text{HN}=\text{C}-\text{OH}$ and $\text{HN}=\text{C}-\text{OH}$ are indicative of the iminolisation of the amide function of the Schiff base. The same phenomenon is observed for

compound (II). The corresponding signals are pointed at 10.750 ppm and 12.360 ppm, respectively. These observations are confirmed by ^{13}C NMR spectra in DMSO- d_6 . Compound (I) gives a signal at 159.198 ppm corresponding to the iminol carbon atom $\text{HN}=\text{C}-\text{OH}$. This signal is identified at 156.540 ppm in the spectrum of (II). The signals due to the hydrazide carbon atoms are at 162.232 ppm and 161.510 ppm for (I) and (II) respectively. This behavior is observed in amide-iminol tautomerism [36].

Table 1. Crystal data and structure refinement for compound (I).

Chemical formula	$\text{C}_{10}\text{H}_{11}\text{N}_3\text{O}_3$
M_r	221.22
Crystal shape/Color	Prismatic/colorless
Crystal system, space group	Triclinic, $P-1$
T (K)	173 (2)
a (\AA)	7.0399 (5)
b (\AA)	8.6252 (8)
c (\AA)	9.5474 (9)
a ($^\circ$)	81.730 (3)
b ($^\circ$)	72.738 (3)
g ($^\circ$)	67.450 (3)
V (\AA^3)	510.99 (8)
Z	2
D_{calc} (g cm^{-3})	1.438
$F(000)$	232
Radiation type	Mo $K\alpha$
μ (mm^{-1})	0.11
Crystal size (mm)	0.12 x 0.10 x 0.10
$T_{\text{min}}, T_{\text{max}}$	0.982, 0.991
h	$-7 \rightarrow 8$
k	$-11 \rightarrow 11$
l	$-12 \rightarrow 12$
Diffractometer	Bruker APEX-II CCD
Absorption correction	Multi-scan SADABS
No. of measured, independent and observed [$I > 2\sigma(I)$] reflections	9791, 2063, 1634
R_{int}	0.028
R [$F^2 > 2\sigma(F^2)$]	0.073
$wR(F^2)$	0.227
S	1.09
No. of parameters/restraints	147/0
$\Delta\rho_{\text{max}}, \Delta\rho_{\text{min}}$ (e \AA^{-3})	0.44, -0.29

3.2. Crystal Structure

The DMF solution of compound $\text{C}_{10}\text{H}_{11}\text{N}_3\text{O}_3$ which was left for slow evaporation for two weeks gave colorless crystals suitable for X-ray analysis. The compound crystallizes in the triclinic group $P-1$. The molecular structure with the atomic-labelling scheme is shown in figure 2. The crystal structure solution and refinement are given in Table 1. Selected bond distances and angles are listed in Table 2. The asymmetric unit contains one organic molecule. The $-\text{NH}_2$ of the amid group is in *anti*-position with respect to the hydrazide group across C9–C10, while the carbonyl groups are in *trans*-position with respect to each other, across C9–C10. The compound assumes an *E* configuration across the C7=N1 bond. The ((2-hydroxyphenyl)–2-ethylidene) imino moiety adopts a *cis* conformation with the oxygen atom O2 of the carbonyl through the C9–N2 bond. The bond lengths values of 1.221 (3) \AA [C9–O2] and 1.234 (3) \AA [C1–O3] are indicative of a double character as

observed for similar compounds which crystallize in their keto forms [37, 7]. Those distances are comparable to the values found for a derivative which has an oxalate group [38]. The bond lengths values of 1.321 (4) Å [C10–N3] and 1.340 (4) Å [C9–N2] bonds are in the normal range observed for single C–N bonds [39]. The oxamic hydrazide fragment N1/N2/C9/O2/C10/O3/N3 is planar with a maximum deviation from the least-squares plane of –0.132 (2) Å for the N3 atom. The oxamic hydrazide moiety of the molecule is slightly twisted as reflected by the torsion angles values of 177.2 (2)° [N1–N2–C9–C10], –171.3 (3)° [N2–C9–C10–N3], –4.6 (4)° [O2–C9–N2–N1] and 8.4 (4)° [O3–C10–C9–N2].

The crystal packing of compound (I) is stabilized by intramolecular O(phenol)–H···N(hydrazide) and intermolecular N(amide)–H···O(amide), N(amide)–H···O(hydrazide) and C–H···O(phenol) hydrogen bonds. The intramolecular hydrogen bond O1(phenol)–H1···N1(hydrazide) which close in *S* (6) ring stabilized the conformation. Intermolecular hydrogen bonds, C3–H3···O1ⁱ(phenol) (i: –x+1, –y, –z+1) N3(amide)–

H3A···O3ⁱⁱ(amide) (ii: –x+1, –y+2, –z) and N3(amide)–H3B···O2ⁱⁱⁱ(hydrazide) (iii: –x+1, –y+1, –z) lead to the formation of sheets parallel to *ac* plane (Figure 3, Table 3).

Table 2. Selected geometric parameters (Å, °).

C2–O1	1.362 (4)	C9–O2	1.221 (3)
C7–N1	1.286 (4)	C10–O3	1.234 (3)
N1–N2	1.371 (3)	C10–N3	1.321 (4)
C9–N2	1.340 (4)		
N1–C7–C1	114.7 (2)	O2–C9–C10	122.9 (2)
N1–C7–C8	123.5 (3)	N2–C9–C10	111.5 (2)
C1–C7–C8	121.8 (3)	O3–C10–N3	126.3 (3)
O2–C9–N2	125.6 (2)	O3–C10–C9	119.6 (2)

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

D–H···A	D–H	H···A	D···A	D–H···A
C3–H3···O1 ⁱ	0.95	2.51	3.395 (4)	154.9
O1–H1···N1	0.84	1.79	2.521 (3)	144.3
N3–H3A···O3 ⁱⁱ	0.88	2.11	2.946 (3)	158.6
N3–H3B···O2 ⁱⁱⁱ	0.88	2.23	3.022 (3)	150.1

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

Table 4. Antioxidant activity of (I) and (II) at different concentration of DPPH.

Concentration (µM)	[DPPH] = 0.1014 mM			[DPPH] = 0.0507 mM		
	Trolox	(I)	(II)	Trolox	(I)	(II)
50	3.92±0.04	1.84±0.08	1.58±0.4	3.35±0.05	4.84±0.11	4.58±0.07
100	8.42±0.06	1.32±0.11	2.63±0.02	11.60±0.012	4.32±0.23	5.63±0.09
200	18.34±0.13	3.42±0.13	3.95±0.05	23.45±0.11	6.42±0.15	6.95±0.06
300	27.22±0.22	5.53±0.08	6.05±0.05	51.03±0.13	8.53±0.18	9.05±0.11
400	42.10±0.18	6.32±0.09	5.26±0.09	81.70±0.09	9.32±0.12	8.26±0.15
500	51.67±0.13	6.32±0.05	4.74±0.06	93.30±0.08	9.32±0.09	7.74±0.07

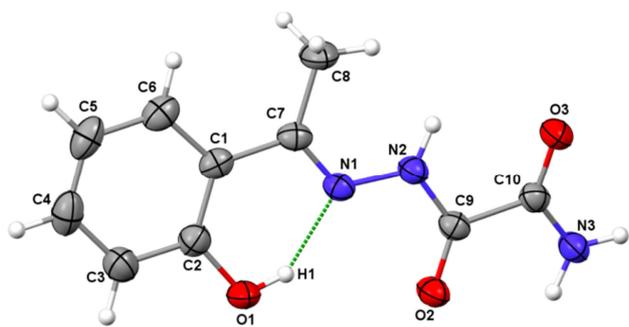


Figure 2. The crystal structure of the compound (I). Displacement ellipsoids are drawn at the 30% probability level and H atoms are shown as small sphere.

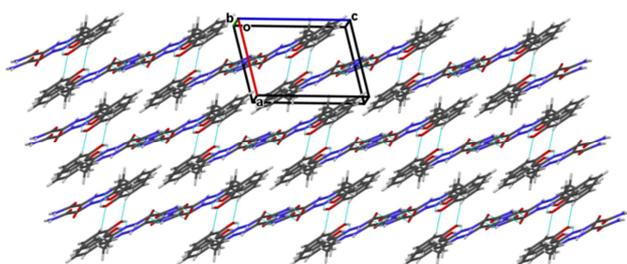


Figure 3. Chain formed by hydrogen bonds in the title compound (I) along *ac* plane.

3.3. Antioxidant Activity

DPPH[•] is a stable free radical which becomes a stable molecule when it accepts an electron or hydrogen radical. DPPH[•] radical scavenging is a method widely used to evaluate the antioxidant activity of compounds [40, 41]. The capacity of scavenging DPPH[•] radical of the two compounds (I) and (II) have been screened (Table 4). The Figure 4 shows the plots of DPPH[•] free radical scavenging activity (%) for the compounds (I) and (II). For compound (I), the scavenging activity increases with increasing the concentration in the range tested (50–500 mmol/L) for the two DPPH[•] initial concentrations. The scavenging activity of (I) varies, for the highest DPPH[•] (0.1014 mM) concentration, in the range 1.84±0.08 – 6.32±0.05% and between 4.84±0.11 and 9.32±0.09% for the lowest DPPH[•] concentration (0.0507 mM). This activity is due to the NH or OH groups which can react with DPPH[•] radical by the typical H–abstraction reaction to form a stable radical. Radical scavenging activity of compound (II) [(1.58±0.15 – 4.74±0.11% for the highest DPPH[•] concentration), (1.58±0.15 – 4.74±0.11% for the lowest DPPH[•] concentration)] is slightly lower than that observed for compound (I) in the concentration range screened (Figure 4). Comparatively to the scavenging activity of Trolox (3.92±0.04 – 51.67±0.13% for the highest DPPH[•] concentration), (3.35±0.05 – 93.30±0.08% for the lowest

DPPH concentration)), the values observed for compounds (I) and (II) are lower than those of Trolox in concentration

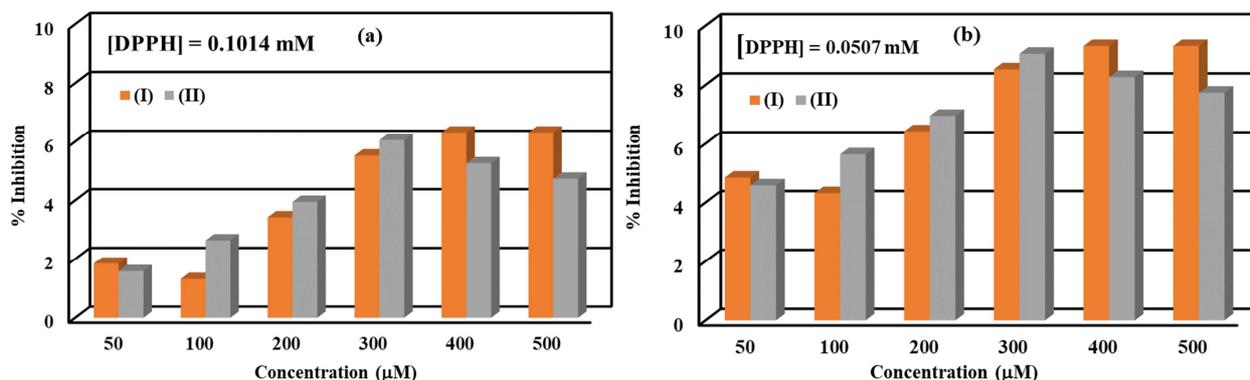


Figure 4. Antioxidant activity of (I) and (II): (a) [DPPH] = 0.1014 mM and (b) [DPPH] = 0.0507 mM.

4. Conclusion

The compounds (*E*)-2-amino-*N'*-(1-(2-hydroxyphenyl)ethylidene)-2-oxoacetohydrazide (I) and (*E*)-*N'*-(2-hydroxy-3-methoxybenzylidene)-2-amino-2-oxoacetohydrazide (II) were successfully synthesized by condensation reaction of oxamic hydrazide and 2-hydroxyacetophenone or *o*-vanillin respectively. The structures of the compounds were confirmed by elemental analysis and spectroscopic techniques (FT-IR, ¹H and ¹³C NMR). The molecular structure of the (I) was also determined using X-ray crystallography technique. Compounds (I) and (II) showed low antioxidant activity of about 10% in the screened concentration range [50–500 ppm].

Supplementary Materials

CCDC-2049222 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, or by contacting The Cambridge Crystallographic Data Centre, 12 Union Road, Cambridge CB2 1EZ, UK; fax: +44 (0) 1223-336033.

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