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

Fatou Faye¹, Amadou Guèye¹, Papa Samba Camara², Aïssatou Alioune Gaye¹, Farba Bouyaguï Tamboura², Nathalie Gruber³, Mohamed Gaye¹,*

¹Department of Chemistry, University Cheikh Anta Diop, Dakar, Senegal
²Department of Chemistry, University Alioune Diop, Bambye, Senegal
³Institut Le Bel, University of Strasbourg, Strasbourg, France

Email address: mohamedl.gaye@ucad.edu.sn (M. Gaye)

*Corresponding author

To cite this article:
Fatou Faye, Amadou Guèye, Papa Samba Camara, Aïssatou Alioune Gaye, Farba Bouyaguï Tamboura, Nathalie Gruber, Mohamed Gaye. Synthesis of Schiff Bases Compounds from Oxamic Hydrazide: Spectroscopic Characterization, X–ray Diffraction Structure and Antioxidant Activity Study. American Journal of Applied Chemistry. Vol. 9, No. 1, 2021, pp. 6-12. doi: 10.11648/j.ajac.20210901.12

Received: January 2, 2021; Accepted: January 11, 2021; Published: January 22, 2021

Abstract: The compounds (E)–2–amino–N¹–(1–(2–hydroxyphenyl)ethylidene)–2–oxoacetohydrazide (I) and (E)–N¹–(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, α = 81.730 (3)°, β = 72.738 (3)°, γ = 67.450 (3)°, V = 510.99 (8) Å³, Z = 2, T = 173 (2) K, m = 0.11 mm⁻¹, Dcalc = 1.438 g/cm³, Rmag = 0.028. 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···O1(phenol) (i: –x+1, –y, –z+1), N3(amide)–H3A···O3(ii: –x+1, –y+2, –z) and N3(amide)–H3B···O2(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
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 DPPH• radical scavenger was 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:

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

where \(A_{\text{control}}\) is the absorbance of the control reaction and \(A_{\text{sample}}\) 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–hydroxy–3–methoxybenzylidene)–2–amino–2–oxoacetohydrazide (I)

Oxamic hydrazide (2.037 g, 19.7 mmol) was suspended in 20 mL of cyclohexanol. o-vanillin (2.997 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 crystals after one week. Yield: 97%. Anal. Calc. for \([\text{C}_{34}\text{H}_{31}\text{N}_5\text{O}_5]\) (%): C, 50.63; H, 4.67; N, 17.71. Found: C, 50.60; H, 4.69; N, 17.75. FT–IR (n, cm\(^{-1}\)):

\[
3375(\text{N–H}), 3221(\text{NH}), 3205(\text{NH}), 1704(\text{C=O}), 1656(\text{C=O}), 1603(\text{C=O}), 1560(\text{C=O}), 1464(\text{C=O}), 1409(\text{C=O}), 1252(\text{C–Ophenol}), 1240(\text{C–Ophenol}), 1080(\text{N–N}), 837(\text{C–H}_3), 669(\text{C–H}_3). \text{RMN } ^1\text{H} (\text{dmso–d}_6, d (ppm)) : 3.80 (S, 3H, –OCH_3), \{6.82–7.10\} (m, 3H, H_A), 7.97 (S, 1H, CONH), 8.30 (S, 1H, Ar–OH), 8.75 (S, 1H, Hc=O), 10.75 (S, 1H, HNCOH), 12.36 (S, 1H, HNCOH). \text{RMN } ^1\text{C} (\text{dmso–d}_6, d (ppm)) : 161.51 (C=O), 156.54 (HN–COH), 150.46 (C=O), 147.87 (C=O), 147.20 (C=O), 120.65, 119.03, 118.72, 113.96 (C_A), 55.76 (–OCH_3).

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 crystals after one week. Yield: 97%. Anal. Calc. for \([\text{C}_{34}\text{H}_{31}\text{N}_5\text{O}_5]\) (%): C, 50.63; H, 4.67; N, 17.71. Found: C, 50.60; H, 4.69; N, 17.75. FT–IR (n, cm\(^{-1}\)):

\[
3375(\text{N–H}), 3221(\text{NH}), 3205(\text{NH}), 1704(\text{C=O}), 1656(\text{C=O}), 1603(\text{C=O}), 1560(\text{C=O}), 1464(\text{C=O}), 1409(\text{C=O}), 1252(\text{C–Ophenol}), 1240(\text{C–Ophenol}), 1080(\text{N–N}), 837(\text{C–H}_3), 669(\text{C–H}_3). \text{RMN } ^1\text{H} (\text{dmso–d}_6, d (ppm)) : 3.80 (S, 3H, –OCH_3), \{6.82–7.10\} (m, 3H, H_A), 7.97 (S, 1H, CONH), 8.30 (S, 1H, Ar–OH), 8.75 (S, 1H, Hc=O), 10.75 (S, 1H, HNCOH), 12.36 (S, 1H, HNCOH). \text{RMN } ^1\text{C} (\text{dmso–d}_6, d (ppm)) : 161.51 (C=O), 156.54 (HN–COH), 150.46 (C=O), 147.87 (C=O), 147.20 (C=O), 120.65, 119.03, 118.72, 113.96 (C_A), 55.76 (–OCH_3).

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 Mo Kα radiation (λ = 0.71073 Å). 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 F² 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 Uiso(H) = 1.2Ueq(C)(1.5 for CH₃ 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 [1570–1405 cm⁻¹] are due to the aromatic groups.

The ¹H NMR spectra of the compounds, in DMSO–d₆ solution, are recorded. Compound (I) gives two signals characteristic of iminolisation. Indeed, the single signal designating the moiety [–C(=O)–NH₂] does not appear on the spectrum. The two signals at 11.428 ppm and 12.925 ppm assigned respectively to HN=C–OH and HN=C–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 ¹³C NMR spectra in DMSO–d₆. Compound (I) gives a signal at 159.198 ppm corresponding to the imino carbon atom HN=C–OH. This signal is identified at 156.540 ppm in the spectrum of (II). The signals due to the hydrazide carbon atoms are at 162.322 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 | C₁₀H₁₁N₂O₂ |
|------------------|-------------|
| Mr               | 221.22      |
| Crystal shape/Color       | Prismatic/colorless |
| Crystal system, space group | Triclinic, P–1 |
| T (K)               | 173 (2)     |
| a (Å)              | 7.0399 (5)  |
| b (Å)              | 8.6252 (8)  |
| c (Å)              | 9.5474 (9)  |
| α (°)              | 81.730 (3)  |
| β (°)              | 72.738 (3)  |
| γ (°)              | 67.450 (3)  |
| V (Å³)             | 510.99 (8)  |
| Z                  | 2           |
| D_cal (g cm⁻³)     | 1.438       |
| F(000)             | 232         |
| Radiation type      | Mo Kα       |
| µ (mm⁻¹)           | 0.11        |
| Crystal size (mm)   | 0.12 x 0.10 x 0.10 |
| Trunc. T_max       | 0.982, 0.991 |
| h                  | –7 → 8     |
| k                  | –11 → 11   |
| l                  | –12 → 12   |
| Diffractometer     | Bruker APEX–II CCD |
| Absorption correction | Multi–scan SADABS |
| No. of measured, independent and observed [I > 2σ(F²)] reflections | 9791, 2063, 1634 |
| R_max              | 0.028       |
| R [I² > 2σ(F²)]    | 0.073       |
| wR(F²)             | 0.227       |
| S                  | 1.09        |
| No. of parameters/restraints | 147/0 |
| Δρ_min, Δρ_max (e Å⁻³) | 0.44, –0.29 |

3.2. Crystal Structure

The DMF solution of compound C₁₀H₁₁N₂O₂, 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 –NH₂ 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 anti–configuration across the C7=N1 bond. The (2–hydroxyphenyl)–2–ethyldiene) 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) Å [C9–O2] and 1.234 (3) Å [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)–H···N1(hydrazide) which close in S (6) ring stabilized the conformation. Intermolecular hydrogen bonds, C3–H3···O1(phenol) (i: −x+1, −y, −z+1) N3(amide)–H3A···O3(amide) (ii: −x+1, −y+2, −z) and N3(amide)–H3B···O2(hydrazide) (iii: −x+1, −y+1, −z) lead to the formation of sheets parallel to ac plane (Figure 3, Table 3).

### Table 3. Hydrogen–bond geometry (Å, °).

| D–H···A  | D–H     | H···A  | D–A     | D–H···A  |
|----------|---------|--------|---------|---------|
| C3–H3···O1′ | 0.95   | 2.51   | 3.395 (4) | 154.9   |
| O1–H1···N1  | 0.84   | 1.79   | 2.521 (3) | 144.3   |
| N3–H3A···O3ii | 0.88  | 2.11   | 2.946 (3) | 158.6   |
| N3–H3B···O2ii | 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.

### 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 concentration. 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

### 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)         | Trolox (II)       | Trolox (I)        | Trolox (II)       |
| 50                 | 1.84±0.08         | 1.58±0.04         | 4.84±0.11         | 4.58±0.07       |
| 100                | 3.22±0.06         | 2.63±0.02         | 4.32±0.23         | 5.63±0.09       |
| 200                | 3.42±0.13         | 3.95±0.05         | 6.42±0.15         | 6.95±0.06       |
| 300                | 5.53±0.08         | 6.05±0.05         | 8.53±0.18         | 9.05±0.11       |
| 400                | 6.32±0.09         | 5.26±0.09         | 9.32±0.12         | 8.26±0.15       |
| 500                | 6.32±0.05         | 4.74±0.06         | 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.

DPPH concentration)), the values observed for compounds (I) and (II) are lower than those of Trolox in concentration range investigated (50–500 mM). The scavenging activity of compounds (I) and (II) are low and do not exceed 10%.

Figure 4. Antioxidant activity of (I) and (II): (a) $[	ext{DPPH}] = 0.1014$ mM and (b) $[	ext{DPPH}] = 0.0507$ 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, $^1$H and $^{13}$C 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.

References
[1] Ćobeljić, B., Pevec, A., Turel, I., Spasojević, V., Milčić, M., Mitić, D., Sladić, D. and Anđelković, K. (2014). Analysis of the structures of the Cu (I) and Cu (II) complexes with 3–acetylpypyridine and thiocyanate. Polyhedron, 69: 77–83.
[2] Gutman, C. T. and Brunold, T. C. (2012). Spectroscopic and computational studies of a small–molecule functional mimic of iron superoxide dismutase, iron 2,6–diacetylpyridine bis (thioxamidicabazole) complexes: structure, anticancer function and mechanism. Dalton Trans, 49 (47): 17207–17220.
[3] Sumar, M., Ivanovic–Burmazovic, I., Hodzic, I. and Andjelkovic, K. (2002). Pentagonal–bipyramidal Mn (II) and Zn (II) complexes with 2′,2′″–(2,6–pyridindiyldi–ethylidene)dioxamohydrazide. Synthesis and Reactivity in Inorganic and Metal–Organic Chemistry, 32 (4): 721–737.
[4] Jarestan, M., Khalatbari, K., pouraei, A., Sadat Shandiz, S. A., Beigi, S., Hedayati, M., Majlesi, A., Akbari, F. and Salehzadeh, A. (2020). Preparation, characterization, and anticancer efficacy of novel cobalt oxide nanoparticles conjugated with thiosemicarbazide. 3 Biotech, 10 (5): 230.
[5] Haba, P., Sow, M. M., Sarr, M., Thiam, J. E., Diaw, M. and Gaye, M. L. (2020). Syntheses, characterization and X–ray crystal structure of polymeric heteronuclear oxo–bridged Fe/Na assembled with salen–type Schiff base and dicyanamide. Sciences Journal of Chemistry, 8 (2): 20–27.
[6] Li, S., Khan, M. H., Wang, X., Cai, M., Zhang, J., Jiang, M., Zhang, Z., Wen, X., Liang, H. and Yang, F. (2020). Synthesis of a series of novel In (III) 2,6–diacetylpyridine bis (thioxemiacbazide) complexes: structure, anticancer function and mechanism. Dalton Trans, 49 (47): 17207–17220.
[7] Kane, C. H., Thiam, E. I., Tamboura, F. B., Gaye, M. and Retailleau, P. (2012). Bis{(2–Amino–2–oxo–$N'$–(1(E))–1–(pyridin–2–yl–kN)ethylidene)acetohydrazidato–kN}$^1$, $O^1$nickel (II). Acta Crystallographica E, 68 (5), m553.
[8] Dimmock, J. R., Vashishtha, S. C. and Stables, J. P. (2000). Anticonvulsant properties of various acetylhydrazones, oxamoylhydrazones and semicarbazones derived from aromatic and unsaturated carboxyl compounds. European Journal of Medicinal Chemistry, 35 (2): 241–248.
[9] Jarestan, M., Khalatbari, K., pouraei, A., Sadat Shandiz, S. A., Beigi, S., Hedayati, M., Majlesi, A., Akbari, F. and Salehzadeh, A. (2020). Preparation, characterization, and anticancer efficacy of novel cobalt oxide nanoparticles conjugated with thiosemicarbazide. 3 Biotech, 10 (5): 230.
behavior of an unusual seven-coordinate iron (III) complex in aqueous solution. Inorganic Chemistry, 41 (20): 5150–5161.

El–Toukhy, A. (1991). Stoichiometry, Products and kinetics of complexes. Inorganic Chemistry, 45 (11): 4518–4525.

Aljahdali, M., El–Sherif, A. A., Shoukry, M. M., Hosny, W. M. and Abd–Elmohgy, M. G. (2013). Complex formation equilibria of unusual seven coordinate Fe (III) complexes with DNA constituents. Journal of Solution Chemistry, 42 (8): 1663–1679.

Sheldrick, G. M. (2015). Crystal structure refinement with SHELXL. Acta Crystallographica C, 71 (1): 3–8.

Sheldrick, G. M. (2015). It WINGX and It ORTEP for Windows: An Update. Journal of Applied Crystallography, 45 (4): 849–854.

Singh, A. K., Pandey, O. P. and Sengupta, S. K. (2013). Synthesis, spectral and antimicrobial activity of Zn (II) complexes with Schiff bases derived from 2–hydrazino–5–[substituted phenyl]–3,4–thiadiazole and benzoaldehyde /2–hydroxyacetophenone / indoline–2,3–dione. Spectrochimica Acta Part A, 113: 393–399.

Saghafiroush, L. A., Hosseinpour, S., Bezpaklo, M. W. and Kassel, W. S. (2019). X–Ray crystal structural and spectral studies of copper (II) and nickel (II) complexes of functionalized bis (thiosemicarbazone) ligands and investigation of their electrochemical behavior. Inorganica Chimica Acta, 484: 527–534.

Tripathi, G. N. R. and Katon, J. E. (1979). Vibrational spectra and structure of crystalline oxalyl hydradize and semioxamazide. Journal of Molecular Structure, 54: 19–29.

Mashevskaya, M. S., Kolla, V. É., Nazmetdinov, F. Ya., Plaksina, A. N. and Semenova, Z. N. (1991). Synthesis of oxalic acid arylidenedihydrazides and their biological activity. Pharmaceutical Chemistry Journal, 25 (2), 83–85.

Chandra, S. and Sharma, A. K. (2009). Nickel (II) and Copper (II) complexes with Schiff base ligand 2,6–diacetylpyridine bis (carboxyhydrazone): synthesis and IR, Mass, 1H NMR, electronic and EPR spectral studies. Spectrochimica Acta Part A, 72 (4): 851–857.

Karimian, R., Hosseini–Monfared, H., Bikas, R., Arslan, N. B., Kazak, C. and Koroglu, A. (2012). (E,E)–N1–4–[(2–benzoylehydrazin–1–ylidene) methyl][benzylidenbenzo hydrazone. Acta Crystallographica E, 68 (5): o1433.

Tai, X.–S., Yin, J. and Kong, F.–Y. (2007). Crystal structure of 2–carboxylbenzaldehyde furan–2–carboxyhydrazone methanol hemisolvate, C13H16N2O2.5CH3OH. Zeitschrift für Kristallographie–New Crystal Structures, 222 (4): 401–402.

Monfared, H. H., Bikas, R. and Mayer, P. (2010). (E)–3–Hydroxy–N–(2–hydroxybenzylidenec)–2–naphthohydrazone. Acta Crystallographica E, 66 (1), o236–o237.
[40] Taha, Z. A., Ajlouni, A. M., Momani, W. A. and Al–Ghawi, A. A. (2011). Syntheses, characterization, biological activities and photophysical properties of lanthanides complexes with a tetradeutate Schiff base ligand. Spectrochimica Acta Part A, 81 (1): 570–577.

[41] Foti, M. C., Daquino, C. and Geraci, C. (2004). Electron–transfer reaction of cinnamic acids and their methyl esters with the DPPH• radical in alcoholic solutions. Journal of Organic Chemistry, 69 (7): 2309–2314.