# Syntheses, Spectral, Thermal and Structural Characterization of 2-Phenyl-2-(1hydroxyiminoethyl)-1,2,3,4-tetrahydroquinazoline and Its Novel Nickel(II) Complex

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**Abstract.** The 2-phenyl-2-(1-hydroxyiminoethyl)-1,2,3,4-tetrahydroquinazoline (HL) and its Ni<sup>II</sup> complex have been prepared and characterized by spectral method (FT-IR, NMR (<sup>13</sup>C and <sup>1</sup>H), UVvis.), elemental analysis, magnetic susceptibility and thermal analysis (TG, DTA) techniques. The crystal structures of HL and Ni<sup>II</sup> complex were also determined by the single crystal X-ray diffraction. The HL and Ni<sup>II</sup> complex crystallizes in the monoclinic and triclinic, space groups  $P2_1/c$  and  $P\overline{1}$ , respectively. The complex was occurred by the elimination of 1 mole of 2-aminobenzylamine from the 2 moles of the HL after the ring opening reaction by the Ni<sup>II</sup> attack. Crystallographic study reveal that Ni<sup>II</sup> atom has a square planer geometry being coordinated by four nitrogen atoms of HL. Two thermal processes of the HL and Ni<sup>II</sup> complex can occur in TG and DTA curves.

Keywords: 1,2,3,4-Tetrahydroquinazoline; Quinazoline; Dioxime; Imine-oxime

benzene or xylene with azeotropic water removal, in re-

## **1** Introduction

2-phenyl-2-(1-hydroxyiminoethyl)-1,2,3,4-tetrahydroquinazoline as a ligand which contains an oxime parts was synthesized from 2-aminobenzylamine and 1-phenyl-1,2-propanedione-2-oxime (Scheme 1). Oximes as ligands have played a significant role in the development of transition metal chemistry. This development has been documented in a number of review articles and we refer the readers to some of these excellent treatises [1-8]. An early treatise by Chakravorty [1] is a comprehensive review on the structural chemistry of simple oximes, vic-dioximes, quinonemonoximes, and carbonyl-, imine-, pyridine-, azo-, hydroxy- and amidoximes. Oxime derivatives are very important compounds because of their biological activity, such as insecticidal, miticidal, nematocidal activities, antidote activities for organophosphor poison. Some oxime complexes have anti cancerogenic activities [9-11], 1.2.3.4-Tetrahydroquinazolines which are heterocyclic compounds, are of interest as dihydrofolate reductase inhibitors, antitubercular and antibacterial agents [13]. 1,2,3,4-Tetrahydroquinazolines can be prepared by the condensation of 1,3diamines and substituted aldehydes or ketones in refluxing

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fluxing ethanol/acetic acid mixtures and by reaction in alkali media or by the condensation of 2-aminobenzylamine with aromatic aldehydes in ionic liquids followed by extraction into diethyl ether [13-15]. The 1,2,3,4-tetrahydroquinazolines may also be prepared by the simple method of slurring the reagents together in water at ambient temperature in a similar manner in the synthesis of aromatic Schiff bases by the *Tanaka*'s method [13-16]. In this work, a 2-phenyl-2-(1-hydroxyiminoethyl)-1,2,3,4-

tetrahydroquinazoline and its Ni<sup>II</sup> complex were synthesized and characterized by spectral (FT-IR, NMR (<sup>13</sup>C and <sup>1</sup>H), UV-vis.) and thermal method. Structures determinations were performed by the X-ray single crystal analyses.

#### 2 Results and Discussion

#### 2.1 Characterization of HL

The NMR measurements were made in DMSO-d<sub>6</sub> and TMS was used as internal standard. The proton spectrum was measured from freshly synthesized compound. The spectrum was repeated in one day period during one week, but no change showing the tautomeric forms, was detected. Chemical shift assignments were based, in addition to form normal proton and carbon measurements, on DEPT experiments. In the <sup>1</sup>H NMR spectrum of the ligand, a singlet peak for the OH proton of oxime group is observed at 8.57 ppm. The N-H proton adjacent to the CH<sub>2</sub> groups in the heterocyclic ring resonates at 2.19 ppm and the other one resonate at 4.94 ppm. The CH<sub>2</sub> group of the hetero-

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cyclic ring of the ligand resonates at 3.82 ppm. The CH<sub>3</sub> protons adjacent to the oxime group resonate at 1.81 ppm. The aromatic C-H protons resonate at 6.62-7.56 ppm.

In the IR spectra of the ligand, bands at 3386, 3320, 3305, 3040–3005, 2900–2840, 1632, 1350–1315, and 960 cm<sup>-1</sup> belong to N1–H, N3–H, O–H, C–H<sub>arom</sub>, C–H<sub>aliph</sub>, C=N, –C–N– and N–O vibrations, respectively. The NMR and IR spectrum of HL is synthesized which contains the oxime part, was in good agreement of with the known oximes [17–20] and 1,2,3,4-tetra-hydroquinazolines [14, 15] and suitable for the synthesized 1,2,3,4-tetrahydroquinazoline ligand.

The crystallographic data of the compound are summarized in Table 1 and selected bond lengths and angles and hydrogen bond geometries are listed in Table 2. The molecular structure and inter molecular hydrogen bonds of HL are shown in Figure 1 and 2, respectively.

The intermolecular hydrogen bonds found at x, y, z-1 for N1-H1...O20 and C9-H9...O20 and at 1-x, -y, 1-z for the O20-H20...N19 and O20-H20...N1. The intermolecular hydrogen bonds are at x, y, z-1, bonded molecules to each other to form molecular sheets through the axis. The intermolecular hydrogen bonds at 1-x, -y, 1-z, are found between the molecular sheets that were formed by the N1-H1...O20 and C9-H9...O20 intermolecular hydrogen bonds. So, the hydrogen bonds formed the two dimensional polymeric structure of the synthesized HL in solid state.

## 2.2 Characterization of the Ni<sup>II</sup> complex

To prepare the complexes of the tautomeric forms of the ligand HL that was only one form in both solid and solu-



Fig. 1 ORTEP-III diagram with 30 % probability ellipsoids of HL



Fig. 2 The intermolecular interactions of the HL ligand with unit cell.

tion, by the template effect of the Ni<sup>II</sup>, the ligand HL was treated with the different equivalent amount of Ni<sup>II</sup>. But, it was found that at each time, the products were red and diamagnetic and had the same IR and UV-Vis. spectra, indicated that was a complex of only one tautomer of HL. In the IR spectrum of the complex, there are no peaks for N-H and O-H belongs to the ligand since complex form upon deprotonation of N-H and O-H hydrogen atoms. The C=N vibration of the ligand shifted to  $1610 \text{ cm}^{-1}$  indicated that the both oxime and imine nitrogen atoms bonded to the Ni<sup>2+</sup> ion. The N-O vibration of the ligand shifted to higher frequency for the nickel(II) complex indicating that the oxime oxygen non-coordinated to the nickel atom. These results may be suitable with complex of the diimine tautomer of the synthesized ligand. But, as unexpected, the complex has a band at  $1380 \text{ cm}^{-1}$  for the -C-N- vibration indicating that the nitrogen atom bonded to the nickel atom. These results indicating that the ligand in the complex is non of the known tautomer of the 1,2,3,4-tetrahydroquinazolines and the IR results do not give any addition knowledge to estimate structure of the complex.

The X-ray single crystal structural studies showed the ligand in the complex has whole different structure from the known tautomer of the 1,2,3,4-tetrahydroquinazolines. The X-ray single crystal structure analysis of the complex showed may be formed by the elimination 1 mole of 2-aminobenzylamine from 2 moles of 1,2,3,4-tetrahydroquinazoline and the ethanol which is presence of the reaction media as solvent, acted as reactant (Scheme 2). The crystallographic data of the complex were summarized in Table 1 and selected bond lengths, angles and hydrogen bond geometries are given Table 3. Tough the ligand in the complex is like the diimine tautomers of 1,2,3,4-tetrahydroquinazolines [13-15], it had completely different bond properties. Fist of all, the ligand in the complex has an ethoxy part bonded to carbon atom of the ketone in the benzylamine type tautomer parts of the diimine tautomer [13, 15]. In this part, N1-C2 and N1-C10 bond lengths are 1.475(4) and 1.371(4) Å, respectively, indicate these bonds are single as unlike diimine tautomers of 1,2,3,4tetrahydroquinazolines. In addition to the difference, the double bond which was between the N atom and the carbon atom of the ketone in the aniline type tautomer parts of the diimine tautomers of 1,2,3,4-tetrahydroquinazolines [13-15], was found between carbon and N atoms of the benzylamine part of the ligand in the complex. The C2-N1 and N3-C4 and N3-C2' bond lengths indicated the double bond is in N3-C4, are 1.475(4) and 1.298(4) and 1.477(4) Å, respectively. Like glyoximes, in the complex the O-H…O hydrogen bond bridge occurres to stabilize the square-planar geometry in around of Ni<sup>II</sup> atom (Fig. 3) [18-21]. Also, at 2-x, 1-y, 1-z to bond complex molecules to each other, the C14 and C15 atoms of the complex act as hydrogen bond donors via their H14 and H15 atoms to O20' and N19 atoms, respectively. In addition to this intra and inter actions, weak C-H··· $\pi$  inter action was found between the H14 atom of the C14 carbon and centre of gravity (Cg) of the C5-C10 ring (phenyl ring of 2-aminobenzylamine) at 1-x, 1-y, 1-z. The hydrogen bonds and C-H··· $\pi$  inter action of the complex were shown in Figure 4.

#### 2.3 Thermal Analyses

The HL ligand exhibits three decomposition stages. The ligand melts at 106.3 °C and then in the temperature range of 115-342 °C, HL ligand involves the consecutive decompositions by giving exothermic effects (DTA = 158 and 224 °C). The third stage is related to the burning of the remain organic residue, in the temperature range of 343-557 °C (DTA = 521 °C).



Fig. 3 An ORTEP-III drawing of complex with displacement ellipsoids shown at the 30 % probability.



Fig. 4 The intra- and intermolecular interactions of the Nickel(II) complex with unit cell.

Table 1 Crystallographic data of the HL and Ni<sup>II</sup> complex

	HL	Ni <sup>II</sup> Complex
Formula	C <sub>16</sub> H <sub>17</sub> N <sub>3</sub> O	C <sub>27</sub> H <sub>28</sub> N <sub>4</sub> O <sub>3</sub> Ni
Formula Weight	267.33	515.24
Crystal System	monoclinic	triclinic
Space group	$P2_1/c$	ΡĪ
a /Å	10.745(5)	10.502(2)
b /Å	20.324(5)	11.050(3)
c /Å	6.687(5)	11.603(2)
α /°	90	79.25(2)
β /°	107.545(5)	72.81(1)
γ /°	90	80.58(2)
$V(Å^3)$	1392.4(13)	1255.3(5)
Z	4	2
Densitiv pcalc $/(g/cm^3)$	1.275	1.363
$\mu(Mo-K_{\alpha})/mm^{-1}$	0.082	0.808
F(000)	568	540
Crystal Size /mm	$0.10 \times 0.49 \times 0.58$	0.18×0.34×0.41
Data Collection		
Temperature /K	293	
Radiation /Å	$M_0 - K_{\alpha} = 0.71073$	
Theta range /°	$2.0 < \theta < 27.0$	$2.4 < \theta < 26.0$
Indis range	$h = -13 \rightarrow 13$ ,	$h = -12 \rightarrow 12$ ,
0	$k = -25 \rightarrow 25$	$k = -13 \rightarrow 13$
	$l = -8 \rightarrow 7$	$1 = -14 \rightarrow 14$
Tot., Uniq. Data, Rint	14993, 3052, 0.077	18099, 4933, 0.080
Observed data $[I > 2\sigma(I)]$	2016	3810
Refinement		
N <sub>ref</sub> , N <sub>par</sub>	3052, 195	4933, 322
$R_1, wR_2 w = 1/[\sigma_2(F_a^2) + (0.0814P)^2]$	0.0481, 0.1405	0.0493, 0.1199
where $\tilde{P} = (F_{0}^{2} + 2F_{c}^{2})/3$	·	
Goodness of fit	1.01	1.05

Table 2Structural parameters /Å, ° for HL

Bond lengths /Å		Bond angles /°	
N1-C2 N1-C10 N3-C4 N3-C2 N1-H1 N3-H3 C2-C17 C17-N19 N19-O20 O20 H20	$\begin{array}{c} 1.468(2) \\ 1.383(2) \\ 1.461(3) \\ 1.465(2) \\ 0.78(2) \\ 0.93(2) \\ 1.538(2) \\ 1.268(2) \\ 1.417(2) \\ 0.96(2) \end{array}$	$\begin{array}{c} C10-N1-C2\\ N1-C2-N3\\ C2-N3-C4\\ N3-C4-C5\\ N1-C2-C11\\ N3-C2-C11\\ C11-C2-C17\\ C2-C17-N19\\ C17-N19-O20\\ N10\\ O20\\ H20\\ D10\\ O20\\ H20\\ H20\\ H20\\ H20\\ H20\\ H20\\ H20\\ H$	121.3(2) 111.6(1) 111.2(1) 113.3(2) 108.0(1) 112.0(1) 107.7(1) 117.5(1) 112.2(1)
Hydrogen-bonding /	Å		102(2)
D-H···A	D…A /Å		
N1-H1O20 <sup>i</sup> C9-H9O20 <sup>i</sup> O20-H20N19 <sup>ii</sup> O20-H20N1 <sup>ii</sup>	3.314(3) 3.510(3) 2.822(2) 3.630(2)		

i: x, y, z-1, ii: 1-x, -y, 1-z

The Ni<sup>II</sup> complex has a high thermal stability. The first decomposition stage begins at 230 °C. In the temperature range of 231-245 °C, the first stage of the complex is related to release of the ethanol group of HL ligand (found = 9.64, calcd. = 8.75), by giving exothermic contribution in the DTA curve (DTA = 241 °C). The following two exothermic stages involve successive decomposition of HL. In the last stage, remain organic part is abruptly burnt in the

Table 3 Structural parameters /Å, ° for Ni<sup>II</sup> complex

Bond lengths /Å		Bond angles /°	
-C2	1.475(4)	N1-C10-C5	120.4(3)
-C10	1.371(4)	C10-C5-C4	123.7(3)
-C4	1.298(4)	C5-C4-N3	126.1(3)
-C2'	1.477(4)	C4-N3-Ni	126.0(2)
-H4	0.930(4)	N1-C2-C17	107.2(2)
7-N19	1.272(4)	N3-C2'-C17'	107.0(3)
7'-N19'	1.281(4)	C17-N19-Ni	117.9(2)
-01	1.431(4)	C17'-N19'-Ni	119.4(2)
9-O20	1.357(3)	N3-Ni-N1	95.9(1)
9'-O20'	1.359(3)	N1-Ni-N19	84.9(1)
-N1	1.858(3)	N3-Ni-N19'	83.0(1)
-N3	1.834(2)	N19-Ni-N19'	96.2(1)
-N19	1.859(3)	N1-Ni-N19'	178.8(1)
-N19′	1.881(3)	N3-Ni-N19'	178.0(1)
9-01 9'-020 9'-020' -N1 -N3 -N19 -N19'	1.451(4) 1.357(3) 1.359(3) 1.858(3) 1.834(2) 1.859(3) 1.881(3)	C17 - N19 - N1 N3-Ni-N1 N1-Ni-N19 N3-Ni-N19' N19-Ni-N19' N1-Ni-N19' N3-Ni-N19'	1 1 1 1

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Hydrogen-bonding /Å

D-H···A	D…A /Å	
020–H20…O20' C14–H14…O20' <sup>i</sup> C15–H15…N19 <sup>i</sup> C14'–H14'…Cg <sup>ii</sup>	2.412(4) 3.464(7) 3.732(6) 3.774(6)	

i: 2-x, 1-y, 1-z, ii: 1-x, 1-y, 1-z.

complex, leading finally to give NiO. The final decomposition products was identified by IR spectroscopy. The overall weight loss agrees well with the proposed structure.

#### **3** Experimental Section

#### 3.1 Materials and measurements

All chemicals used were analytical reagent. Elemental analyses for C, H, and N were carried out at the TÜBİTAK Marmara Research Centre. Magnetic susceptibility measurements at room temperatures were performed using a Sherwood Scientific MK1 model Gouy magnetic balance. UV-vis. spectrum was obtained in the methanol solutions  $(10^{-3} \text{ mol/L})$  of the complex with a Unicam UV2 spectrometer in the range of 900-190 nm. The <sup>1</sup>H-NMR and <sup>13</sup>C-NMR spectra were recorded on a Varian Mercury Plus 400 MHz FT-NMR spectrometer, utilizing deuterated dimethylsulphoxide as a solvent and TMS was used as an internal standard. FT-IR spectra were recorded in the 4000-400 cm<sup>-1</sup> region with a Mattson 1000 FT-IR spectrometer using KBr pellets. Thermal analysis curves (TG and DTA) were recorded simultaneously in a static air atmosphere with a Shimadzu DTA 50 thermal analyzer. The heating rate was 10 °C min<sup>-1</sup>. The crystal data were collected using  $\omega - 2\theta$  scan techniques on Stoe IPDS-2 diffractometer with a graphite-filtered Mo-K $\alpha$  radiation ( $\lambda = 0.71073$  Å). Software program(s) used, for data collection and cell refinement: X-AREA [22]; for data reduction: X-RED32 [22]; to solve structure: SHELXS86 [23]; to refinement structure: SHELXL97 [24]; for molecular graphics: ORTEPIII [25]; to prepare material for publication: WinGX [26].

## 3.2 Synthesis of HL

The compound can be alternatively named as 1-(2-phenyl-1,2,3,4-tetrahydro quinazolin-2-yl)ethanone oxime. A solution of 2-aminobenzylamine (10 mmol, 1.25 g) in EtOH (30 ml) was added drop wise to a solution of 1-phenyl-1,2-propanedione-2-oxime (10 mmol, 1.64 g) in ethanol at -5 °C. After 1 hour, glacial acetic acid (1.5 ml) was added drop wise to the mixture. The solutions were cooled to -15 °C in a temperature-controlled bath and stirred for 4 h. The solutions were kept 12 h and then the crystalline products were collected by filtration, dried on air (Scheme 1). The precipitated product was filtered, washed with water and then recrystallized from ethanol. Yield 84 % (beige needless crystals); mp 106.3 °C; *Analytical data:* C<sub>16</sub>H<sub>17</sub>N<sub>3</sub>O (267.33 gmol<sup>-1</sup>); C 71.85 (calc. 71.89); H 6.41 (6.38); N 15.75 (15.72) %.

<sup>1</sup>H NMR (DMSO-d<sub>6</sub>) δ = 1.81 (s, 3H, -CH<sub>3</sub>), 2.19 (s, 1H, -N<sub>3</sub>H), 3.82 (m, 2H, -CH<sub>2</sub>), 4.94 (s, 1H, -N<sub>1</sub>H), 8.57 (s, 1H, -OH), 6.62 (d, 1H, J = 8 Hz, H-9), 6.66 (t, 1H, J = 7.3 Hz, H-7), 6.85 (d, 1H, J = 7.3 Hz, H-6), 7.04 (t, 1H, J = 7.3 Hz, H-8), 7.30 (d, 2H, J = 10 Hz, H-12), 7.31 (t, 1H, J = 7.3 Hz, H-14), 7.56 (t, 2H, J = 8 Hz, H-13), <sup>13</sup>C NMR (D<sub>6</sub>]DMSO) δ = 10.38 (C-18), 42.91 (C-4), 74.35 (C-2), 115.47 (C-9), 118.15 (C-7), 121.39 (C-5), 122.57 (C-14), 126.17 (C-8), 126.80 (C-12), 127.87 (C-16), 128.21 (C-13), 128.55 (C-15), 128.89 (C-6), 141.96 (C-11), 142.03 (C-10), 158.78 (C-17); IR (KBr): v(N1-H) 3386 s, v(N3-H) 3320 s, v(OH) 3305 s, v(C-H<sub>arom</sub>) 3040-3005 w, v(C-H<sub>aliph</sub>) 2900-2840 w, v(C=N) 1632 s, v(C-N-) 1350-1315 m, v(N-O) 960 m cm<sup>-1</sup>.



Scheme 1 Synthesis of HL

## 3.3 Synthesis of the Ni<sup>II</sup> complex

A solution of HL (10 mmol, 2.67 g,) in ethanole (20 ml) was added drop wise with stirring at 60 °C to a solution of NiCl<sub>2</sub>·6H<sub>2</sub>O (5 mmol, 1.188 g) in ethanol (20 mL). The mixture was stirred for 4 h at 60 °C and then slowly cooled to room temperature (Scheme 2). The crystals formed were filtered and washed with 10 mL of cold ethanol and dried on air. The complex named as 1-ethoxy-1-{2- [(2-hydroxyimino-1-phenyl-propylimino)methyl-phenylamino}-1-phenyl-propan-2-oneoximatonickel(II). Yield 71 % (red crystals); d.p. 231 °C; *Analytical data:* C<sub>27</sub>H<sub>28</sub>N<sub>4</sub>O<sub>3</sub>Ni (515.24 gmol<sup>-1</sup>); C 62.91 (calc. 62.94); H 5.53 (5.48); N 10.91 (10.87) %.

IR(KBr): ν(C-H<sub>arom.</sub>) 3100–3050 w, ν(C-H<sub>aliph.</sub>) 2877 w, ν(C=N) 1610 s, ν(C-N) 1380, ν(N-O) 1008 m, ν(Ni-N) 474 cm<sup>-1</sup>; UV/VIS:  $λ_{max}$  (lg ε): 452 nm (1342);  $μ_{eff}$ : diamagnetic;



Scheme 2 Synthesis of the Nickel(II) complex

#### 3.4 Supplementary Data

Crystallographic data (excluding structure factors) have been deposited with the Cambridge Crystallographic Data Centre as the supplementary publication No. CCDC 616665, 616666. Copies of the data can be obtained, free of charge, on application to CCDC, 12 Union Road, Cambridge CB2 1EZ, UK (fax: +44-1223-336033 or e-mail: deposit@ccdc.cam.ac.uk).

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