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SYNTHESIS AND BIOLOGICAL ACTIVITIES OF SOME 3-(AMINO OR ARYLIDINE)-2-PYRIDINYL-4(3*H*)-QUINAZOLINONE AND SOME OF THEIR COPPER COMPLEXES

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Abstract

Several 4(3*H*)-quinazolinones containing pyridine moiety were synthesized starting from substituted anthranilic acid derivatives. Copper complexes of 3-amino-2-pyridinyl-4(3*H*)-quinazolinones and some of their derivatives were synthesized and characterized. The synthesized compounds were screened for their anticonvulsant, analgesic, antitumor, as well as, their antimicrobial activities.

Keywords: 4(3*H*)-Quinazolinones; Copper complexes; Anticonvulsant; Analgesic; Antitumor and antimicrobial activities

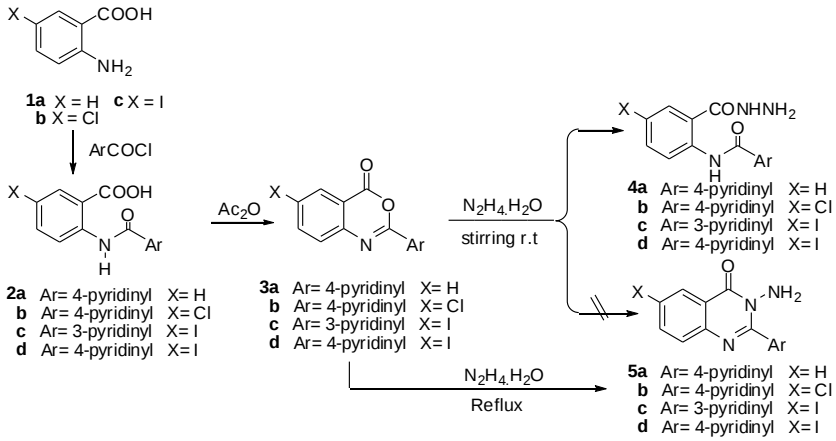
Introduction

Several compounds containing quinazoline moiety are well known in medicinal chemistry as important compounds for their therapeutic value. The chemistry of 4(3*H*)-quinazolinone system had received an increasing interest due to its biological significance. Also, many derivatives of this system showed antifungal[1], antibacterial[2], antitumor[3], anti-inflammatory[4], anticonvulsant[5-7], analgesic[8,9] and anti-tuberculosis[10,11] activities. Moreover, numerous heterocyclic compounds containing the pyridine nucleus exhibit various pharmacological activities. Furthermore, pyridine nucleus is well known to be found in a broad variety of drugs such as nicotinamide (pyridine-3-carboxamide) which is well-known drug used as respiratory analeptic[12], as well as, fungicides[13] pesticides[14] or for treatment of benign prostatic hyperplasia[15]. Therefore, it was aimed in the present work to prepare a new series of 4(3*H*)-quinazolinone derivatives incorporating biologically active pyridine moiety and the corresponding copper complexes for their expected biological value.

Results and Discussion

The most common approaches to synthesize 3,2-disubstituted-4(3*H*)-quinazolinone derivatives involves amidation of 2-aminobenzoic acid derivatives, then treatment of the amidated anthranilic acid derivatives with acetic anhydride to afford benzoxazinone, followed by condensation with nitrogen nucleophiles. Thus, the amide derivatives **2a-d** were prepared by heating the corresponding pyridine

carboxylic acid chloride with the anthranilic acid derivatives **1a-c** in toluene. Cyclodehydration of the amides **2a-d** was carried out upon heating in acetic anhydride to afford the 3,1-benzoxazin-4-ones **3a-d** (85-90% yield). Structures of the amides **2** and benzoxazines **3** were supported on the basis of correct analytical, as well as, spectral data. IR spectra of products **3** showed absence of bands for OH, NH groups and the presence of strong band at: 1773-1753 cm^{-1}, characteristic of the lactone group. ^{1}HNMR spectra of the benzoxazines **3c,d** were compatible with the assigned structure. Mass spectrum of the benzoxazine derivative **3c** showed a molecular ion peak at: *m/z* = 258 (M^+; 87.1%), with a base peak at 106 (100 %). When 2-pyridinyl-3,1-benzoxazinones **3a-d** were reacted with hydrazine hydrate in dioxane at room temperature the only isolatable produced **4a-d** were obtained rather than the expected 3-amino-2- pyridinyl-4(3*H*)-quinazolinone derivatives **5**. The latter products were obtained in good yield upon heating of **3a-d** with hydrazine hydrate in *n*-butanol (Scheme **1**).



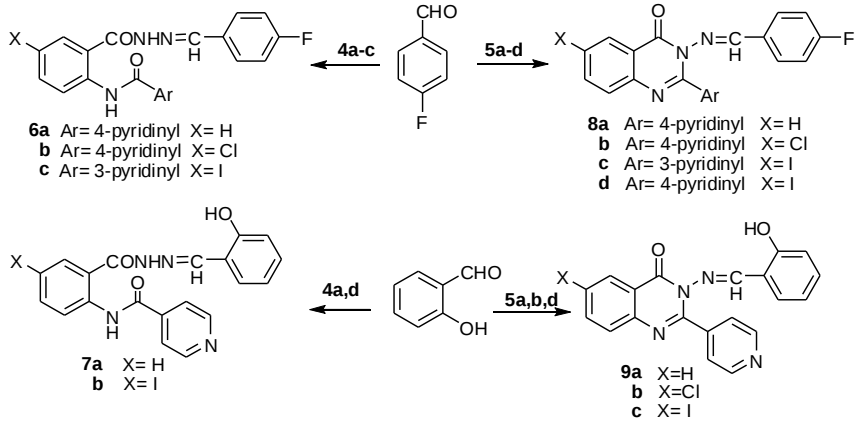
Scheme 1

IR spectra of the hydrazides **4a-d** revealed the presence of NH,NH_2 group at 3318-3164 cm^{-1}. ^{1}HNMR spectra of **4a** and **d** showed broad bands at 4.72, 4.71 ppm respectively characteristic for amino group and two singlets at 12.22, 12.72 ppm for 2NH, (D_2O-exchangeable). Mass spectrum of compound **4c**: showed a molecular ion peak at: *m/z* = 382 (17.1 %) with a base peak at: m/z = 351 (100%). IR spectra of products **5a-d** showed absence of the characterisitic bands for lactone group and revealed, instead of, bands at 3293-3210 and 1677 cm^{-1} regions for amino and lactam C=O (quinazoline) groups. ^{1}HNMR spectra of **5a, c** and **d** showed broad signals at 5.63, 5.71, and 5.70 ppm characterized for amino group. Mass spectrum of

compound **5b**: showed a molecular ion peak at: *m/z* = 272 (5.8%). Also, mass spectrum of **5c**: exhibited a molecular ion peak (base peak) at: *m/z* = 364 (100%).

Structure of the hydrazides **4a-d** was confirmed chemically by further condensation with aromatic character aldehydes. Thus, the hydrazones **6a-c, 7a-c** were obtained by condensation of the hydrazide derivatives **4a-c** with *p*-fluorobenzaldehyde and salicylaldehyde respectively. IR spectra of **6a-c** showed bands for NH and C=O groups. ^{1}HNMR spectra of **6a, b** showed signals for NH at 11.92-12.15 ppm. Mass spectrum of **6a** showed a molecular ion peak at: *m/z* = 362(2.3%) with a base peak at: m/z = 225 (100%). IR spectra of **7a,b** showed bands for OH, NH and C=O groups (c.f. experimental part). ^{1}HNMR spectrum of **7a** and **b** displayed the expected signals for NH and OH groups.

The present work was continued to synthesize a new series of quinazolinone **8a-d, 9a-c** through condensation of the 3-aminoquinazolinone derivatives **5a-d** with *p*-fluorobenzaldehyde and/or salicylaldehyde in dioxane. Structures of compounds **8a-d** and **9a-c** were realized by careful inspections of their spectral data. IR spectra of **8a-d** showed disappearance of bands for amino group. ^{1}HNMR spectrum of **8a** showed absence of the amino group signal and revealed signal at 9.11 ppm for azomethine group. Mass spectra of **8b,c** showed a molecular ion peak at: m/z = 378(M^{+};4.5 %) and 470(M^{+}; 9.8 %) respectively. IR spectra of **9a-c** showed the presence of OH in 3395-3149 cm^{-1} region. ^{1}HNMR spectra of **9b,c** showed absence of amino group and revealed signal at: 9.20, 15.50 ppm for azomethine and OH group respectively. Mass spectrum of **9b:** showed a molecular ion peak at: m/z = 376 with a base peak at: m/z = 257(M^{+}; 59.6 %) (Scheme **2**).

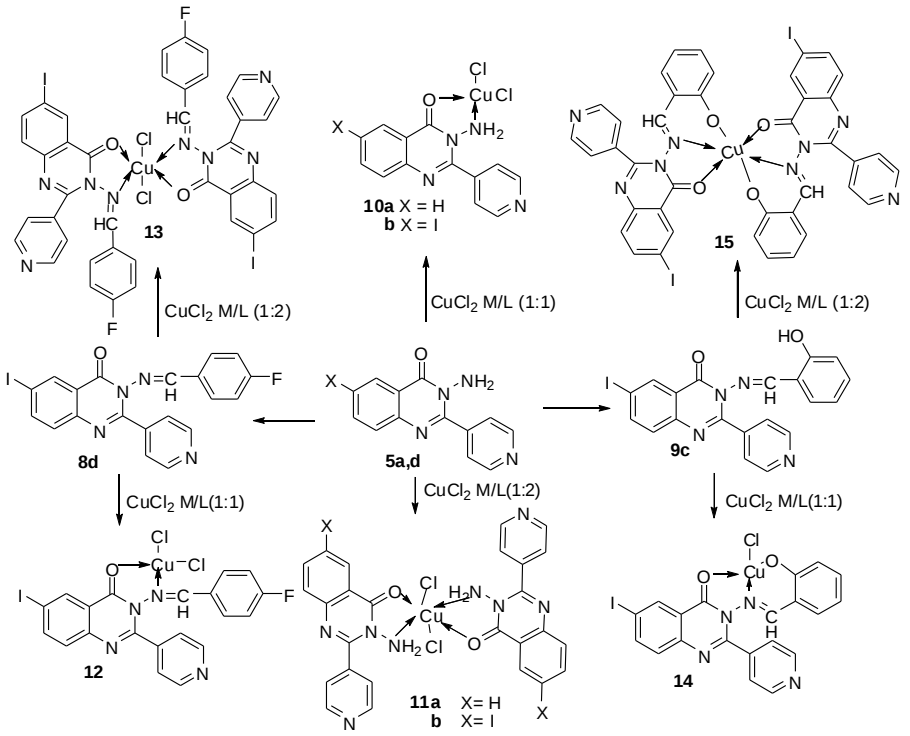


Scheme 2

Due to the successful preparation of 3-amino-2- pyridinyl-4(3*H*)-quinazolinone derivatives **5** and its aryldine derivatives **8** the attention was directed, in the present work, to investigate their chemical behavior towards metal cations. Therefore, it was

encouraged to synthesize some new copper complexes from the compounds **8** for their expected biological activities. Thus, when a solution of the divalent metal ions $CuCl_2$ (0.01 mole) was added to a stirred solution of **5a, d, 8d** and **9c** (0.01 mole) in dioxane, at reflux temperature, the corresponding complexes **10a, b, 12, 14** were formed. On the other hand, the corresponding bis-complexes **11a,b, 13** and **15** were obtained when a solution of the $CuCl_2$ (0.01 mole) was added to a stirred solution of **5a,d, 8d** and **9c** (0.02 mole) in dioxane at reflux temperature (Scheme **3**).

All the synthesized copper complexes are stable at room temperature, non-hygroscopic, decompose at higher temperature >250°C, insoluble in water and many of the common organic solvents, but soluble in DMF. Elemental analyses of the isolated complexes indicated that the composition of products **10a, b** can be represented as $CuL.Cl_2$ and of **11a, b** as CuL_2Cl_2.



Scheme 3

IR spectra of the complexes **10 a, b** and **11 a, b** showed shifted NH_2 and C=O bands around 3179-3156 , 3214-3177 and 1767-1766 , 1763-1749 cm^{-1} respectively, when compared to the bands of their ligands **5a,d**; which indicated the participation

of NH_2 , C=O in the metal-ligand bond and thus suggesting that the ligand 3-amino-4(3*H*)-quinazolinone **5** can act as bidentate ligand towards Cu(II) through nitrogen and oxygen atoms of the amino and carbonyl groups. This result was compatible with a previous work on some similar 3-amino-4(3*H*)-quinazolinone derivatives[16-19]. Electronic spectra (DMF solution) of the ligand **5a** and **5d** showed bands at λmax: 288 and 301 nm respectively; these bands could be assigned to $\pi \rightarrow \pi^*$ and $n \rightarrow \pi^*$ transitions. These bands were noticed to be shifted in UV spectra of the complexes **10a, b, 11a, b** and appeared around at λmax: 285 nm. In addition, the spectra of these complexes exhibited a weak band around λmax: 420 nm probably due to charge transfer (CT) from both ligands to the Cu atom.

IR spectral determination of the copper complexes **12** and **13** suggested that the compound **8d** act as bidentate ligand towards Cu (II) through nitrogen and oxygen atoms of the amino and carbonyl groups. This result agreed well with a previous work on some similar derivatives derived from some 3-amino-4(3*H*)-quinazolinones[20-23]. Electronic spectra (DMF solution) of ligand **8d** showed band at λmax: 302 nm which correspond to $\pi \rightarrow \pi^*$ transition of C=N group. The spectrum showed another band at λmax: 270 nm which can be due to $n \rightarrow \pi^*$ transitions of azomethine group[20-23]. One the other hand, the UV spectra of complexes **12** and **13** exhibited two bands around λmax: 303 and 260 nm respectively.

IR spectrum of the free ligand **9c** showed band at: 3288(OH) which disappeared in the IR spectra of the complexes **14** and **15**. Accordingly, the infrared spectral determinations of the copper complexes **14** and **15** suggested that the 3-(2-hydroxybenzylideneamino)-4(3*H*)-quinazolinone acts as tridentate ligand towards Cu (II) through oxygen of C=O group, nitrogen of azomethine and oxygen atom of OH group. This result is in accordance with a previous work on some similar derivatives of 4(3*H*)-quinazolinone[24]. The electronic absorption spectrum (DMF solution) of the free ligand **9c** exhibited bands at: λmax: 304 and 266 nm. The probable assignment for these bands can be due to $n \rightarrow \pi^*$ transitions and associating with the azomethine functions. The spectra of the complexes **14** and **15** showed absorption bands around: λmax: 273 nm.

The acute toxicity is usually measured by the median lethal dose (LD_{50}) which is the dose that kills 50% of the experimental animals under specified conditions. The acute LD_{50} of the screened compounds were determined by Spearman-Karber method[25]. All the compounds **5a-d, 6a-c, 7a,b, 8a-d** and **9a-c** were well tolerated up to the doses of 1200 mg/kg without any toxic manifestations. This observation indicated the non-toxicity of these compounds and some of their complexes.

Therefore, the main of calculated therapeutic index of the tested compounds was high and this seems a good guide for their safety depending on the rule which states the higher the index, the safer the drug. Thus, the therapeutic index is a good guide to the safety of a drug.

Anticonvulsant activity was screened for some synthesized compounds. Most of the experimental methods used for screening and bioassay of anticonvulsants involved artificial induction of convulsion and their inhibition by the test compounds. Convulsion was carried out using pentylenetetrazole method[26]. The tested compounds (please see the tabulated compounds numbers, Table 1) were administrated to a group of adult rats, 45 min. later *i.p* injection of pentylenetetrazole is given in a dose of 100 mg/Kg. The anterior of anticonvulsant activity was complete protection against convulsion of any kind observations were made for at least 60 minutes after the administration of pentylenetetrazole. Then, the ED_{50} was calculated. Benzodiazepine (Diazepam) was used as standard.

Table 1: The Anticonvulsant activity of the screened compounds:

Compounds Number	Latency (sec)	Duration of seizure (sec)	Mortal
Control (CMC 1%)	118	312	Dead
5a	288	466	Dead
5d	368	783	Dead
8a	128	328	Dead
8d	121	350	Dead
9c	129	317	Dead
Diazepam	3600	0	Alive

The latency time (s) to shocks and convulsions was evaluated. The onset of seizure was found to be 118 sec. and the mean seizure duration was 312 sec. Diazepam protected the animals from developing convulsions. Compound **5d** showed some protection for the animals from developing seizure in comparison with control group; and were of 766, and 783 sec. duration seizure. While compounds **5a**, **8a,d** and **9c** was devoid of any effect on PTZ-shock and convulsion latency. Animals were less active after receiving all of the test compounds than the control rat. The results were given in Table 1. The results of the present study revealed that this compound **5a** exhibited possible activity a matter which needs further careful investigation in this respect in the future.

Analgesic activity was carried out for some synthesized compounds. Experimental model used in this study were selected to investigate Narcotic

analgesic activity of some tested compounds. For this purpose, the hot-plate test[27] to reveal Narcotic analgesic activity. In this method, albino mice (Swiss strain) were put on a hot plate with constant temperature 55 ºC., the time taken by the mice to lick its feet or to jump within a cylinder placed on a hot plate surface was determined. Five test compounds were injected *i.p*. at a dose level of 50 mg/kg into mice. Control group of animals was similarly treated with 1 % CMC. The reaction time was evaluated directly after 0.5 and 1 hrs of injection. Comparison between the narcotic analgesic activity of the tested compounds and the standard Novalgen from weak, moderate and potent analgesic activity was carried out (Table 2).

Table 2: Narcotic analgesic activity at the tested compounds

Compounds Number	Analgesic activity in seconds (Mean)	
	0.5 h.	1.0 h.
Control	7.4	7.5
5a	12.5	16.6
5c	20.1	27.2
5d	20.6	29.0
8c	12.4	15.2
8d	13.8	16.5
9c	7.3	8.4
Novalgen	62.9	65.9

From the tabulated data the following points could be picked out: Compounds **5c,d** showed generally remarkable analgesic activity. Moderate to weak analgesic activity was shown for all other tested compounds. Compound **9c** was excluded because it gave no significant result compared with the Novalgen.

The compounds were tested using the short term *in vitro* cytotoxicity towards Ehrlich Ascites Carcinoma cells (EAC) as a preliminary screening technique of Tryphan Blue Exclusion Method (Cell Viability Test) for their potential cytotoxic activity[28]. This is one of the methods to assess cytotoxicity of anticancer activity. This test is based on the principle that living cell membrane has ability to prevent the entry of dye. Hence, they remain unstained and can be easily distinguished from dead cells, which take the dye. The percentage of viable cells was determined. Results of the short term *in vitro* cytotoxicity of the compounds with the percentage

of nonviable cells are presented in the following Tables. These experiments were carried out mainly with three different concentrations of the compounds.

Table 3: Antitumor activity of the screened compounds:

.Compounds No	cell death at different concentrations after 2 h %		
	µg /ml		
	100	50	25
5a	10	0	0
5d	10	0	0
8d	0	0	0
9b	0	0	0
10a	60	20	10

The results of antitumor activity for some of synthesized 4(3*H*)-quinazolinone containing pyridine moiety **5a,d, 8d, 9b** and the complex **10a** indicated that compounds **5a,d** incorporating NH_2 and pyridinyl groups at C-3 & C-2 and with & without iodine at C-6 position showed weak activity against EAC cells (10%) only at 100 µg/ml. Also the corresponding derivatives **8d** and **10b** were inactive towards the tested EAC cells. Meanwhile, copper complex **10a** of the ligand product **5a** was found to be of high (60%) moderate (20%) and low (10%) activity at the used concentration 100, 50 and 25 µg/ml.

Antimicrobial screening: (*Paper disc method*[29,30]) Six test organisms representing three different microbial groups were used: Group 1: (Gram positive bacteria) *Bacillus subtilis* (ATCC 6633) and *Staphylococcus aureus.* (ATCC 6538) Group 2: (Gram negative bacteria) *Escherichia* coli (ATCC 7839) and *Pseudomonas aeruginosa*. (ATCC 9027) Group 3: (Fungi) *Aspergillus ochraceus wilhelm* and *Fusarium oxysporium* (local strain identified in Regional Center for fungi and its applications in Al-Azher University). The results concerning *in vitro* antimicrobial activities of some synthesized compounds and some of their complexes together with the inhibition zone (mm) and (MIC) values of compared antibiotic and antifungal activities were presented in (Table 4).

Table 4. Antimicrobial activities data of some synthesized compounds.

Compd. **No.**	*Gram +ve*		*Gram –ve*		*Fungi*	
	B. Sub.	*Staph.*	*E. coli*	*Psedom.*	*Asperg.*	*fuzarium*
3c	07	0	8	15	0	16 (12.5)
3e	08	12	10	18 (25)	0	0
5b	6	0	0	0	0	0
5c	0	0	0	0	19 (12.5)	16 (12.5)
10a	7	17	0	0	0	0
11b	7	19 (12.5)	0	0	0	13
Erythromycin	30	19	15	16	-	-
Noroxin	35	27	17	15	-	-
Nystatin	-	-	-	-	12	20

(A) The tabulated antimicrobial screening results showed the following:

Benzoxazine derivatives **3c,e** were of slight to high activity against Gram +ve bacteria and Gram –ve tested bacteria. Aminoquinazolinone derivatives **5b,c** showed no activity against all tested bacteria species except **5b** was of slight activity against *B. sub*. Complexes **10a** and **11b** were slightly active against *B. sub*., highly active against *Staph. sp*. and of no activity against Gram –ve bacteria (*E. coli* & *pseudomonus*).

(B) Antifungal screening results showed the following:

Benzoxazinone **3c,e** showed no activity against the tested fungi; except **3c** which showed high activity towards *Fuzarium sp*. Aminoquinazolinone **5b** was of no activity towards all two tested organism but compound **5c** showed high activity. Copper complex **10a** showed no activity towards all the tested organism. On the other hand, biscomplex **11b** was highly active against *fuzarium sp.* and of no activity against *Aspergillus sp*.

Conclusions

The present work included the synthesis of a range of biologically important heterocycles, including 3,1-benzoxazine, 3-amino-4(3*H*)-quinazolinone, their a new series of corresponding azomethine derivatives and their corresponding copper complexes. The syntheses of these compounds have permitted us to further analyze the SAR for screened compounds. Animal toxicity studies indicated the non-toxicity of 4(3*H*)-quinazolinone derivatives. From the tabulated data the following points could be picked out: 3-amino-4(3*H*)-quinazolinone containing pyridine moiety

showed activities as anticonvulsant, analgesic and antitumor, while the corresponding azomethine derivatives were inactive as anticonvulsant, analgesic and antitumor. Meanwhile, its corresponding copper complex was found to be of high activity as antitumor. Antimicrobial screening results showed the following: benzoxazine derivatives were high activity against tested bacteria. 3-Amino-4(3*H*)-quinazolinone derivatives showed no activity against all tested bacteria species While, their complexes were active against *Staphylococcus aureus.*

Experimental Section

All melting points are uncorrected. Microanalyses were carried out by the Microanalytical Laboratory, National Research Center, Cairo, Egypt and the Microanalytical Research Center, Faculty of Science, Cairo University. Infrared spectra (KBr-disc) were recorded using a Jasco FT/IR-300E spectrophotometer and FTIR 5300 spectrometer (ν, cm^{-1}). ^1H NMR spectra were recorded using Varian mercury 300 MHz & Varian Gemini 200 MHz with chemical shift in δ from Me_4Si and Jeol 270, 500 MHz. Mass spectra were recorded on GC/MS finnigan SSQ 7000 spectrophotometer & GC Ms-QP 1000 EX mass spectrometer at 70 ev. UV spectra were recorded on UV-2401PC, UV-Vis Recording spectrophotometer.

The 2-(pyridine-4-ylamido)-benzoic acid (**2a**) and 2-(pyridin-4-yl)-3,1-benzoxazin-4-ones (**3a**) were synthesized according to a previously reported method[31]

General procedure for the synthesis of 2-(pyridinylamido)-5-substituted–benzoic acid (2b-d)

To a suspension of anthranilic acid derivative (0.01 mole) in toluene (20 ml), pyridine carboxylic acid chloride[31] (0.01 mol) was added dropwise while stirring. After complete addition, the reaction mixture was heated under reflux for 0.5 hour. The solid product obtained was collected by filtration, washed with hot acetic acid then with water and crystallized from DMF to give amide derivatives:

5-Chloro-2-(isonicotinamido) benzoic acid (2b)

M.P. >300 °C, Yield (55%), IR: ν/cm^{-1}: 3350-3200 (OH, NH) and 1681 (C=O). MS, m/z 276 (M^+; 72.5%), 106(13.7%), 278 (23.3%), 153 (35.4%), 124(11.6), 78 (63.9%). Anal. Calcd. for $C_{13}H_9ClN_2O_3$ (276.68): C, 56.43; H, 3.28; N, 10.13; Found: C, 56.40; H, 3.30; N, 10.10.

5-Iodo-2-(nicotinamido)benzoic acid (2c)

M.P. >300 °C, Yield (50%), IR: ν/cm^{-1}: 3447(OH), 3210(NH) and 1676 (C=O). ^{1}HNMR (DMSO-d_6): δ/ppm: 7.58 – 8.80 (m, 7H, ArH + NH), 9.11 (s, 1H, ArH at C_2-H of pyridine), 12.08 (s, 1H, OH). Anal. Calcd for $C_{13}H_9IN_2O_3$ (368.13): C, 42.41; H, 2.46; N, 7.61. Found: C, 42.40; H, 2.50; N, 7.60

5-Iodo-2-(isonicotinamido)benzoic acid (2d)

M.P. >300 °C, Yield (55%) IR: ν/cm^{-1}: 3426(OH), 3257(NH) and 1681 (C=O). MS, m/z 368 (M^+), 369 (16.4%), 245(53.7%), 218(7.0%), 106(78.8%) Anal. Calcd for $C_{13}H_9IN_2O_3$ (368.13): C, 42.41; H, 2.46; N, 7.61. Found: C, 42.40; H, 2.50; N, 7.60.

General procedure for the synthesis of 2-(pyridinyl)-6-(substituted)-3,1-benzoxazin-4-ones (3b-d)

A solution of each of compound **2b** or **c** or **d** (0.01 mole) in acetic anhydride (20 ml) was heated under reflux for 3 hr. and then allowed to cool. The precipitated product was filtrated and crystallized from acetic acid to give benzoxazine derivatives **3b-d.**

6-Chloro-2-(pyridin-4-yl)-4H-benzo[d][1,3]oxazin-4-one (3b*):*

M.P. 173-175 °C, Yield (90%) IR: ν/cm^{-1}: 1773 (C=O). ^{1}HNMR (DMSO-d_6): δ/ppm: 7.78 (d, 1H, *J*= 9.30 Hz, ArH at C_8-H), 8.00 (m, 3H, ArH at C_7-H, AB system $C_{3,5}$-H pyridine), 8.03 (s, 1H, ArH at C_5-H), 8.83 (d, 2H, *J*= 5.90 Hz, AB system $C_{2,6}$-H pyridine). MS, m/z 258 (M^+; 87.1%), 106 (100 %). 259 (24.2%), 260 (33.3%), 261 (7.9%), 231 (7.3%), 214 (34.3%), 78 (70%). Anal. Calcd for $C_{13}H_7ClN_2O_2$ (258.66): C, 60.36; H, 2.73; N, 10.83. Found: C, 60.40; H, 2.70; N, 10.80.

6-Iodo-2-(pyridin-3-yl)-4H-benzo[d][1,3]oxazin-4-one (3c):

M.P. 169-170°C, Yield (85%) IR: ν/cm^{-1}: 1753 (C=O). ^{1}HNMR (DMSO-d_6): δ/ppm: 7.53 (d, 1H, *J*= 8.40 Hz, ArH at C_8-H), 8.05 (d, 2H, *J*= 5.80 Hz, AB system $C_{3,5}$-H pyridine), 8.23 (d, 1H, *J*= 8.40 Hz, ArH at C_7-H), 8.40 (s, 1H, ArH at C_5-H), 8.85 (d, 2H, *J*= 5.80 Hz, AB system $C_{2,6}$-H pyridine) Anal. Calcd for $C_{13}H_7IN_2O_2$ (350.11): C, 44.60; H, 2.02; N, 8.00. Found: C, 44.60; H, 2.00; N, 8.00.

6-Iodo-2-(pyridin-4-yl)-4H-benzo[d][1,3]oxazin-4-one (3d):

M.P. 233-235°C, Yield (90%) IR: ν/cm^{-1}: 1764 (C=O). MS, m/z 350 (90.5%). 351 (58.1%), 352 (7.6%), 306 (29.1%), 272 (28.5%), 78 (96.1%). Anal. Calcd for $C_{13}H_7IN_2O_2$ (350.11): C, 44.60; H, 2.02; N, 8.00. Found: C, 44.60; H, 2.00; N, 8.00.

General procedure for the synthesis of 2-(Pyridinylamido)-5-(substituted)-benzohydrazides (4a-d)

To a solution of the benzoxazine derivative **3a-d** (0.01 mole) in dioxane (30 ml), hydrazine hydrate (0.012 mol) was added. The reaction mixture was stirred for 5 min. at room temperature. The solid product formed was filtered off and crystallized from dioxane to give **4a-d**

N-(2-(hydrazinecarbonyl)-phenyl)isonicotinamide (4a)

M.P. 183-185 °C, Yield (70%) IR: ν/cm^{-1}: 3296, 3214, 3164 (NH, NH_2) and 1676 (C=O). ^{1}HNMR (DMSO-d_6): δ/ppm: 4.72 (b, 2H, NH_2, D_2O-exchangeable), 7.27 (t, 1H, *J*= 7.60 Hz, ArH at C_5-H), 7. 60 (t, 1H, *J*= 7.60 Hz, ArH at C_4-H), 7. 81 (m, 3H, ArH at C_3-H + AB-system $C_{3,5}$-H pyridine), 8.60 (d, 1H, *J*= 8.10 Hz, ArH at C_6-H), 8.82 (d, 2H, *J*= 5.30 Hz, AB-system $C_{2,6}$-H pyridine) 10.22 (b, 1H, NH, D_2O-exchangeable), 12.70 (b, 1H, NH, D_2O-exchangeable). Anal. Calcd for $C_{13}H_{12}N_4O_2$ (256.26) : C, 60.93; H, 4.72; N, 21.86. Found: C, 60.90; H, 4.70; N, 21.90.

N-(4-chloro-2-(hydrazinecarbonyl) phenyl) isonicotinamide (4b):

M.P. 218-220°C, Yield (75%) IR: ν/cm^{-1}: 3272, 3202 (NH, NH_2) and 1682 (C=O). Anal. Calcd for $C_{13}H_{11}ClN_4O_2$ (290.71): C, 53.71; H, 3.81; N, 19.27. Found: C, 53.70; H, 3.80; N, 19.30.

N-(2-(hydrazinecarbonyl)-4-iodophenyl) nicotinamide (4c):

M.P. 188-190°C, Yield (75%) IR: ν/cm^{-1}: 3318, 3257 (NH, NH_2) and 1682 (C=O). MS, m/z 382 (17.1 %), 351 (100%), 383 (4.32%), 363 (33.8%), 246 (19.5%), 78(16.9%). Anal. Calcd for $C_{13}H_{11}IN_4O_2$ (382.16): C, 40.86; H, 2.90; N, 14.66. Found: C, 40.80; H, 2.90; N, 14.70.

N-(2-(hydrazinecarbonyl)-4-iodophenyl) isonicotinamide (4d)

M.P. 248-250°C, Yield (80%) IR: ν/cm^{-1}: 3314, 3274 (NH, NH_2) and 1667 (C=O). ^{1}HNMR (DMSO-d_6): δ/ppm: 4.71 (b, 2H, NH_2, D_2O-exchangeable), 7.81(d, 2H, *J*= 5.50 Hz, AB-system $C_{3,5}$-H pyridine), 7.89 (d, 1H, *J*= 8.50 Hz, ArH at C_3-H), 8.11 (s, 1H, ArH at C_6-H), 8.40 (d, 1H, *J*= 8.50 Hz, ArH at C_4-H), 8.82 (d, 2H, *J*= 5.50 Hz, AB-system $C_{2,6}$-H pyridine), 10.32 (b, 1H, NH, D_2O-exchangeable), 12.60

(b, 1H, NH, D_2O-exchangeable). Anal. Calcd for $C_{13}H_{11}IN_4O_2$ (382.16): C, 40.86; H, 2.90; N, 14.66. Found: C, 40.80; H, 2.90; N, 14.70.

General procedure for the synthesis of 3-amino-2-pyridinyl-6-substituted-4(3*H*)-quinazolinones (5a-d)

A mixture of the benzoxazines **3b-e** (0.01 mol) and hydrazine hydrate (0.012 mol) in *n*-butanol (30 ml) was heated under reflux for 3 hrs. and then allowed to cool. The obtained solid product was filtered off and recrystallized from ethanol to give 3-aminoquinazolinone derivatives **5a-d**

3-Amino-2-(pyridin-4-yl)-quinazolin-4(3*H*)-one (5a)

M.P. 204-205 °C, Yield (85%) IR: ν/cm^{-1}: 3300, 3216 (NH_2) and 1675 (C=O). ^{1}HNMR (DMSO-d_6): δ/ppm: 5.63 (b, 2H, NH_2, D_2O-exchangeable), 7.63 (t, 1H, *J*= 7.60 Hz, ArH at C_7-H), 7. 74 (m, 3H, ArH, AB-system $C_{3,5}$-H pyridine + 1H at C_6-H), 7. 76 (d, 1H, *J*= 7.60 Hz, ArH at C_8-H), 8.21 (d, 1H, *J*= 7.60 Hz, ArH at C_5-H), 8.71 (d, 2H, *J*= 5.90 Hz, AB-system $C_{2,6}$-H pyridine). Anal. Calcd for $C_{13}H_{10}N_4O$ (238.24): C, 65.54; H, 4.23; N, 23.52. Found: C, 65.50; H, 4.20; N, 23.50.

3-Amino-6-chloro-2-(pyridin-4-yl) quinazolin-4(3*H*)-one (5b)

M.P. 221-222°C, Yield (85%) IR: ν/cm^{-1}: 3393, 3312 (NH_2) and 1671 (C=O). MS, m/z 272 (M^+; 5.8%) 78 (100). Anal. Calcd for $C_{13}H_9ClN_4O$ (272.69): C, 57.26; H, 3.33; N, 20.55. Found: C, 57.30; H, 3.30; N, 20.50.

3-Amino-6-iodo-2-(pyridin-3-yl)-quinazolin-4(3*H*)-one (5c*)*

M.P. 184-185°C, Yield (85%) IR: ν/cm^{-1}: 3336, 3227 (NH_2) and 1677 (C=O). ^{1}HNMR (DMSO-d_6): δ/ppm:5.71 (b, 2H, NH_2, D_2O-exchangeable), 7.53 (m, 2H, ArH at C_5-pyidine, ArH at C_8-H quinazoline), 8.10 (d, 1H, *J*= 7.70 Hz, ArH at C_4-H pyidine), 8. 20 (d, 1H, *J*= 8.40 Hz, ArH at C_7-H quinazoline), 8.43 (s, 1H, ArH at C_5-H quinazoline), 8.66 (d, 1H, *J*= 7.70 Hz, ArH at C_6-H pyidine), 8.97 (s, 1H, ArH at C_2-H pyidine). MS, m/z 364(M^+; 100). 365 (26.3%), 363 (62.5%), 347 (28.4%), 335 (26.9%), 245 (26.3%), 208(22.4%), 179 (18.5%). Anal. Calcd for $C_{13}H_9IN_4O$ (364.14): C, 42.88; H, 2.49; N, 15.39. Found: C, 42.90; H, 2.50; N, 15.40.

3-Amino-6-iodo-2-(pyridin-4-yl)-quinazolin-4(3*H*)-one (5d)

M.P. 243-245°C, Yield (90%) IR: ν/cm^{-1}: 3308, 3210 (NH, NH_2) and 1670 (C=O). ^{1}HNMR (DMSO-d_6): δ/ppm: 5.70 (b, 2H, NH_2, D_2O-exchangeable), 7.50 (d, 1H, *J*= 8.40 Hz, ArH at C_8-H), 7.72 (d, 2H, *J*= 5.50 Hz, AB-system $C_{3,5}$-H pyridine), 8.13 (d, 1H, *J*= 8.40 Hz, ArH at C_7-H), 8.45 (s, 1H, ArH at C_5-H), 8.7 (d,

2H, *J*= 5.50 Hz, AB-system $C_{2,6}$-H pyridine. Anal. Calcd for $C_{13}H_9IN_4O$ (364.14): C, 42.88; H, 2.49; N, 15.39. Found: C, 42.90; H, 2.50; N, 15.40.

General procedure for the synthesis of N'-(arylidine)-2-(pyridinylamido)-5-(substituted)-benzohydrazides (6a-c and 7a,b)

A mixture of the hydrazide derivatives **4a-c** (0.01 mol) and the corresponding aldehyde derivative (0.01 mol) in dioxane (30 ml) was heated under reflux for 6 hrs. and left to cool. The solid product obtained was filtered off and recrystallized from ethanol to give the Schiff's bases derivatives **6a-c** and **7a,b** .

N-(2-(2-(4-fluorobenzylidene) hydrazinecarbonyl) phenyl) isonicotinamide (6a)

M.P. 218-220°C, Yield (70%) IR: ν/cm^{-1}: 3186 (NH) and 1677, 1657 (C=O). ^{1}HNMR (DMSO-d_6): δ/ppm: 7.25 (m, 3H, ArH]), 7.60 (t, 1H, *J*= 7.60 Hz, ArH), 7.70 – 7.95 (m, 5H, ArH]), 8.43 (d, 2H, *J*= 5.30 Hz, AB-system), 8.84 (m, 2H, ArH + CH=N), 12.00 (s, 1H, NH, D_2O-exchangeable), 12.15 (s, 1H, NH, D_2O-exchangeable).. MS, m/z 362(M^+; 2.3%) 225 (100%), m/z = 363 (15.4%), 137 (8.9%), 119 (3.5%), 105 (11.7%), 77 (7.6%). Anal. Calcd for $C_{20}H_{15}FN_4O_2$ (362.36): C, 66.29; H, 4.17; N, 15.46. Found: C, 66.30; H, 4.20; N, 15.50

N-(4-chloro-2-(2-(4-fluorobenzylidene) hydrazinecarbonyl) phenyl) isonicotinamide (6b)

M.P. 184-185°C, Yield (85%) IR: ν/cm^{-1}: 3184 (NH) and 1677 (C=O). ^{1}HNMR (DMSO-d_6): δ/ppm: 7.3 (m, 2H, ArH), 7.6 (d, 2H, J= 8.40 Hz, AB-system), 7.8 (m, 3H, [AB-system + ArH]), 8.0 (s, 1H, ArH), 8.5 (m, 2H, [ArH + CH=N), 8.9 (d, 2H, J= 5.30 Hz, AB-system), 11.92 (b, 1H, NH, D_2O-exchangeable), 12.22 (b, 1H, NH, D_2O-exchangeable). Anal. Calcd for $C_{20}H_{14}ClFN_4O_2$ (396.80): C, 60.54; H, 3.56; N, 14.12. Found: C, 60.50; H, 3.60; N, 14.10.

N-(2-(2-(4-fluorobenzylidene)-hydrazinecarbonyl)-4-iodophenyl)nicotinamide (6c*)*

M.P. 223-225°C, Yield (70%) IR: ν/cm^{-1}: 3400, 3200 (NH) and 1677, 1645 (C=O). Anal. Calcd for $C_{20}H_{14}FIN_4O_2$ (488.25): C, 49.20; H, 2.89; N, 11.47. Found: C, 49.20; H, 2.90; N, 11.50.

N-(2-(2-(2-hydroxybenzylidene)-hydrazinecarbonyl)-phenyl) isonicotinamide (7a)

Ethanol M.P. 215-217°C, Yield (70%) IR: ν/cm^{-1}: 3324, 3176 (OH, NH) and 1683 (C=O). ^{1}HNMR (DMSO-d_6): δ/ppm: 6.92-7.02 (m, 2H, ArH), 7.31-7.40 (m, 2H, ArH), 7.60 (d, 1H, *J*= 6.70 Hz, ArH), 7.66 (t, 1H, *J*= 8.40 Hz, ArH), 7.86 (d, 2H, *J*= 5.10 Hz, AB-system), 7.95 (d, 1H, *J*= 7.55 Hz, ArH), 8.46 (d, 1H, *J*= 8.40 Hz, ArH), 8.69 (s, 1H, CH=N), 8.85 (d, 2H, *J*= 5.10 Hz, AB-system), 11.12 (s, 1H, NH, D_2O-exchangeable) 11.97 (b, 1H, NH, D_2O-exchangeable), 12.31 (b, 1H, OH, D_2O-exchangeable). Anal. Calcd for $C_{20}H_{16}N_4O_3$ (360.37): C, 66.66; H, 4.48; N, 15.55. Found: C, 66.70; H, 4.50; N, 15.50.

N-(2-(2-(2-hydroxybenzylidene)-hydrazinecarbonyl)-4-iodophenyl) isonicotinamide (7b)

Dioxane M.P. 248-250°C, Yield (75%) IR: ν/cm^{-1}: 3152 (OH, NH) and 1682 (C=O). ^{1}HNMR (DMSO-d_6): δ/ppm: 6.9-7.0 (m, 2H, ArH), 7.3 (t, 1H, ArH), 7.6 (d, 1H, ArH), 7.8 (d, 2H, AB-system), 7.9 (d, 1H, ArH), 8.1-8.3 (m, 2H, ArH), 8.6 (s, 1H, CH=N), 8.9 (d, 2H, AB-system), 11.0 (s, 1H, NH, D_2O-exchangeable 11.92 (b, 1H, NH, D_2O-exchangeable), 12.30 (b, 1H, OH, D_2O-exchangeable). Anal. Calcd for $C_{20}H_{15}N_4O_3$ (486.26): C, 49.40; H, 3.11; N, 11.52. Found: C, 49.40; H, 3.10; N, 11.50.

General procedure for the preparation of 3-[4-Aryleneamino]-2-(pyridinyl)-6-substituted-4(3*H*) quinazolinones (8a-d and 9a-c)

A mixture of 3-aminoquinazolinone derivative **5a-d** (0.01 mol) and the desired aldehyde (0.01 mol) in dioxane (30 ml) was heated under reflux for 10 hrs. and left to cool. The solid obtained was filtered and crystallized from the proper solvent to give:

3-(4-Fluorobenzylideneamino)-2-(pyridin-4-yl) quinazolin-4(3*H*)-one (8a):

Ethanol M.P. 193-195°C, Yield (70%) IR: ν/cm^{-1}: 1678 (C=O). ^{1}HNMR (DMSO-d_6): δ/ppm: 7.37 (t, 2H, *J*= 8.82 Hz, ArH), 7.64 – 7.69 (m, 3H, ArH), 7.77 – 7.82 (m, 3H, ArH), 7.93 (t, 1H, *J*= 7.12 Hz, ArH), 8.27 (d, 1H, ArH at C_5-H), 8.70 (d, 2H, *J*= 5.90 Hz, AB-system), 9.11(s, 1H, CH=N). Anal. Calcd for $C_{20}H_{13}FN_4O$ (344.34): C, 69.76; H, 3.81; N, 16.27. Found: C, 69.80; H, 3.80; N, 16.30.

6-Chloro-3-(4-fluorobenzylideneamino)-2-(pyridin-4-yl)-quinazolin-4(3*H*)-one (8b):

Dioxane M.P. 243-245°C, Yield (70%) IR: ν/cm^{-1}: 1684 (C=O). MS, m/z 378 (M^+; 4.5%), 179 (11.5%), 152 (49.5 %), 110 (12.4%), 75 (13.1%). Anal. Calcd for

$C_{20}H_{12}ClFN_4O$ (378.79): C, 63.42; H, 3.19; N, 14.79. Found: C, 63.40; H, 3.20; N, 14.80.

3-(4-Fluorobenzylideneamino)-6-iodo-2-(pyridin-3-yl)-quinazolin-4(3*H*)-one(8c):

Dioxane M.P. 214-215 °C, Yield (70%) IR: ν/cm^{-1}: 1681 (C=O). MS, m/z 470 (M^+; 9.8%), 349 (100%), 375(15.0%), 350 (18.3%), 245 (86.0%), 179 (21.6%). Anal. Calcd for $C_{20}H_{12}FIN_4O$ (470.24): C, 51.08; H, 2.57; N, 11.91. Found: C, 51.10; H, 2.60; N, 11.90.

3-(4-Fluorobenzylideneamino)-6-iodo-2-(pyridin-4-yl)-quinazolin-4(3*H*)-one (8d)

Dioxane M.P. >300°C, Yield (70%) IR: ν/cm^{-1}: 1684 (C=O). MS, m/z 470 (M; 3.4%), 349 (100%), 471(1.8%), 306 15.9%), 245 (24.5%), 179 (20.9%), 108 (50%). Anal. Calcd for $C_{20}H_{12}$ FIN_4O (470.24): C, 51.08; H, 2.57; N, 11.91. Found: C, 51.10; H, 2.60; N, 11.90.

3-(2-Hydroxybenzylideneamino)-2-(pyridin-4-yl)-quinazolin-4(3*H*)-one (9a)

MeOH M.P. 169-170 °C, Yield (75%) IR: ν/cm^{-1}: 3149(OH) and1675 (C=O). Anal. Calcd for $C_{20}H_{14}N_4O_2$ (342.35): C, 70.17; H, 4.12; N, 16.37. Found: C, 70.20; H, 4.10; N, 16.40.

6-Chloro-3-(2-hydroxybenzylideneamino)-2-(pyridin-4-yl)-quinazolin-4(3*H*)-one (9b)

MeOH M.P. 223-225 °C, Yield (80%) IR: ν/cm^{-1}: 3395 (OH) and1691 (C=O). ^{1}HNMR (DMSO-d_6): δ/ppm: 6.80 (t, 1H, ArH at C_4-H phenol), 6.90 (d, 1H, ArH at C_3-H phenol), 7.30 (t, 1H, ArH at C_5-H phenol), 7.40(d, 1H, ArH at C_6-H phenol), 7.50 (d, 2H, *J*= 5.50 Hz, AB-system $C_{3,5}$-H pyridine), 7.80 (d, 1H, ArH at C_8-H quinazoline), 7.90 (d, 1H, ArH at C_7-H quinazoline), 8.10 (s, 1H, ArH at C_5-H quinazoline), 8.60 (d, 2H, *J*= 5.50 Hz, AB-system $C_{2,6}$-H pyridine), 9.20 (s, 1H, CH=N), 10.50 (s, 1H, OH). MS, m/z 376 (M^+; 59.6%) 257(100), 377 (2.3%), 214 (6.4%), 179(12.4%), 153(71.6%), 78 (10.7%).Anal. Calcd for $C_{20}H_{13}ClN_4O_2$ (376.80): C, 63.75; H, 3.48; N, 14.87. Found: C, 63.70; H, 3.48; N, 14.92.

3-(2-Hydroxybenzylideneamino)-6-iodo-2-(pyridin-4-yl)-quinazolin-4(3*H*)-one (9c)

Dioxane M.P. 269-270°C, Yield (85%) IR: ν/cm^{-1}: 3152 (OH) and1682(C=O). ^{1}HNMR (DMSO-d_6): δ/ppm: 6.80 (t, 1H, ArH at C_4-H phenol), 6.90 (d, 1H, ArH at C_3-H phenol), 7.39 (t, 1H, ArH at C_5-H phenol), 7.50 (d, 1H, ArH at C_6-H phenol), 7.55 (d, 1H, *J*= 8.40 Hz, ArH at C_8-H quinazoline), 7.65 (d, 2H, *J*= 5.50 Hz, AB-

system $C_{3,5}$-H pyridine), 8.18 (d, 1H, *J*= 8.40 Hz, ArH at C_7-H quinazoline), 8.50 (s, 1H, ArH at C_5-H quinazoline), 8.70 (d, 2H, *J*= 5.50 Hz, AB-system $C_{2,6}$-H pyridine), 9.20 (s, 1H, CH=N), 10.45 (s, 1H, OH). MS, m/z 468 (M^+; 0.2%) 364(100) 208 (22.7%), 179 (8.8%), 286(58.9%), 272(53.8%), 185 (23.8%). Anal. Calcd for $C_{20}H_{13}IN_4O_2$: C, 51.30; H, 2.80; N, 11.97. Found: C, 51.30; H, 2.80; N, 12.00.

General procedure for the preparation of Cu (II) complexes (10a,b, 12 and 14)

A solution of cupric chloride (0.01 mol) dissolved in minimum quantity of ethanol-water (1:½, v/v) was added while stirring to a solution of **5a,d , 8d** or **9c (**0.01 mol) in dioxane (20 ml) and the reaction mixture was heated under reflux for 3 h. The solid product which formed while hot was collected by filtration, washed with hot water (10 ml) followed by small quantity of dioxane and dried to give **10a,b,12 and 14,** respectively as greenish crystals.

Dichloro 3-Amino-2-(pyridin-4-yl)-quinazolin-4(3*H*)-one copper(II) complex (10a)

D.P. 257-261 °C Yield (90%). IR: ν/cm^{-1}: 3246 (asNH_2), 3156(sNH_2), 1603 (NH_2), 1767(C=O), 534(Cu-N).and 422(Cu-O).UV-vis (DMSO), λmax: 420 and 285 nm Anal. Calcd for $C_{13}H_{12}Cl_2CuN_4O_2$ (390.5): C, 39.96; H, 3.10; N, 14.34; Cu, 16.26. Found: C, 40.00; H, 3.10; N, 14.30; Cu, 16.20.

Dichloro 3-Amino-6-iodo-2-(pyridin-4-yl)-quinazolin-4(3*H*)-one copper(II) complex (10b)

D.P. 268-271°C Yield (95%). IR: ν/cm^{-1}: 3258 (asNH_2), 3179(sNH_2), 1593 (NH_2), 1766(C=O), 509(Cu-N).and 444(Cu-O). UV-vis (DMSO), λmax: 285 nm Anal. Calcd for $C_{13}H_{11}Cl_2CuIN_4O_2$ (516.5): C, 30.22; H, 2.15; N, 10.85; Cu, 12.30; Found: C, 30.20; H, 2.10; N, 10.80; Cu, 12.30.

Dichloro 3-(4-Fluorobenzylideneamino)-6-iodo-2-(pyridin-4-yl)-quinazolin-4(3*H*)-one copper(II) complex (12)

D.P. < 300 °C Yield (90(IR: ν/cm^{-1}: 1684(C=O), 1590(C=N), 580(Cu-N) and 449(Cu-O). UV-vis (DMSO), λmax: 303 and 267 nm Anal. Calcd for $C_{20}H_{14}Cl_2CuFIN_4O_2$ (622.7): C, 38.58; H, 2.27; N, 9.00; Cu, 10.20; Found: C, 38.60; H, 2.30; N, 9.00; Cu, 10.20

Monochloro 3-(2-Hydroxybenzylideneamino)-6-iodo-2-(pyridin-4-yl)-quinazolin-4(3*H*)-one copper(II) complex (14)

D.P.< 300 °C Yield (90(IR: ν/cm^{-1}: 3408(OH),1673(C=O), 1595(C=N), 589(Cu-N) and 431(Cu-O). UV-vis (DMSO), λmax: 274 nm Anal. Calcd for $C_{21}H_{17}ClCuIN_4O_3$ (599.3): C, 42.09; H, 2.86; N, 9.35; Cu, 10.60; Found: C, 42.10; H, 2.90; N, 9.40; Cu, 10.60

General procedure for the preparation of bis Cu (II) complexes (11a,b,13 and 15)

To a solution of **5a,d,8d** or **9c** (0.02 mol) in dioxane (25 ml), a solution of $CuCl_2$ (0.01 mol) in ethanol-water (1:½, v/v) 3 ml was added. The reaction mixture was heated under reflux while stirring for 10 h. The solid product that formed while hot was separated by filtration, washed with dioxane and dried to gave **11a,b,13 and 15** respectively as greenish crystals.

Dichloro Bis [3-Amino-2-(pyridin-4-yl)-quinazolin-4(3*H*)-one] copper(II) complex (11a)

D.P. 277-281°C Yield (80%). IR: ν/cm-1: 3295 (asNH_2), 3177(sNH_2), 1601 (NH_2), 1763(C=O), 535(Cu-N) and 438(Cu-O). UV-vis (DMSO), λmax: 285 nm Anal. Calcd for $C_{26}H_{22}Cl_2CuN_8O_3$ (629): C, 49.65; H, 3.53; N, 17.82; Cu, 10.10; Found: C, 49.70; H, 3.50; N, 17.80; Cu, 10.10

Dichloro Bis [3-Amino-6-iodo-2-(pyridin-4-yl)-quinazolin-4(3*H*)-one] copper(II) complex (11b)

D.P. 279-283°C Yield (90%). IR: ν/cm-1: 3303 (asNH_2), 3214(sNH_2), 1593 (NH_2), 1749(C=O), 511(Cu-N) and 441(Cu-O). UV-vis (DMSO), λmax: 422 and 286 nm Anal. Calcd for $C_{26}H_{20}Cl_2CuI_2N_8O_3$ (880.7): C, 35.46; H, 2.29; N, 12.72; Cu, 7.21; Found: C, 35.50; H, 2.30; N, 12.70; Cu, 7.20

Dichloro Bis [3-(4-Fluorobenzylideneamino)-6-iodo-2-(pyridin-4-yl)-quinazolin-4(3*H*)-one] copper(II) complex (13)

D.P. <300 °C Yield (90(IR: ν/cm^{-1}: 1684(C=O), 1590(C=N), 580(Cu-N) and 447(Cu-O). UV-vis (DMSO), λmax: 303 and 248 nm Anal. Calcd for $C_{40}H_{26}Cl_2CuF_2I_2N_8O_3$ (1092.9): C, 43.96; H, 2.40; N, 10.25; Cu, 5.8; Found: C, 44.00; H, 2.40; N, 10.30; Cu, 5.80

Bis [3-(2-Hydroxybenzylideneamino)-6-iodo-2-(pyridin-4-yl)-quinazolin-4(3*H*)-one] copper(II) complex (15)

D.P. < 300 °C Yield (85(IR: ν/cm^{-1}: 3408(OH), 1669(C=O), 1595(C=N), 584(Cu-N) and 422(Cu-O). UV-vis (DMSO), λmax: 273 nm Anal. Calcd for $C_{41}H_{29}CuI_2N_8O_5$ (1031): C, 47.76; H, 2.83; N, 10.87; Cu, 6.16; Found: C, 47.80; H, 2.80; N, 10.90; Cu, 6.20

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