# An Efficient Synthesis of Quinazoline Derivatives With the Aid of Low-valent Titanium Reagent

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Quinazolin-4(3*H*)-ones, 1,2-dihydroquinazolin-4(3*H*)-ones, 3,4-dihydroquinazolines, imidazo[1,2-*c*]-quinazolines and 5,6-dihydroimidazo[1,2-*c*]-quinazolines were synthesized by the novel reductive reaction of nitro group, N-H bond and ortho-ester, aldehydes or ketones promoted by the low-valent titanium reagent (TiCl<sub>4</sub>-Zn system). The structures of these compounds were characterized by elemental analysis, IR and <sup>1</sup>HNMR spectra and further confirmed by single crystal X-ray diffraction analysis.

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The pyrimidine ring constitutes a basic heteroaromatic structure. It is a vital building block for the construction of other heterocyclic ring compounds and alkloids. A large number of compounds containing the pyrimidine ring possess a wide range of pharmacological activities. Quinazolin-4(3H)-one derivatives (I) have been reported to exhibit anticonvulsant [1], antihypertensive [2], antidiabetic [3], antibacterial [4], antitumor [5], antihistaminic [6] and anti-inflammatory [7] activities. Some polyheterocyclic structures such as indole[1,2-c]quinazoline (II) and benzimidazo[1,2-c]quinazoline (III) skeletons have been reported to represent potent cytotoxic agents [8] (Figure 1). Therefore, the development of novel and convenient synthetic methods for the preparation of quinazolin-4(3H)-ones and quinazolines still remains an active research area.

Low-valent titanium reagents have an exceeding high ability to promote reductive coupling of carbonyl compounds and are attracting increasing interest in organic synthesis [9]. Many other functional groups can be reacted [10]. Recently, we have reported the low-valent titanium-induced intermolecular reductive coupling reaction of carboxylic derivatives with aromatic ketones [11], the intramolecular reductive coupling reaction of 4,4-dicyano-1,3-diaryl-1- butanone [12] and the cyclodimerization of  $\alpha,\beta$ -unsaturated ketones [13]. In the course of our work on the application of low-valent titanium reagents in the preparation of bioactive heterocyclic compounds, we have reported the synthesis of indoles [14], 2-aminoquinolines

[15] and 2-arylquinolines [16] with the aid of low-valent titanium reagent. We have reported the primary results for the synthesis of the quinazolin-4(3H)-ones and 1,2-dihydroquinazolin-4(3H)-ones with the aid of a low-valent titanium reagent [17]. Here, we wish to describe in detail the methods induced by the TiCl<sub>4</sub>-Zn system for the preparation of quinazolin-4(3H)-ones, quinazolines and imidazo[1,2-c]quinazolines.

When *N*-substituted-*o*-nitrobenzamides **1** and triethyl orthoformate **2** were treated with low-valent titanium prepared from titanium tetrachloride and zinc powder in anhydrous THF, the intermolecular reductive cross-coupling products 3-substituted quinazolin- 4(3*H*)-ones **3** were obtained (Scheme 1).

## Scheme 1

Table 1 summarized our results on the reaction of N-substituted-o-nitrobenzamides and triethyl orthoformate with low-valent titanium reagent. The chloro and bromo groups of the substrates could not be reduced under the reaction conditions and no influence on the rate of reductive coupling reaction was observed. On treating triethyl orthoformate with N-phenyl-o-aminobenzamide under the same reaction conditions, no reaction took place and no 3-phenylquinazolin-4(3H)-one could be detected.

Treatment of o-nitrobenzyl amines **4** and triethyl orthoformate **2** with TiCl<sub>4</sub>/Zn in anhydrous THF under the same reaction conditions afforded 3,4-dihydroquinazolines **5** in good yields (Scheme 2). Table 2 summarized our results.

Table 1
Synthesis of 3-Substituted Quinazolin-4(3H)-ones Induced by Low-valent Titanium Reagent

Entry	$\mathbb{R}^1$	$\mathbb{R}^2$	Yield(%)
3a	Н	$C_6H_5$	84
3b	H	$4-CH_3C_6H_4$	84
3c	H	C <sub>6</sub> H <sub>5</sub> CH <sub>2</sub>	71
3d	Cl	$C_6H_5CH_2$	79
3e	Cl	$4-ClC_6H_4CH_2$	72
3f	Cl	C <sub>6</sub> H <sub>11</sub> -c	46
3g	H	$4\text{-IC}_6\text{H}_4$	73
3h	Cl	$4-IC_6H_4$	81
3i	H	$2-\text{ClC}_6\dot{\text{H}}_4$	72
3j	Cl	$2-ClC_6H_4$	86

Table 2 Synthesis of 3,4-Dihydroquinazolines Promoted by Low-valent Titanium Reagent

Entry	R	Ar	Yield(%)
5a	Н	4-ClC <sub>6</sub> H <sub>4</sub>	73
5b	Н	$4-BrC_6H_4$	84
5c	Cl	C <sub>6</sub> H <sub>5</sub>	71
5d	Cl	4-CH3C6H4	83
5e	C1	4-ClC <sub>6</sub> H <sub>4</sub>	79
5f	Cl	$4-BrC_6H_4$	82

Similarly, 2-(o-nitrophenyl)imidazole **6** and ortho-ester **7** were treated with low-valent titanium reagent in anhydrous THF under the same reaction conditions. The desired products imidazo[1,2-c]quinazolines **8** were obtained in moderate yields (Scheme 3). The results are summarized in Table 3.

Scheme 2

Moroever, treatment of o-nitrobenzamides  $\mathbf{1}$  and ketones or aromatic aldehydes  $\mathbf{9}$  with  $\mathrm{TiCl_4/Zn}$  in anhydrous THF under the same reaction conditions afforded 1,2-dihydro-

Scheme 3

Table 3 Synthesis of IImidazo[1,2-c]quinazolines Induced by Low-valent Titanium Reagent

Entry	Ar	X	Y	R	Yield (%)
8a	$C_6H_5$	Н	Н	Н	68
8b	$C_6H_5$	Cl	Н	H	74
8c	$C_6H_5$	CH <sub>3</sub> O	CH <sub>3</sub> O	H	62
8d	$C_6H_5$	Н	Н	$CH_3$	70
8e	$C_6H_5$	$CH_3O$	$CH_3O$	$CH_3$	61
8f	$C_6H_5$	Cl	Н	$CH_3$	65
8g	$4-CH_3C_6H_4$	Н	Н	Н	68
8h	$4-CH_3C_6H_4$	Cl	Н	H	72
8i	4-BrC <sub>6</sub> H <sub>4</sub>	Н	Н	H	63
8j	$4$ -BrC $_6$ H $_4$	Cl	Н	H	69

quinazolin-4(3*H*)-ones **10** in good yields (Scheme 4). Table 4 summarized our results. All reactions could be carried out under mild conditions. However, *N*-phenyl-*o*-nitrobenzamide failed to react with butanone, 3-pentanone, cyclopentanone, cyclohexanone, benzaldehyde or acetophenone under these conditions, although the reaction of *o*-nitrobenzamide **1** and the cyclic ketone **11** with the same reagent system afford 2,2-polymethylene-1,2-dihydroquinazolin-4(3*H*)-ones **12** (Scheme 5) and the results are summarized in Table 5. However, *o*-nitrobenzamide failed to react with acetophenone or 1-tetralone.

Scheme 4

Scheme 5

However, treatment of 2-(o-nitrophenyl)imidazolines **6** and ketones or aromatic aldehydes **9** with TiCl<sub>4</sub>/Zn system in dry THF under the same reaction conditions gave the desired cross-coupling products 5,6-dihydroimidazo[1,2-c]quinazolines **13** were obtained in moderated yields (Scheme 6).

 $\begin{tabular}{l} Table 4 \\ Synthesis of 1,2-Dihydroquinazolin-4(3H)-ones Promoted by Low-valent Titanium Reagent \\ \end{tabular}$ 

Entry	$\mathbb{R}^1$	$\mathbb{R}^2$	$\mathbb{R}^3$	$\mathbb{R}^4$	Yield (%)
10a	Н	4-CH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	$CH_3$	CH <sub>3</sub>	88
10b	H	4-ClC <sub>6</sub> H <sub>4</sub>	CH <sub>3</sub>	CH <sub>3</sub>	71
10c	Н	$4-BrC_6H_4$	$CH_3$	CH <sub>3</sub>	83
10d	C1	$C_6H_5$	$CH_3$	CH <sub>3</sub>	85
10e	C1	$4-CH_3C_6H_4$	$CH_3$	CH <sub>3</sub>	89
10f	C1	$4-BrC_6H_4$	CH <sub>3</sub>	CH <sub>3</sub>	87
10g	Н	CH <sub>3</sub> (CH <sub>2</sub> ) <sub>7</sub>	$CH_3$	CH <sub>3</sub>	72
10h	C1	$CH_3(CH_2)_7$	$CH_3$	CH <sub>3</sub>	73
10i	H	$C_6H_5CH_2$	CH <sub>3</sub>	CH <sub>3</sub>	71
10j	Н	H	$CH_3$	$C_2H_5$	79
10k	Н	Н	$C_2H_5$	$C_2H_5$	86
<b>101</b>	C1	Н	CH <sub>3</sub>	$C_2H_5$	74
10m	C1	Н	$C_2H_5$	$C_2H_5$	71
10n	Н	Н	Η	$4-CH_3C_6H_4$	91
10o	Н	Н	Н	3,4-OCH <sub>2</sub> OC <sub>6</sub> H <sub>3</sub>	93
10p	C1	Н	Н	4-CH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	80
10q	Cl	Н	Н	4-CH <sub>3</sub> OC <sub>6</sub> H <sub>4</sub>	87
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Table 5
Synthesis of 2,2-Polymethylene-1,2-dihydroquinazolin-4(3*H*)-ones Induced by Low-valent Titanium Reagent

Entry	R	n	Yield (%)
12a	Н	1	84
12b	H	2	63
12c	C1	1	89
12d	Cl	2	83

Scheme 6

Table 6 summarizes our results. All reactions could be carried out under mild conditions. However, 2-(o-nitrophenyl)imidazoles failed to react with butanone, 3-pentanone, cyclohexanone or acetophenone under these conditions.

 ${\it Table \ 6}$  Synthesis of 5,6-Dihydroimidazo[1,2-c]quioazolines Promoted by Lowvalent Titanium Reagent

Entry	Ar	X	Y	$R^1$	$\mathbb{R}^2$	Yield (%
13a	$C_6H_5$	Н	Н	$CH_3$	$CH_3$	71
13b	$C_6H_5$	Cl	Н	$CH_3$	$CH_3$	74
13c	$C_6H_5$	$OCH_2$	O	$CH_3$	$CH_3$	62
13d	$4-CH_3C_6H_4$	Н	Η	$CH_3$	$CH_3$	82
13e	$4-CH_3C_6H_4$	Cl	Н	$CH_3$	CH <sub>3</sub>	84

Table 6

Entry	Ar	X	Y	$\mathbb{R}^1$	$\mathbb{R}^2$	Yield (%)
13f	$4$ -BrC $_6$ H $_4$	Н	Н	$CH_3$	CH <sub>3</sub>	80
13g	$4$ -BrC $_6$ H $_4$	C1	H	$CH_3$	CH <sub>3</sub>	83
13h	$4-CH_3OC_6H_4$	H	H	$CH_3$	CH <sub>3</sub>	69
13i	$4-CH_3OC_6H_4$	Cl	H	$CH_3$	CH <sub>3</sub>	73
13j	$4-FC_6H_4$	Н	H	$CH_3$	CH <sub>3</sub>	75
13k	$C_6H_5$	Н	H	Н	$4-CH_3C_6H_4$	76
131	$C_6H_5$	Н	H	Н	3,4-(CH <sub>3</sub> O) <sub>2</sub> C <sub>6</sub> H	3 81
13m	$C_6H_5$	Cl	Н	Н	$4-CH_3C_6H_4$	79
13n	$C_6H_5$	Cl	Н	Н	$4\text{-CH}_3\text{OC}_6\text{H}_4$	70

All the products 3, 5, 8, 10, 12 and 13 were characterized by IR, <sup>1</sup>HNMR and elemental analysis. The structures of 8f, 10a, 12b and 13c were further confirmed by single crystal X-ray diffraction analysis. Fig.2 to Fig.5 show the molecular structures of 8f, 10a, 12b and 13c, respectively. The crystallographic data of these compounds are summarized in Table 7

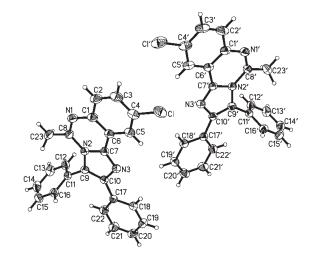


Figure 2 X-ray structure of 8f.

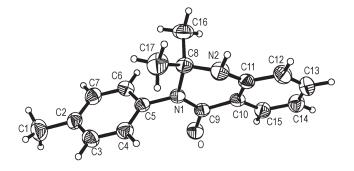


Figure 3 X-ray structure of 10a.

Table 7
Crystallographic Data for **8f**, **10a**, **12b** and **13c** 

	8f	10a	12b	13c
Empirical formula	$C_{23}H_{16}CIN_3$	$C_{17}H_{18}N_2O$	$C_{13}H_{16}N_2O$	$C_{25}H_{21}N_3O_2$
Formula weight	369.84	266.33	216.28	395.45
Temperature (K)	291(2)	295(2)	296(2)	296(2)
Wave length (Å)	0.71073	0.71073	0.71073	0.71073
Crystal system	Monoclinic	Monoclinic	Monoclinic	Monoclinic
Space group	P2 <sub>1</sub> /c	$P2_1/n$	$P2_1/n$	P2 <sub>1</sub> /c
a (Å)	7.789(1)	11.917(2)	10.387(1)	9.594(2)
b (Å)	17.777(2)	6.911(1)	10.954(2)	16.928(4)
c (Å)	26.040(3)	17.821(4)	10.827(2)	12.865(3)
α (°)	90	90	90	90
β (°)	94.70(1)	98.81(1)	110.77(1)	95.73(2)
γ(°)	90	90	90	90
V (Å <sup>3</sup> )	3593.7(9)	1450.4(5)	1151.8(4)	2079.1(8)
Z	8	4	4	4
Dcalc. (Mg/m <sup>3</sup> )	1.367	1.220	1.247	1.263
Absorption coefficient(mm <sup>-1</sup> )	0.225	0.077	0.080	0.082
F(000)	1536	568	464	832
Crystal size (mm)	0.54x0.48x0.34	0.56x0.52x0.32	0.52x0.48x0.44	0.48x0.38x0.26
θ Range (°)	1.39 to 25.00	1.93 to 25.50	2.34 to 25.00	1.99 to 25.50
Limiting indices	0≤h≤9	0≤h≤14	0≤h≤12	0≤h≤11
	0≤k≤21	0≤k≤8	0≤k≤13	0≤k≤20
	-30≤1≤30	-21≤1≤21	-12≤1≤12	-15≤l≤15
Reflections collected	7325	3202	2681	4424
Independent reflections	6330	2706	2033	3873
Data/restraints/parameters	6330/0/490	2706/0/189	2033/0/146	3873/0/278
Goodness-of-fit on F <sup>2</sup>	0.795	0.921	1.087	0.817
Final R indices [I>2σ(I)]	R <sub>1</sub> =0.0391	$R_1 = 0.0433$	$R_1 = 0.0449$	$R_1 = 0.0482$
	wR=0.0747	wR=0.1076	wR=0.1217	wR=0.1134
R indices (all data)	$R_1 = 0.0983$	$R_1 = 0.0770$	$R_1 = 0.0572$	$R_1 = 0.1065$
	w R=0.0868	w R=0.1194	w R=0.1276	wR=0.1274
Extinction coefficient	0.0035(2)	0.0114(16)	0.068(6)	0.0096(12)
Largest diff. Peak and hole (e•Å-3)	0.136 and -0.225	0.136 and -0.136	0.296 and -0.310	0.281 and -0.204

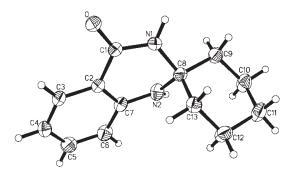


Figure 4. X-ray structure of 12b.

In conclusion, a series of quinazolin-4(3H)-ones, 1,2-dihydroquinazolin-4(3H)-ones, 3,4-dihydroquinazolines, imidazo[1,2-c]quinazolines and 5,6-dihydroimidazo[1,2-c]quinazolines were synthesized via novel reductive reaction of nitro group, N-H bond and ortho-ester or carbonyl group induced by the TiCl<sub>4</sub>/Zn system. The advantages of our methods are the easily accessible starting materials, convenient manipulation and moderate to high yields.

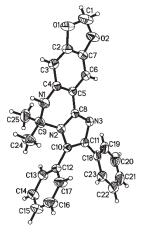


Figure 5. X-ray structure of 13c.

# **EXPERIMENTAL**

Melting point were determined in open capillaries and are uncorrected. IR spectra were recorded on a FTIR-8101 spectrometer. <sup>1</sup>HNMR spectra were measured on an Inova-400 MHz spectrometer

using TMS as internal standard, CDCl<sub>3</sub> as solvent. Microanalysis were carried out on Perkin-Elmer 2400 II instruments. X-ray diffraction was recorded on a Siemens P4 diffractometer.

## General Procedure for the Synthesis of Quinazolin-4(3*H*)-ones.

 $TiCl_4$  (2.2 mL, 20 mmol) was added dropwise using a syringe to a stirred suspension of zinc dust (2.6 g, 40 mmol) in freshly distilled anhydrous THF (20 mL) at room temperature under a dry nitrogen atmosphere. After completion of the addition, the mixture was refluxed for 2 h. The suspension of the low-valent titanium reagent formed was cooled to room temperature and a solution of N-aryl-o-nitrobenzamide (5 mmol) and triethyl orthoformate (10 mmol) in THF was added dropwise. The mixture was refluxed for 5 h under  $N_2$  (the reaction was monitored by TLC). The reaction mixture was quenched with 10% HCl (50 mL) and extracted with CHCl $_3$  (3x50 mL). The combined extracts were washed with water (3x50 mL) and dried over anhydrous  $Na_2SO_4$ . After evaporation of the solvent under reduced pressure, the crude products 3a-j were purified by recrystallization from 95% ethanol.

## 3-Phenylquinazolin-4(3*H*)-one (**3a**).

This compound was obtained as solid with mp 140-142 °C (Lit.18 138-140 °C); <sup>1</sup>HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 8.38 (d, J = 8.4 Hz, 1H, H5), 8.15 (s, 1H, H2), 7.81 (dd,  $J_1$  = 8.4 Hz,  $J_2$  = 7.6 Hz, 1H, H7), 7.78 (d, J = 8.4 Hz, 1H, H8), 7.57 (dd,  $J_1$  = 8.4 Hz,  $J_2$  = 7.6 Hz, 1H, H6), 7.54-7.51 (m, 3H), 7.46-7.43 (m, 2H); IR (KBr): 3030, 1672, 1610, 1473, 1402, 1262, 1181, 1111, 1024, 933, 913, 767, 699 cm<sup>-1</sup>.

## 3-(4'-Methylphenyl)quinazolin-4(3*H*)-one (**3b**).

This compound was obtained as solid with mp 147-149 °C (Lit.18 148-149 °C);  $^1\mathrm{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta=8.38$  ( d, J=8.4 Hz, 1H, H5), 8.13 (s, 1H, H2), 7.81 (dd,  $J_1=8.4$  Hz,  $J_2=8.4$  Hz, 1H, H7), 7.77 (d, J=8.4 Hz, H8), 7.55 (dd,  $J_1=8.4$  Hz,  $J_2=8.4$  Hz, 1H, H6), 7.36 (d, J=8.4 Hz, 2H), 7.31 (d, J=8.4 Hz, 2H), 2.44 (s, 3H, CH<sub>3</sub>); IR (KBr): 3030, 1689, 1600, 1514, 1471, 1323, 1293, 1192, 1114, 1025, 917, 836, 817, 770, 749, 694 cm $^{-1}$ .

## 3-Benzylquinazolin-4(3*H*)-one (3c).

This compound was obtained as solid with mp 114-115 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 8.36 (d, J = 7.6 Hz, 1H, H5), 8.15 (s, 1H, H2), 7.80-7.72 (m, 2H), 7.54 (dd,  $J_1$  = 7.2 Hz,  $J_2$  = 7.6 Hz, 1H), 7.37-7.29 (m, 5H), 5.21 (s, 2H, CH<sub>2</sub>); IR (KBr): 3036, 2945, 1677, 1604, 1473, 1440, 1411, 1365, 1320, 1289, 1161, 1149, 1076, 937, 869, 776, 706, 692 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{15}H_{12}N_2O$ : C, 76.25; H, 5.12; N, 11.86. Found: C, 76.42; H, 4.93; N, 11.90.

## 7-Chloro-3-benzylquinazolin-4(3H)-one (**3d**).

This compound was obtained as solid with mp 119-120 °C;  $^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta = 8.25$  (d, J = 8.8 Hz, 1H, H5), 8.13 (s, 1H, H2), 7.77 (s, 1H, H8), 7.47 (d, J = 8.8 Hz, 1H, H6), 7.39-7.31 (m, 5H), 5.19 (s, 2H, CH<sub>2</sub>); IR (KBr): 2947, 1690, 1601, 1554, 1456, 1400, 1365, 1316, 1216, 1167, 1110, 1070, 946, 894, 876, 819, 781, 756, 715, 699 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{15}H_{11}CIN_2O$ : C, 66.55; H, 4.10; N, 10.35. Found: C, 66.78; H, 3.86; N, 10.51.

## 7-Chloro-3-(4'-chlorobenzyl)quinazolin-4(3*H*)-one (**3e**).

This compound was obtained as solid with mp 139-140 °C;  $^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta = 8.23$  (d, J = 8.4 Hz, 1H, H5),

8.10 (s, 1H, H2), 7.70 (s, 1H, H8), 7.46 (d, J= 8.4 Hz, 1H, H6), 7.34-7.21 (m, 4H), 5.14 (s, 2H, CH<sub>2</sub>); IR (KBr): 3050, 2946, 1672, 1626, 1602, 1530, 1456, 1409, 1350, 1317, 1303, 1232, 1106, 1089, 1015, 910, 841, 798 cm<sup>-1</sup>.

Anal. Calcd. for  $C_{15}H_{10}Cl_2N_2O$ : C, 59.04; H, 3.30; N, 9.18. Found: C, 58.89; H, 3.53; N, 8.96.

## 7-Chloro-3-cyclohexylquinazolin-4(3*H*)-one (**3f**).

This compound was obtained as solid with mp 120-121 °C;  $^1\text{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta = 8.24$  (d, J = 8.8 Hz, 1H, H5), 8.13 (s, 1H, H2), 7.70 (s, 1H, H8), 7.44 (d, J = 8.8 Hz, 1H, H6), 4.79 (m, 1H, NCH), 2.02-1.94 (m, 6H, 3xCH<sub>2</sub>), 1.65-1.51 (m, 4H, 2xCH<sub>2</sub>); IR (KBr): 2933, 2854, 1678, 1600, 1461, 1402, 1352, 1318, 1270, 1162, 1130, 1068, 910, 891, 872, 827, 778 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{14}H_{15}CIN_2O$ : C, 64.00; H, 5.75; N, 10.66. Found: C, 64.25; H, 5.63; N, 10.58.

## 3-(4'-Iodophenyl)quinazolin-4(3*H*)-one (**3g**).

This compound was obtained as solid with mp 194-195 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta = 8.36$  (d, J = 7.2 Hz, 1H, H5), 8.09 (s, 1H, H2), 7.89 (d, J = 8.4 Hz, 2H), 7.84-7.77 (m, 2H), 7.59-7.55 (m, 1H), 7.18 (d, J = 8.4 Hz, 2H);IR (KBr): 3041, 1674, 1607, 1487, 1469, 1397, 1322, 1295, 1185, 1109, 1006, 911, 849, 812, 769, 690 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{14}H_9IN_2O$ : C, 48.30; H, 2.61; N, 8.05. Found: C, 48.47; H, 2.76; N, 7.81.

#### 7-Chloro-3-(4'-iodophenyl)quinazolin-4(3*H*)-one (**3h**).

This compound was obtained as solid with mp 239-240 °C;  $^1\text{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta = 8.30$  (d, J = 8.4 Hz, 1H, H5), 8.11 (s, 1H, H2), 7.92 (d, J = 8.4 Hz, 2H), 7.79 (s, 1H, H8), 7.55 (d, J = 8.4 Hz, 1H, H6), 7.20 (d, J = 8.4 Hz, 2H); IR (KBr): 3049, 1688, 1605, 1483, 1464, 1428, 1396, 1307, 1294, 1085, 1059, 1008, 915, 895, 835, 815, 784, 693 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{14}H_8CIIN_2O$ : C, 43.95; H, 2.11; N, 7.32. Found: C, 44.19; H, 1.93; N, 7.58.

## 3-(2'-Chlorophenyl)quinazolin-4(3H)-one (3i).

This compound was obtained as solid with mp 221-222 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 8.38 (d, J = 8.0 Hz, 1H, H5), 7.98 (s, 1H, H2), 7.86-7.79 (m, 2H), 7.64-7.43 (m, 5H); IR (KBr): 3062, 1681, 1605, 1470, 1394, 1373, 1301, 1266, 1186, 1114, 1083, 1021, 917, 867, 776, 755, 726, 696 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{14}H_9ClN_2O$ : C, 65.51; H, 3.53; N, 10.91. Found: C, 65.72; H, 3.28; N, 11.15.

# 7-Chloro-3-(2'-chlorophenyl)quinazolin-4(3*H*)-one (3**j**).

This compound was obtained as solid with mp 188-189 °C;  ${}^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 8.30 (d, J = 8.8 Hz, 1H, H5), 7.99 (s, 1H, H2), 7.81 (s, 1H, H8), 7.63 (d, J = 8.8 Hz, 1H, H6), 7.55-7.45 (m, 4H); IR (KBr): 3032, 1682, 1646, 1604, 1528, 1466, 1440, 1390, 1302, 1266, 1087, 900, 836, 761, 692 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{14}H_8Cl_2N_2O$ : C, 57.76; H, 2.77; N, 9.62. Found: C, 57.94; H, 2.47; N, 9.76.

## General Procedure for the Synthesis of 3,4-Dihydroquinazolines.

A solution of o-nitrobenzyl amine 4 (5 mmol) and triethyl orthoformate (10 mmol) in anhydrous THF (10 mL) was added carefully at room temperature to a suspension of low-valent titanium reagent (20 mmol) prepared as mentioned above. When the reaction was completed (at refluxing under  $N_2$ ), most of the

solvent was removed *in vacuo*. The residue was poured into 10% HCl (100 mL), and extracted with CHCl<sub>3</sub> (3x50 mL). The combined organic layers were washed with water (3x50 mL), dried (Na<sub>2</sub>SO<sub>4</sub>), and the solvent was removed *in vacuo* to give the crude product. The crude product was recrystallized from ethanol to give the pure product.

## 3-(4'-Chlorophenyl)-3,4-dihydroquinazoline (5a).

This compound was obtained as solid with mp 140-141 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.56 (s, 1H, H2), 7.40 (d, J = 8.8 Hz, 2H), 7.29-7.21(m, 2H, H6, H7), 7.15 (d, J = 7.6 Hz, 1H, H8), 7.10 (d, J = 8.8 Hz, 2H), 7.01 (d, J = 6.4 Hz, 1H, H5), 4.92 (s, 2H, H4); IR (KBr): 3067, 2810, 1600, 1565, 1550, 1470, 1290, 1230, 1160, 920, 800, 735 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{14}H_{11}ClN_2$ : C, 69.29; H, 4.57; N, 11.54. Found: C, 69.53; H, 4.26; N, 11.72.

## 3-(4'-Bromophenyl)-3,4-dihydroquinazoline (**5b**).

This compound was obtained as solid with mp 144-145 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.58 (s, 1H, H2), 7.55 (d, J = 8.8 Hz, 2H), 7.27-7.13 (m, 3H, H6, H7, H8), 7.05 (d, J = 8.8 Hz, 2H), 7.01 (d, J = 7.2 Hz, 1H, H5), 4.92 (s, 2H, H4); IR (KBr): 3055, 1610, 1560, 1550, 1490, 1370, 1300, 1235, 1165, 1080, 1000, 920, 800, 750 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{14}H_{11}BrN_2$ : C, 58.56; H, 3.86; N, 9.76. Found: C, 58.75; H, 3.68; N, 9.54.

## 6-Chloro-3-phenyl-3,4-dihydroquinazoline (5c).

This compound was obtained as solid with mp 126-127 °C;  $^1\text{HNMR}$  (400 MHz,  $\text{CDCl}_3$ ):  $\delta = 7.59$  (s, 1H, H2), 7.47-7.43 (m, 2H), 7.24-7.14 (m, 5H), 6.99 (s, 1H, H5), 4.93 (s, 2H, H4); IR (KBr): 3060, 1593, 1580, 1550, 1500, 1367, 1283, 1217, 810, 740 cm<sup>-1</sup>.

*Anal.* Calcd. for C<sub>14</sub>H<sub>11</sub>ClN<sub>2</sub>: C, 69.29; H, 4.57; N, 11.54. Found: C, 69.45; H, 4.38; N, 11.39.

## 6-Chloro-3-(4'-methylphenyl)-3,4-dihydroquinazoline (5d).

This compound was obtained as solid with mp 181-182 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.55 (s, 1H, H2), 7.24 (d, J = 8.0 Hz, 2H), 7.20 (d, J = 8.0 Hz, 1H, H7), 7.13 (d, J = 8.0 Hz, 1H, H8), 7.06 (d, J = 8.0 Hz, 2H), 6.98 (s, 1H, H5), 4.91 (s, 2H, H4), 2.37 (s, 3H, CH<sub>3</sub>); IR (KBr): 3045, 1593, 1550, 1490, 1380, 1283, 1250, 1207, 907, 817, 800 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{15}H_{13}ClN_2$ : C, 70.18; H, 5.10; N, 10.91. Found: C, 70.41; H, 4.86; N, 11.07.

## 6-Chloro-3-(4'-chlorophenyl)-3,4-dihydroquinazoline (5e).

This compound was obtained as solid with mp 187-188 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.53 (s, 1H, H2), 7.41 (d, J = 8.8 Hz, 2H), 7.22 (d, J = 8.8 Hz, 1H, H7), 7.14 (d, J = 8.8 Hz, 1H, H8), 7.10 (d, J = 8.8 Hz, 2H), 7.00 (s, 1H, H5), 4.88 (s, 2H, H4); IR (KBr): 3060, 1600, 1565, 1550, 1480, 1380, 1280, 1230, 1160, 1100, 930, 870, 810 cm<sup>-1</sup>.

Anal. Calcd. for  $C_{14}H_{10}Cl_2N_2$ : C, 60.67; H, 3.64; N, 10.11. Found: C, 60.83; H, 3.54; N, 10.36.

# 6-Chloro-3-(4'-bromophenyl)-3,4-dihydroquinazoline (5f).

This compound was obtained as solid with mp 187-188 °C;  ${}^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.58 (s, 1H, H2), 7.56 (d, J = 8.8 Hz, 2H), 7.23 (d, J = 8.4 Hz, 1H, H7), 7.17 (d, J = 8.4 Hz, 1H, H8), 7.05 (d, J = 8.8 Hz, 2H), 7.01 (s, 1H, H5), 4.90 (s, 2H, H4);

IR (KBr): 3060, 1600, 1560, 1550, 1490, 1280, 1230, 1170, 1090, 925, 820 cm $^{-1}$ .

*Anal.* Calcd. for  $C_{14}H_{10}B_rClN_2$ : C, 52.29; H, 3.13; N, 8.71. Found: C, 52.53; H, 2.94; N, 8.97.

General Procedure for the Synthesis of Imidazo[1,2-c]quinazolines.

A solution of 2-(o-nitrophenyl)imidazole **6** (2 mmol) and ortho-ester **7** (4 mmol) in anhydrous THF (10 mL) was added carefully at room temperature to a suspension of low-valent titanium reagent (10 mmol) prepared as mentioned above. When the reaction was completed (at room temperature under  $N_2$ ), most of the solvent was removed *in vacuo*. The residue was poured into 10% HCl (50 mL), and extracted with CHCl $_3$  (3x50 mL). The combined organic layers were washed with water (3x50 mL), dried (Na $_2$ SO $_4$ ), and the solvent was removed *in vacuo* to give the crude product. The crude product was purified by column chromatography on silica gel (200-300 mesh) using petroleum ether (b. p. 60-90 °C) – acetone (5:1) as eluent.

## 2,3-Diphenylimidazo[1,2-c]quinazoline (8a).

This compound was obtained as solid with mp 193-195 °C;  ${}^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 8.72 (s, 1H), 7.97 (d, J = 7.6 Hz, 1H), 7.86 (d, J = 8.0 Hz, 1H), 7.4-7.70 (m, 4H), 7.59-7.50 (m, 5H), 7.41-7.30 (m, 3H); IR (KBr): 3058, 1603, 1473, 1379, 1353, 1310, 1262, 1235, 894, 778, 746, 704, 693 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{22}H_{15}N_3$ : C, 82.22; H, 4.70; N, 13.08. Found: C, 82.41; H, 4.46; N, 13.16.

## 9-Chloro-2,3-diphenylimidazo[1,2-c]quinazoline (**8b**).

This compound was obtained as solid with mp 216-217 °C;  ${}^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 8.75 (s, 1H), 8.49 (s, 1H), 7.98 (d, J = 8.4 Hz, 1H), 7.81 (d, J = 8.4 Hz, 1H), 7.67-7.61 (m, 7H), 7.38-7.31 (m, 3H); IR (KBr): 3049, 1601, 1468, 1404, 1352, 1309, 1082, 1024, 901, 831, 781, 750, 700, 683 cm<sup>-1</sup>.

Anal. Calcd. for  $C_{22}H_{14}ClN_3$ : C, 74.26; H, 3.97; N, 11.81. Found: C, 74.42; H, 3.75; N, 11.69.

## 8,9-Dimethoxy-2,3-diphenylimidazo[1,2-c]quinazoline (8c).

This compound was obtained as solid with mp 206-207 °C;  $^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 8.63 (s, 1H), 7.82 (s, 1H), 7.68-7.56 (m, 7H), 7.43 (s, 1H), 7.36-7.29 (m, 3H), 4.02 (s, 3H, CH<sub>3</sub>O), 3.94 (s, 3H, CH<sub>3</sub>O); IR (KBr): 3049, 2933, 1622, 1495, 1462, 1383, 1340, 1271, 1217, 1134, 1030, 984, 883, 860, 818, 787, 744, 704 cm<sup>-1</sup>.

Anal. Calcd. for  $C_{24}H_{19}N_3O_2$ : C, 75.57; H, 5.02; N, 11.02. Found: C, 75.71; H, 4.86; N, 11.23.

## 5-Methyl-2,3-diphenylimidazo[1,2-c]quinazoline (8d).

This compound was obtained as solid with mp 188-189 °C;  $^1\mathrm{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta=8.02\text{-}7.84$  (m, 4H), 7.70-7.50 (m, 7H), 7.40-7.36 (m, 3H), 2.66 (s, 3H, CH<sub>3</sub>); IR (KBr): 3050, 1697, 1623, 1575, 1537, 1358, 1333, 1280, 1242, 1174, 823, 766, 743, 703 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{23}H_{17}N_3$ : C, 82.36; H, 5.11; N, 12.53. Found: C, 82.56; H, 4.83; N, 12.47.

5-Methyl-8,9-dimethoxy-2,3-diphenylimidazo[1,2-c]quinazoline (**8e**).

This compound was obtained as solid with mp 254-256 °C;  $^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta = 7.62$ -7.53 (m, 9H), 7.29-7.26

(m, 3H), 4.04 (s, 6H, 2xCH<sub>3</sub>O), 2.64(s, 3H, CH<sub>3</sub>); IR (KBr): 3057, 2957, 1628, 1535, 1495, 1438, 1380, 1339, 1265, 1228, 1206, 1182, 1025, 925, 784, 768, 707 cm<sup>-1</sup>.

*Anal.* Calcd. for C<sub>25</sub>H<sub>21</sub>N<sub>3</sub>O<sub>2</sub>: C, 75.93; H, 5.35; N, 10.63. Found: C, 75.89; H, 5.28; N, 10.81.

5-Methyl-9-chloro-2,3-diphenylimidazo[1,2-c]quinazoline (**8f**).

This compound was obtained as solid with mp 179-180 °C; <sup>1</sup>HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 8.66 (s, 1H), 7.76 (d, J = 8.0 Hz, 1H), 7.60-7.52 (m, 8H), 7.26-7.22 (m, 3H), 2.29 (s, 3H, CH<sub>3</sub>); IR (KBr): 3054, 1607, 1538, 1499, 1467, 1442, 1379, 1331, 1258, 1186, 1071, 1019, 920, 886, 871, 820, 778, 761, 703 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{23}H_{16}ClN_3$ : C, 74.69; H, 4.36; N, 11.36. Found: C, 74.83; H, 4.15; N, 11.29.

2,3-Di(4'-methylphenyl)imidazo[1,2-c]quinazoline (8g).

This compound was obtained as solid with mp 227-229 °C;  $^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 8.71 (s, 1H), 8.52 (d, J = 7.6 Hz, 1H), 7.95 (d, J = 8.8 Hz, 1H), 7.79-7.75 (m, 2H), 7.55 (d, J = 8.4 Hz, 2H), 7.51 (d, J = 8.0 Hz, 2H), 7.43 (d, J = 8.0 Hz, 2H), 7.16 (d, J = 8.4 Hz, 2H), 2.45 (s, 3H, CH<sub>3</sub>), 2.31 (s, 3H, CH<sub>3</sub>); IR (KBr): 3050, 1619, 1600, 1530, 1491, 1472, 1453, 1379, 1351, 1308, 1268, 1176, 894, 835, 822, 759, 733, 704 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{24}H_{19}N_3$ : C, 82.49; H, 5.48; N, 12.03. Found: C, 82.56; H, 5.34; N, 11.98.

9-Chloro-2,3-di(4'-methylphenyl)imidazo[1,2-c]quinazoline (8h).

This compound was obtained as solid with mp 237-239°C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 8.74 (s, 1H), 8.46 (s, 1H), 7.97 (d, J = 8.8 Hz, 1H), 7.80 (d, J = 8.8 Hz, 1H), 7.55 (d, J = 8.0 Hz, 2H), 7.51 (d, J = 8.0 Hz, 2H), 7.43 (d, J = 8.0 Hz, 2H), 7.17 (d, J = 8.0 Hz, 2H), 2.46 (s, 3H, CH<sub>3</sub>), 2.31 (s, 3H, CH<sub>3</sub>); IR (KBr): 3040, 1603, 1530, 1500, 1468, 1377, 1345, 1174, 1078, 900, 821, 734 cm<sup>-1</sup>.

Anal. Calcd. for  $C_{24}H_{18}ClN_3$ : C, 75.09; H, 4.73; N, 10.95. Found: C, 75.21; H, 4.66; N, 11.06.

2,3-Di(4'-bromophenyl)imidazo[1,2-c]quinazoline (8i).

This compound was obtained as solid with mp 239-240 °C;  $^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 8.81 (s, 1H), 7.80 (d, J = 8.4 Hz, 1H), 7.97 (d, J = 8.4 Hz, 1H), 7.84-7.77 (m, 5H), 7.62-7.59 (m, 5H); IR (KBr): 3050, 1613, 1530, 1493, 1471, 1396, 1374, 1351, 1301, 1261, 1175, 1121, 1009, 959, 894, 827, 732 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{22}H_{13}Br_2N_3$ : C, 55.14; H, 2.73; N, 8.77. Found: C, 55.32; H, 2.64; N, 8.54.

9-Chloro-2,3-di(4'-bromophenyl)imidazo[1,2-c]quinazoline (**8j**).

This compound was obtained as solid with mp 248-250 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 8.83 (s, 1H), 8.48 (s, 1H), 7.99 (d, J = 8.0 Hz, 1H), 7.83 (d, J = 8.4 Hz, 2H), 7.65-7.54 (m, 7H); IR (KBr): 3050, 1608, 1358, 1494, 1469, 1397, 1375, 1345, 1124, 1079, 1009, 900, 821, 734, 715 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{22}H_{12}Br_2ClN_3$ : C, 51.45; H, 2.35; N, 8.18. Found: C, 51.68; H, 2.28; N, 8.40.

General Procedure for the Synthesis of 1,2-Dihydroquinazolin-4(3H)-ones.

A solution of *o*-nitrobenzamide **1** (3 mmol) and ketone or aldehyde **9** (3 mmol) in anhydrous THF (10 mL) was added carefully at room temperature to a suspension of low-valent titanium

reagent (10 mmol) prepared as mentioned above. When the reaction was completed (at room temperature under  $N_2$ ), most of the solvent was removed *in vacuo*. The residue was poured into 10% HCl (50 mL), and extracted with CHCl $_3$  (3x50 mL). The combined organic layers were washed with water (3x50 mL), dried (Na $_2$ SO $_4$ ), and the solvent was removed *in vacuo* to give the crude product. The crude product was recrystallized from ethanol to give the pure product.

2,2-Dimethyl-3-(4'-methylphenyl)-1,2-dihydroquinazolin-4(3*H*)-one (**10a**).

This compound was obtained as solid with mp 255-256 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.95 (d, J = 8.4 Hz, 1H, H5), 7.32 (dd,  $J_1$  = 8.8 Hz,  $J_2$  = 7.2 Hz, 1H, H7), 7.23 (d, J = 8.4 Hz, 2H), 7.12 (d, J = 8.4 Hz, 2H), 6.87 (dd,  $J_1$  = 8.4 Hz,  $J_2$  = 7.2 Hz, 1H, H6), 6.67 (d, J = 8.8 Hz, 1H, H8), 2.38 (s, 3H, CH<sub>3</sub>), 1.49 (s, 6H, 2xCH<sub>3</sub>); IR (KBr): 3305, 2973, 1627, 1575, 1519, 1489, 1464, 1433, 1399, 1378, 1277, 1225, 1175, 1107, 1022, 947, 813, 784, 755,718, 697 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{17}H_{18}N_2O$ : C, 76.66; H, 6.81; N, 10.52. Found: C, 76.83; H, 6.59; N, 10.63.

2,2-Dimethyl-3-(4'-chlorophenyl)-1,2-dihydroquinazolin-4(3*H*)-one (**10b**).

This compound was obtained as solid with mp 255-256 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.94 (d, J = 7.2Hz, 1H, H5), 7.41 (d, J = 8.4 Hz, 2H), 7.35 (dd,  $J_1$  = 8.4 Hz,  $J_2$  = 8.0 Hz, 1H, H7), 7.19 (d, J = 8.4 Hz, 2H), 6.89 (dd,  $J_1$  = 8.0 Hz,  $J_2$  = 7.2 Hz, 1H, H6), 6.69 (d, J = 8.4 Hz, 1H, H8), 1.50 (s, 6H, 2xCH<sub>3</sub>); IR (KBr): 3307, 2972, 1628, 1518, 1489, 1465, 1433, 1398, 1377, 1338, 1273, 1179, 1088, 1016, 872, 818, 756, 727, 696 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{16}H_{15}ClN_2O$ : C, 67.02; H, 5.27; N, 9.77. Found: C, 67.18; H, 5.03; N, 9.83.

2,2-Dimethyl-3-(4'-bromophenyl)-1,2-dihydroquinazolin-4(3H)-one ( $\mathbf{10c}$ ).

This compound was obtained as solid with mp 264-265 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.94 (d, J = 8.0 Hz, 1H, H5), 756 (d, J = 8.0 Hz, 2H), 7.34 (dd,  $J_1$  = 8.0 Hz,  $J_2$  = 7.2 Hz, 1H, H7), 7.13 (d, J = 8.0 Hz, 2H), 6.88 (dd,  $J_1$  = 8.0 Hz,  $J_2$  = 7.2 Hz, 1H, H6), 6.68 (d, J = 8.0 Hz, 1H, H8), 1.49 (s, 6H, 2xCH<sub>3</sub>); IR (KBr): 3307, 2968, 1627, 1579, 1489, 1465, 1433, 1397, 1377, 1271, 1171, 1098, 1067, 1012, 871,756,696 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{16}H_{15}BrN_2O$ : C, 58.02; H, 4.56; N, 8.46. Found: C, 58.18; H, 4.54; N, 8.65.

7-Chloro-2,2-dimethyl-3-phenyl-1,2-dihydroquinazolin-4(3H)-one (10d).

This compound was obtained as solid with mp 270-272 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.87 (d, J = 8.4 Hz, 1H, H5), 7.55-7.22 (m, 5H), 6.83 (d, J = 8.4 Hz, 1H H6), 6.68 (s, 1H, H8), 1.49 (s, 6H, 2xCH<sub>3</sub>); IR (KBr): 3286, 2968, 1628, 1606, 1516, 1488, 1454, 1411, 1389, 1371 1279, 1224, 1178, 1077, 1032, 1002, 988, 950, 931, 898, 850, 809, 758, 733, 699 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{16}H_{15}ClN_2O$ : C, 67.02; H, 5.27; N, 9.77. Found: C, 67.25; H, 5.18; N, 9.69.

7-Chloro-2,2-dimethyl-3-(4'-methylphenyl)-1,2-dihydroquinazolin-4(3*H*)-one (**10e**).

This compound was obtained as solid with mp 278-280 °C;  $^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.87 (d, J = 8.0 Hz, 1H, H5),

7.23 (d, J = 7.2 Hz, 2H), 7.10 (d, J = 7.2 Hz, 2H), 6.82 (d, J = 8.0 Hz, 1H H6), 6.67 (s, 1H, H8), 2.38 (s, 3H, CH<sub>3</sub>), 1.49 (s, 6H, 2xCH<sub>3</sub>); IR (KBr):3300, 2971, 1633, 1510, 1484, 1457, 1413, 1366, 1284, 1258, 1175, 1106, 1077, 1025, 992, 901, 849, 811, 786, 765, 709, 693 cm<sup>-1</sup>.

*Anal.* Calcd. for C<sub>17</sub>H<sub>17</sub>ClN<sub>2</sub>O: C, 67.88; H, 5.70; N, 9.31. Found: C, 67.93; H, 5.54; N, 9.49.

7-Chloro-2,2-dimethyl-3-(4'-bromophenyl)-1,2-dihydroquinazolin-4(3*H*)-one (**10f**).

This compound was obtained as solid with mp 270-272 °C;  $^1\text{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta = 7.86$  (d, J = 8.0 Hz, 1H, H5), 7.57 (d, J = 8.0 Hz, 2H), 7.11 (d, J = 8.0 Hz, 2H), 6.84 (d, J = 8.0 Hz, 1H H6), 6.69 (s, 1H, H8), 1.49 (s, 6H, 2xCH<sub>3</sub>); IR (KBr): 3297, 2965, 1625, 1515, 1486, 1455, 1409, 1372, 1279, 1174, 1097, 1068, 1023, 1010, 903, 856, 814, 768, 715, 695 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{16}H_{14}BrClN_2O$ : C, 52.56; H, 3.86; N, 7.66. Found: C, 52.68; H, 3.74; N, 7.54.

## 2,2-Dimethyl-3-(*n*-octyl)-1,2-dihydroquinazolin-4(3*H*)-one (**10g**).

This compound was obtained as solid with mp 110-112 °C; 

<sup>1</sup>HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.90 (d, J = 7.2 Hz, 1H, H5), 7.21 (dd,  $J_1$  = 7.6 Hz,  $J_2$  = 8.0 Hz, 1H, H7), 6.72 (dd,  $J_1$  = 7.2 Hz,  $J_2$  = 7.6 Hz, 1H, H6), 6.60 (d, J = 8.0 Hz, 1H, H8), 3.48-3.38 (m, 2H, CH<sub>2</sub>N), 1.66-1.56 (m, 2H, CH<sub>2</sub>), 1.55 (s, 6H, 2xCH<sub>3</sub>), 1.40-1.23 (m, 10H, 5xCH<sub>2</sub>), 0.92-0.85 (m, 3H, CH<sub>3</sub>); IR (KBr):3281, 2925, 2851, 1621, 1517, 1487, 1465, 1434, 1401, 1367, 1352, 1326, 1282, 1191, 1148, 1029, 752, 699 cm<sup>-1</sup>.

Anal. Calcd. for  $C_{18}H_{28}N_2O$ : C, 74.96; H, 9.78; N, 9.71. Found: C, 75.03; H, 9.57; N, 9.62.

7-Chloro-2,2-dimethyl-3-(n-octyl)-1,2-dihydroquinazolin-4(3H)-one (10h).

This compound was obtained as solid with mp 110-111 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.82 (d, J = 8.4 Hz, 1H, H5), 7.21 (d, J = 8.4 Hz, 1H, H6), 6.68 (s, 1H, H8), 3.45-3.35 (m, 2H, CH<sub>2</sub>N), 1.65-1.58 (m, 2H, CH<sub>2</sub>), 1.55 (s, 6H, 2xCH<sub>3</sub>), 1.38-1.22 (m, 10H, 5xCH<sub>2</sub>), 0.92-0.85 (m, 3H, CH<sub>3</sub>); IR (KBr): 3299, 2960, 2928, 2854, 1623, 1516, 1468, 1417, 1367, 1324, 1279, 1181, 1079, 987, 847, 768, 693 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{18}H_{27}ClN_2O$ : C, 66.96; H, 8.43; N, 8.68. Found: C, 67.21; H, 8.27; N, 8.56.

# 2,2-Dimethyl-3-benzyl-1,2-dihydroquinazolin-4(3H)-one ( $\mathbf{10i}$ ).

This compound was obtained as solid with mp 200-201 °C;  $^1\mathrm{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta=7.98$  (d, J=7.2 Hz, 1H, H5), 7.30-7.23 (m, 5H), 7.22 (dd,  $J_1=7.6$  Hz,  $J_2=8.4$  Hz, 1H, H7), 6.87 (dd,  $J_1=7.2$  Hz,  $J_2=7.6$  Hz, 1H, H6), 6.64 (d, J=8.4 Hz, 1H, H8), 4.82 (s, 2H, CH2), 1.55 (s, 6H, 2xCH<sub>3</sub>); IR (KBr): 3321, 3030, 2997, 2927, 1625, 1514, 1458, 1364, 1284, 1179, 1073, 1025, 918, 858, 756, 694 cm $^{-1}$ .

*Anal.* Calcd. for  $C_{17}H_{18}N_2O$ : C, 76.66; H, 6.81; N, 10.52. Found: C, 76.59; H, 7.05; N, 10.67.

## 2-Methyl-2-ethyl-1,2-dihydroquinazolin-4(3*H*)-one (**10j**).

This compound was obtained as solid with mp 184-186 °C;  ${}^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.87 (d, J = 8.0 Hz, 1H, H5), 7.30 (dd,  $J_{1}$  = 8.0 Hz,  $J_{2}$  = 7.2 Hz, 1H, H7), 6.81 (dd,  $J_{1}$  = 8.0 Hz,  $J_{2}$  = 7.2 Hz, 1H, H8), 6.16 (br s, 1H, NH), 1.81 (q, J = 8.0 Hz, 2H, CH<sub>2</sub>), 1.50 (s, 3H, CH<sub>3</sub>), 0.99 (t, J = 8.0 Hz, 3H, CH<sub>3</sub>); IR (KBr):3279, 3178, 2974, 1643, 1609,

1512, 1489, 1430, 1395, 1331, 1275, 1182, 1153, 758 cm<sup>-1</sup>. Anal. Calcd. for  $C_{11}H_{14}N_2O:$  C, 69.45; H, 7.42; N, 14.73. Found: C, 69.58; H, 7.14; N, 14.89.

## 2,2-Diethyl-1,2-dihydroquinazolin-4(3*H*)-one (**10k**).

This compound was obtained as solid with mp 190-191 °C;  $^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.85 (d, J = 8.0 Hz, 1H, H5), 7.28 (dd,  $J_{1}$  = 8.0 Hz,  $J_{2}$  = 8.0 Hz, 1H, H7), 6.78 (dd,  $J_{1}$  = 8.0 Hz,  $J_{2}$  = 8.0 Hz, 1H, H8), 5.98 (br s, 1H, NH), 1.76 (q, J = 8.0 Hz, 4H, 2xCH<sub>2</sub>), 0.97 (t, J = 8.0 Hz, 6H, 2xCH<sub>3</sub>); IR (KBr):3320, 3175, 2974, 1646, 1607, 1510, 1489, 1463, 1429, 1395, 1329, 1274, 1150, 758 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{12}H_{16}N_2O$ : C, 70.56; H, 7.90; N, 13.71. Found: C, 70.83; H, 7.84; N, 13.62.

7-Chloro-2-methyl-2-ethyl-1,2-dihydroquinazolin-4(3*H*)-one (101).

This compound was obtained as solid with mp 173-174 °C;  ${}^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.78 (d, J = 8.0 Hz, 1H, H5), 6.96 (br s, 1H, NH), 6.75 (d, J = 8.0, 1H, H6), 6.63 (s, 1H, H8), 1.79 (q, J = 8.0 Hz, 2H, CH<sub>2</sub>), 1.50 (s, 3H, CH<sub>3</sub>), 0.98 (t, J = 8.0 Hz, 3H, CH<sub>3</sub>); IR (KBr):3304, 3191, 2974, 1642, 1608, 1510, 1480, 1454, 1419, 1320, 1276, 1156, 1079, 896, 854, 778 cm<sup>-1</sup>.

*Anal.* Calcd. for C<sub>11</sub>H<sub>13</sub>ClN<sub>2</sub>O: C, 58.80; H, 5.83; N, 12.47. Found: C, 58.92; H, 5.71; N, 12.56.

#### 7-Chloro-2,2-diethyl-1,2-dihydroquinazolin-4(3H)-one (10m).

This compound was obtained as solid with mp 164-166 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.76 (d, J = 8.0 Hz, 1H, H5), 6.71 (d, J = 8.0 Hz, 1H, H6), 6.61 (s, 1H, H8), 6.59 (br s, 1H, NH),1.75 (q, J = 8.0 Hz, 4H, 2xCH<sub>2</sub>), 0.96 (t, J = 8.0 Hz, 6H, 2xCH<sub>3</sub>); IR (KBr):3286, 3215, 2967, 1644, 1608, 1513, 1483, 1461, 1420, 1363, 1324, 1276, 1175, 1155, 1082, 986, 912, 874, 774 cm<sup>-1</sup>.

*Anal.* Calcd. for C<sub>12</sub>H<sub>15</sub>ClN<sub>2</sub>O: C, 60.38; H, 6.33; N, 11.74. Found: C, 60.51; H, 6.31; N, 11.59.

#### 2-(4'-Methylphenyl)-1,2-dihydroquinazolin-4(3H)-one (**10n**).

This compound was obtained as solid with mp 231-233 °C; <sup>1</sup>HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.94 (d, J = 7.2 Hz, 1H, H5), 7.48 (d, J = 8.0 Hz, 2H), 7.34 (dd, J<sub>1</sub> = 8.4 Hz, J<sub>2</sub>=8.4 Hz, 1H, H7), 7.25 (d, J = 8.0 Hz, 2H), 6.90 (dd, J<sub>1</sub> = 8.4 Hz, J<sub>2</sub> = 7.2 Hz, 1H, H6), 6.67 (d, J = 8.4 Hz, 1H, H8), 5.87 (s, 1H, H2), 5.78 (br s, 1H, NH), 2.40 (s, 3H, CH<sub>3</sub>); IR (KBr): 3312, 3194, 1657, 1611, 1509, 1486, 1438, 1385, 1328, 1297, 1151, 1133, 1022, 948, 909, 859, 800, 751 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{15}H_{14}N_2O$ : C, 75.61; H, 5.92; N, 11.76. Found: C, 75.82; H, 5.74; N, 11.85.

2-(3',4'-Methylenedioxylphenyl)-1,2-dihydroquinazolin-4(3*H*)-one (**10o**).

This compound was obtained as solid with mp 199-201 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.94 (d, J = 7.2 Hz, 1H, H5), 7.34 (dd,  $J_1$  = 7.6 Hz,  $J_2$  = 8.4 Hz, 1H, H7), 7.15 (s, 1H), 6.99 (d, J = 8.0 Hz, 1H), 6.91 (dd,  $J_1$  = 7.2 Hz,  $J_2$  = 7.6 Hz, 1H, H6), 6.83 (d, J = 8.0 Hz, 1H), 6.67 (d, J = 8.0 Hz, 1H, H8), 6.02 (s, 2H, OCH<sub>2</sub>O), 5.82 (s, 1H, H2), 5.80 (br s, 1H, NH); IR (KBr):3282, 3181, 1654, 1612, 1486, 1446, 1388, 1327, 1297, 1248, 1187, 1164, 1149, 1121, 1105, 1036, 930, 864, 786, 754 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{15}H_{12}N_2O_3$ : C, 67.16; H, 4.51; N, 10.44. Found: C, 67.27; H, 4.38; N, 10.49.

7-Chloro-2-(4'-methylphenyl)-1,2-dihydroquinazolin-4(3*H*)-one (**10p**).

This compound was obtained as solid with mp 242-244 °C;  ${}^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta = 7.87$  (d, J = 8.8 Hz, 1H, H5), 7.45 (d, J = 8.0 Hz, 2H), 7.25 (d, J = 8.0 Hz, 2H), 6.86 (d, J = 8.8 Hz, 1H, H6), 6.67 (s, 1H, H8), 5.87 (s, 1H, H2), 5.78 (br s,1H, NH), 2.40 (s, 3H, CH<sub>3</sub>); IR (KBr): 3294, 3181, 1654, 1606, 1510, 1471, 1432, 1370, 1297, 1172, 1133, 1079, 1014, 922, 858, 816, 748, 721, 678 cm<sup>-1</sup>.

Anal. Calcd. for  $C_{15}H_{13}CIN_2O$ : C, 66.06; H, 4.80; N, 10.27. Found: C, 66.24; H, 4.76; N, 10.19.

7-Chloro-2-(4'-methoxyphenyl)-1,2-dihydroquinazolin-4(3*H*)-one (**10q**).

This compound was obtained as solid: with mp 215-216 °C;  $^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.86 (d, J = 8.8 Hz, 1H, H5), 7.51 (d, J = 8.0 Hz, 2H), 6.96 (d, J = 8.0 Hz, 2H), 6.86 (d, J = 8.0 Hz, 1H, H6), 6.67 (s, 1H, H8), 5.86 (s, 1H, H2), 5.77 (br s, 1H, NH), 3.85 (s, 3H, CH<sub>3</sub>O); IR (KBr): 3295, 3185, 1654, 1610, 1510, 1480, 1427, 1369, 1294, 1254, 1171, 1135, 1110, 1079, 1038, 923, 863, 834, 782, 748, 680 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{15}H_{13}ClN_2O_2$ : C, 62.40; H, 4.54; N, 9.70. Found: C, 62.56; H, 4.32; N, 9.81.

## 2,2-Tetramethylene-1,2-dihydroquinazolin-4(3H)-one (12a).

This compound was obtained as solid with mp 251-253 °C;  $^1\text{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta = 7.88$  (d, J = 7.2 Hz, 1H, H5), 7.31 (dd,  $J_1 = 8.4$  Hz,  $J_2 = 7.2$  Hz, 1H, H7), 6.85 (dd,  $J_1 = 7.2$  Hz,  $J_2 = 7.2$  Hz, 1H, H6), 6.65 (d, J = 8.4 Hz, 1H, H8), 6.17 (br s, 1H, NH), 1.97-1.88 (m, 4H, 2xCH<sub>2</sub>), 1.80-1.79 (m, 4H, CH<sub>2</sub>CH<sub>2</sub>); IR (KBr): 3292, 3159, 2972, 1638, 1606, 1517, 1485, 1431, 1385, 1334, 1270, 1149, 1088, 1049, 954, 849, 803, 781, 753 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{12}H_{14}N_2O$ : C, 71.26; H, 6.98; N, 13.85. Found: C, 71.38; H, 6.71; N, 14.02.

## 2,2-Pentamethylene-1,2-dihydroquinazolin-4(3*H*)-one (**12b**).

This compound was obtained as solid with mp 224-225 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.87 (d, J = 7.6 Hz, 1H, H5), 7.30 (dd,  $J_1$  = 8.4Hz,  $J_2$  = 7.2 Hz, 1H, H7), 6.82 (dd,  $J_1$  = 7.6 Hz  $J_2$  = 7.2 Hz, 1H, H6), 6.65 (d, J = 8.4 Hz, 1H, H8), 6.19 (br s, 1H, NH), 1.84-1.80 (m, 4H, 2xCH<sub>2</sub>), 1.60-1.53 (m, 4H, 2xCH<sub>2</sub>), 1.48-1.47 (m, 2H, CH<sub>2</sub>); IR (KBr): 3367, 3170, 2923, 1651, 1612, 1507, 1484, 1417, 1382, 1269, 1210, 1178, 1145, 1093, 1040, 1004, 951, 914, 855, 802, 760 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{13}H_{16}N_2O$ : C, 72.19; H, 7.46; N, 12.95. Found: C, 72.32; H, 7.28; N, 13.15.

7-Chloro-2,2-tetramethylene-1,2-dihydroquinazolin-4(3*H*)-one (12c).

This compound was obtained as solid with mp 223-225 °C;  $^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.80 (d, J = 8.8 Hz, 1H, H5), 6.80 (d, J = 8.8 Hz, 1H, H6), 6.65 (s, 1H, H8), 6.23 (br s, 1H, NH), 1.96-1.81 (m, 4H, 2\_CH<sub>2</sub>), 1.80-1.62 (m, 4H, 2xCH<sub>2</sub>); IR (KBr): 3260, 3189, 1650, 1608, 1519, 1480, 1421, 1361, 1318, 1277, 1154, 1078, 1044, 936, 899, 855, 768 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{12}H_{13}CIN_2O$ : C, 60.98; H, 5.54; N, 11.84. Found: C, 61.02; H, 5.36; N, 11.97.

7-Chloro-2,2-pentamethylene-1,2-dihydroquinazolin-4(3*H*)-one (12d).

This compound was obtained as solid with mp 221-222 °C;  $^1\text{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.79 (d, J = 8.8 Hz, 1H, H5), 6.77 (d, J = 8.8 Hz, 1H, H6), 6.66 (s, 1H, H8), 6.17 (br s, 1H, NH), 1.83-1.78 (m, 4H, 2xCH<sub>2</sub>), 1.67-1.53 (m, 4H, 2xCH<sub>2</sub>), 1.49-1.45 (m, 2H, CH<sub>2</sub>); IR (KBr): 3362, 3249, 1699, 1600, 1576, 1507, 1464, 1336, 1152, 1044, 890, 751 cm<sup>-1</sup>.

*Anal.* Calcd. for C<sub>13</sub>H<sub>15</sub>ClN<sub>2</sub>O: C, 62.28; H, 6.03; N, 11.17. Found: C, 62.43; H, 5.96; N, 11.33.

General Procedure for the Synthesis of 5,6-Dihydroimidazo[1,2-c]-quinazolines.

A solution of 2-(o-nitrophenyl)imidazole **6** (3 mmol) and ketone or aldehyde **9** (3 mmol) in anhydrous THF (10 mL) was added carefully at room temperature to a suspension of low-valent titanium reagent (10 mmol) prepared as mentioned above. When the reaction was completed (at room temperature under N<sub>2</sub>), most of the solvent was removed *in vacuo*. The residue was poured into 10% HCl (50 mL), and extracted with CHCl<sub>3</sub> (3\_50 mL). The combined organic layers were washed with water (3x50 mL), dried (Na<sub>2</sub>SO<sub>4</sub>), and the solvent was removed *in vacuo* to give the crude product. The crude product was purified by column chromatography on silica gel (200-300 mesh) using petroleum ether (b.p. 60-90 °C) – acetone (5:1) as eluent.

5,5-Dimethyl-2,3-diphenyl-5,6-dihydroimidazo[1,2-*c*]quinazoline (**13a**).

This compound was obtained as solid with mp 240-241 °C;  $^1\text{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.84 (d, J = 7.2 Hz, 1H), 7.51-7.54 (m, 5H), 7.36 (d, J = 7.2 Hz, 2H), 7.08-7.19 (m, 4H), 6.83 (d, J = 7.2 Hz, 1H), 6.79 (d, J = 8.8 Hz, 1H), 6.52 (br s, 1H, NH), 1.36 (s, 6H, 2xCH<sub>3</sub>); IR (KBr): 3240, 3012, 2979, 1614, 1512, 1479, 1444, 1367, 1275, 1211, 1161, 1072, 964, 916, 791, 772, 752, 698 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{24}H_{21}N_3$ : C, 82.02; H, 6.02; N, 11.96. Found: C, 82.25; H, 5.89; N, 12.10.

5,5-Dimethyl-9-chloro-2,3-diphenyl-5,6-dihydroimidazo[1,2-*c*]-quinazoline (**13b**).

This compound was obtained as solid with mp 228-229 °C;  $^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.77 (s, 1H), 7.58-7.48 (m, 5H), 7.36 (d, J = 7.6 Hz, 2H), 7.21-7.10 (m, 4H), 6.80 (d, J = 8.4 Hz, 1H), 6.76 (br s, 1H, NH), 1.35 (s, 6H, 2xCH<sub>3</sub>); IR (KBr): 3247, 3087, 2979, 1603, 1557, 1525, 1508, 1477, 1444, 1389, 1368, 1309, 1226, 1161, 814, 774, 697 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{24}H_{20}ClN_3$ : C, 74.70; H, 5.22; N, 10.89. Found: C, 74.82; H, 5.06; N, 10.96.

5,5-Dimethyl-8,9-methylenedioxy-2,3-diphenyl-5,6-dihydroimidazo[1,2-c]quinazoline (13c).

This compound was obtained as solid with mp 239-240 °C;  $^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.48-7.53 (m, 5H), 7.33 (d, J = 7.2 Hz, 2H), 7.29 (s, 1H), 7.16-7.06 (m, 3H), 6.41 (s, 1H), 6.32 (br s, 1H, NH), 5.97 (s, 2H, CH<sub>2</sub>), 1.31 (s, 6H, 2xCH<sub>3</sub>). IR (KBr): 3230, 2976, 1626, 1502, 1469, 1354, 1265, 1198, 1148, 1039, 941, 864, 831, 781, 700 cm<sup>-1</sup>.

Anal. Calcd. for  $C_{25}H_{21}N_3O_2$ : C, 75.93; H, 5.35; N, 10.63. Found: C, 76.03; H, 5.28; N, 10.55.

5,5-Dimethyl-2,3-di(4'-methylphenyl)-5,6-dihydroimidazo[1,2-*c*]-quinazoline (**13d**).

This compound was obtained as solid with mp 222-224 °C;  $^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.81 (d, J = 7.6 Hz, 1H), 7.40-

7.14 (m, 7H), 6.97 (d, J = 8.4 Hz, 2H), 6.96-6.77 (m, 2H), 6.52 (br s, 1H, NH), 2.42 (s, 3H, CH<sub>3</sub>), 2.21 (s, 3H, CH<sub>3</sub>), 1.34 (s, 6H, 2xCH<sub>3</sub>); IR (KBr): 3224, 3012, 1613, 1513, 1500, 1490, 1388, 1367, 1311, 1278, 1232, 1211, 1167, 1107, 1022, 968, 824, 754, 705 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{26}H_{25}N_3$ : C, 82.29; H, 6.64; N, 11.07. Found: C, 82.17; H, 6.75; N, 11.23.

5,5-Dimethyl-2,3-di(4'-methylphenyl)-9-chloro-5,6-dihydroimidazo[1,2-*c*]quinazoline (**13e**).

This compound was obtained as solid with mp 215-217 °C;  $^1\text{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.73 (d, J = 8.8 Hz, 1H), 7.32-7.24 (m, 5H), 7.19 (d, J = 7.6 Hz, 1H), 7.04 (d, J = 8.4 Hz, 1H), 6.98 (d, J = 8.0 Hz, 1H), 6.88 (d, J = 8.8 Hz, 1H), 6.79 (d, J = 8.4 Hz, 1H), 6.75 (br s, 1H, NH), 2.42 (s, 3H, CH<sub>3</sub>), 2.21(s, 3H, CH<sub>3</sub>), 1.35 (s, 6H, 2xCH<sub>3</sub>); IR (KBr): 3212, 1613, 1508, 1490, 1389, 1369, 1306, 1258, 1229, 1205, 1163, 1108, 1083, 1019, 971, 813, 767, 738, 678 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{26}H_{24}ClN_3$ : C, 75.44; H, 5.84; N, 10.15. Found: C, 75.59; H, 5.66; N, 10.37.

5,5-Dimethyl-2,3-di(4'-bromophenyl)-5,6-dihydroimidazo[1,2-c]quinazoline (13f).

This compound was obtained as solid with mp 262-264 °C;  $^1\text{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.83 (d, J = 7.6 Hz, 1H), 7.74 (d, J = 8.4 Hz, 1H), 7.50-7.40 (m, 6H), 7.27 (d, J = 8.4 Hz, 1H), 7.17 (d, J = 7.2 Hz, 1H), 6.83 (d, J = 7.6 Hz, 1H), 6.79 (d, J = 7.2 Hz, 1H), 6.61 (br s, 1H, NH), 1.39 (s, 6H, 2xCH<sub>3</sub>); IR (KBr): 3354, 3050, 1610, 1532, 1496, 1401, 1328, 1152, 1098, 1079, 1009, 969, 810, 748, 717 cm<sup>-1</sup>.

Anal. Calcd. for  $C_{24}H_{19}Br_2N_3$ : C, 56.61; H, 3.76; N, 8.25. Found: C, 56.83; H, 3.51; N, 8.09.

5,5-Dimethyl-2,3-di(4'-bromophenyl)-9-chloro-5,6-dihydroimidazo[1,2-*c*]quinazoline (**13g**).

This compound was obtained as solid with mp 286-288 °C;  $^1$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.74 (d, J = 8.4 H, 2H), 7.50-7.45 (m, 3H), 7.42 (d, J = 8.8 Hz, 2H), 7.28 (d, J = 8.8 Hz, 2H), 7.22 (d, J = 8.4 Hz, 1H), 6.83 (br s, 1H, NH), 6.81 (d, J = 8.4 Hz, 1H), 1.41 (s, 6H, 2xCH<sub>3</sub>); IR (KBr): 3390, 3012, 2973, 1610, 1592, 1536, 1491, 1387, 1307, 1195, 1167, 1099, 1070, 1009, 829 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{24}H_{18}Br_2ClN_3$ : C, 53.02; H, 3.34; N, 7.73. Found: C, 53.26; H, 3.15; N, 7.96.

5,5-Dimethyl-2,3-di(4'-methoxyphenyl)-5,6-dihydroimidazo[1,2-c]quinazoline (13h).

This compound was obtained as solid with mp 187-189 °C;  $^1\text{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.82 (d, J = 7.2 Hz, 1H), 7.44 (d, J = 8.8 Hz, 2H), 7.38 (d, J = 8.4 Hz, 1H), 7.31 (d, J = 8.0 Hz, 1H), 7.14 (t, J = 7.2 Hz, 1H), 7.08-7.05 (m, 2H), 6.82 (d, J = 7.2 Hz, 1H), 6.77 (d, J = 8.4 Hz, 2H), 6.58 (t, J = 7.2 Hz, 1H), 6.53 (br s, 1H, NH), 3.84 (s, 3H, CH<sub>3</sub>O), 3.69 (s, 3H, CH<sub>3</sub>O), 1.35 (s, 6H, 2xCH<sub>3</sub>); IR (KBr): 3305, 1613, 1540, 1489, 1384, 1363, 1287, 1176, 1106, 1030, 966, 837, 742 cm<sup>-1</sup>.

Anal. Calcd. for  $C_{26}H_{25}N_3O_2$ : C, 75.89; H, 6.12; N, 10.21. Found: C, 75.94; H, 5.96; N, 10.37.

5,5-Dimethyl-2,3-di(4'-methoxyphenyl)-5,6-dihydroimidazo[1,2-c]quinazoline (13i).

This compound was obtained as solid with mp 196-197 °C;  $^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  =7 .74 (s, 1H), 7.39 (d, J = 8.8 Hz,

2H), 7.31 (d, J = 8.8 Hz, 2H), 7.17 (d, J = 8.4 Hz, 1H), 7.07 (d, J = 8.8 Hz, 2H), 6.80-6.75 (m, 4H), 6.74 (br s, 1H, NH), 3.84 (s, 3H, CH3), 3.69 (s, 3H, CH3), 1.35 (s, 6H,  $2xCH_3$ ); IR (KBr): 3389, 1611, 1530, 1493, 1385, 1367, 1238, 1107, 1029, 966, 879, 841, 803, 774, 745, 713, 681 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{26}H_{24}ClN_3O_2$ : C, 70.03; H, 5.42; N, 9.42. Found: C, 70.18; H, 5.27; N, 9.53.

5,5-Dimethyl-2,3-di(4'-fluorophenyl)-5,6-dihydroimidazo[1,2-c]quinazoline (13**j**).

This compound was obtained as solid with mp 261-262 °C;  $^1\text{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta = 7.83$  (d, J = 7.6 Hz, 1H), 7.57 (dd, J<sub>1</sub> = 8.8 Hz, J<sub>2</sub> = 5.6 Hz, 2H), 7.39-7.34 (m, 4H), 7.20-7.15 (m, 1H), 7.04 (dd, J<sub>1</sub> = 8.8 Hz, J<sub>2</sub> = 5.6 Hz, 2H), 6.83 (d, J = 7.6 Hz, 1H), 6.79 (d, J = 8.4 Hz, 1H), 6.58 (br s, 1H, NH), 1.36 (s, 6H, 2xCH<sub>3</sub>). IR (KBr): 3416, 2987, 1619, 1595, 1538, 1485, 1385, 1364, 1321, 1279, 1214, 1159, 855, 843, 818, 748, 703 cm<sup>-1</sup>.

Anal. Calcd. for  $C_{24}H_{19}F_{2}N_{3}$ : C, 74.40; H, 4.94; N, 10.85. Found: C, 74.37; H, 4.89; N, 11.03.

5-(4'-Methylphenyl)-2,3-diphenyl-5,6-dihydroimidazo[1,2-<math>c]-quinazoline (13k).

This compound was obtained as solid with mp 239-240 °C;  $^1\mathrm{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta=7.52\text{-}7.51$  (m, 5H), 7.40-7.38 (m, 2H), 7.33-7.31 (m, 5H), 7.21-7.12 (m, 4H), 6.98-6.78 (m, 1H), 6.75-6.77 (m, 1H), 4.47 (s, 1H), 2.42 (s, 3H, CH<sub>3</sub>); IR (KBr): 3367, 3055, 2920, 1609, 1584, 1535, 1517, 1505, 1477, 1441, 1334, 1306, 1283, 1067, 817, 765, 739, 714, 695 cm $^{-1}$ .

*Anal.* Calcd. for  $C_{29}H_{23}N_3$ : C, 84.23; H, 5.61; N, 10.16. Found: C, 84.46; H, 5.38; N, 10.24.

5-(3',4'-Dimethoxyphenyl)-2,3-diphenyl-5,6-dihydroimidazo[1,2-c]quinazoline (13l).

This compound was obtained as solid with mp 237-239 °C;  $^{1}$ HNMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.51-7.37 (m, 11H), 7.17-7.14 (m, 3H), 7.02 (s, 1H), 6.87 (d, J = 8.0 Hz, 1H), 6.70 (d, J = 8.0 Hz, 1H), 4.44 (s, 1H),3.91 (s, 3H, CH<sub>3</sub>O), 3.80 (s, 3H, CH<sub>3</sub>O); IR (KBr): 3241, 3066, 2982, 1611, 1512, 1476, 1443, 1386, 1371, 1312, 1277, 1229, 1211, 1164, 1029, 967, 770, 752, 722, 700 cm<sup>-1</sup>.

Anal. Calcd. for  $C_{30}H_{25}N_3O_2$ : C, 78.41; H, 5.48; N, 9.14. Found: C, 78.57; H, 5.23; N, 9.29.

5-(4'-Methylphenyl)-2,3-diphenyl-9-chloro-5,6-dihydroimidazo[1,2-<math>c]quinazoline (13m).

This compound was obtained as solid with mp 217-218 °C;  $^1\text{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta$  = 7.55-7.42 (m, 6H), 7.25-7.00 (m, 7H), 6.89 (d, J = 8.0 Hz, 1H), 6.81 (d, J = 8.4 Hz, 1H), 6.39 (d, J = 7.2 Hz, 2H), 4.29 (s, 1H), 2.18 (s, 3H, CH<sub>3</sub>); IR (KBr): 3265, 3066, 2951, 1608, 1529, 1496, 1443, 1118, 1065, 1015, 887, 815, 775, 698 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{29}H_{22}ClN_3$ : C, 77.76; H, 4.95; N, 9.38. Found: C, 77.85; H, 4.81; N, 9.25.

5-(4'-Methoxylphenyl)-2,3-diphenyl-9-chloro-5,6-dihydroimidazo[1,2-*c*]quinazoline (**13n**).

This compound was obtained as solid with mp 206-207 °C;  $^1\text{HNMR}$  (400 MHz, CDCl<sub>3</sub>):  $\delta = 7.58\text{-}7.49$  (m, 6H), 7.41-7.32 (m, 7H), 7.08 (d, J = 8.4 Hz, 2H), 6.73 (d, J = 8.0 Hz, 2H), 4.43 (s, 1H), 3.84 (s, 3H, CH<sub>3</sub>O); IR (KBr): 3455, 3057, 2925, 1612, 1500, 1442, 1323, 1250, 1155, 1072, 1028, 971, 916, 860, 813, 769, 697 cm<sup>-1</sup>.

*Anal.* Calcd. for  $C_{29}H_{22}CIN_3O$ : C, 75.07; H, 4.78; N, 9.06. Found: C, 75.26; H, 4.59; N, 9.13.

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