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Recyclable $[Ce(L\text{-}Pro)_2]_2$ (Oxa) used as Heterogeneous Catalyst: One-Pot Synthesis of 2,3-Dihydroquinazolin-4(1*H*)-ones in Ethanol

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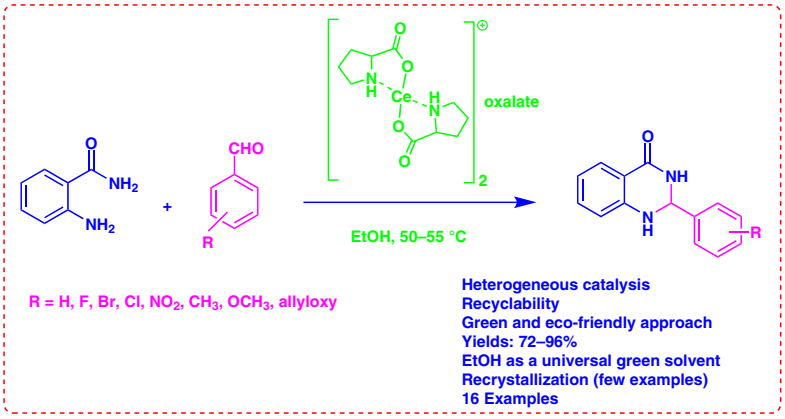

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Abstract $[Ce(L\text{-}Pro)_2]_2$ (Oxa) was used as a recyclable heterogeneous catalyst under mild conditions for the preparation of 2-aryl-2,3-dihydroquinazolin-4(1*H*)-one derivatives. The one-pot protocol proceeds in ethanol using anthranilamide with several aldehydes at 50–55 °C. The catalyst can be recycled and reused three cycles without significant loss of catalytic activity.

Key words heterogeneous catalysis, anthranilamide, benzaldehydes, hybrid catalysts

Heterocyclic compounds are vital in daily life, and they play an essential role in the metabolism of living cells. These compounds contain one or more hetero atoms in their structure, and they are biosynthesized by plants and animals.[1] Moreover, numerous natural products are used as antibiotics such as penicillins and cephalosporin, and some alkaloids (morphine, vinblastine, and reserpine) comprise a heterocyclic moiety.[2] A large number of heterocyclic compounds show a range of applications in pharmaceuticals, agrochemicals, and veterinary products.[3] Some of the heterocyclic compounds can be used as sanitizers, antioxidants, developers, copolymers, corrosion inhibitors and dyestuff.[4] DNA and RNA compounds also contain heterocyclic bases such as pyrimidines and purines.[5]

We have been interested in the synthesis of heterocyclic compounds such as 2-aryl-2,3-dihydroquinazolin-4(1*H*)-ones, which belong to the family of six-membered heterocycles. In particular, 2,3-dihydroquinazolin-4(1*H*)-one derivatives have proven to provide a useful framework for bioactive compounds, as shown in Figure 1. These analogues exhibit significant biological properties that can be effectively utilized as antitumor, anticancer, herbicidal, diuretic compounds, and in plant growth regulation.[6] Moreover, these compounds are oxidized to the quinazolin-4(3*H*)-one moiety, which constitutes an important core structure in many natural products.[7] Very recently, our group has been developing a synthesis of 2-aryl-2,3-dihydroquinazolin-4(1*H*)-ones by using a one-pot approach mediated by β-cyclodextrin under aqueous conditions.[8] In view of the importance of these heterocycles, several methods were reported for the synthesis of 2-aryl-2,3-dihydroquinazolin-4(1*H*)-one derivatives;[9] approaches include the use of silica sulfuric acid,[10] gallium(III)triflate,[11] montmorillonite K-10,[12] Amberlyst-15,[13] $KAl(SO_4)_2{\cdot}12H_2O$,[14] $Al(H_2PO_4)_3$,[15] zinc(II) perfluorooctanoate $[Zn(PFO)_2]$,[16] MCM-41-SO_3H,[17] 1-butyl-3-methylimidazolium tetrafluoroborate ($[bmim]BF_4$),[18] and molecular iodine.[19] However, these methods are associated with some limitations such as hazardous organic solvents, expensive catalysts, low yields, long reaction time, elevated temperature, non-recyclability, toxic metals, stoichiometric loadings, or low conversions.

Therefore, it is essential to establish a greener method for the preparation of 2,3-dihydroquinazolin-4(1*H*)-ones. Fortunately, we have overcome all the limitations noted

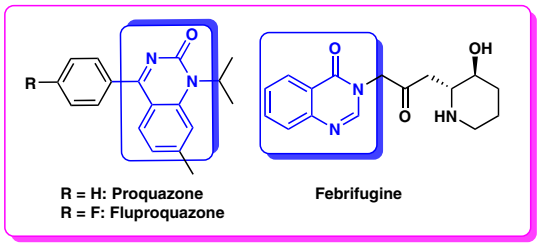
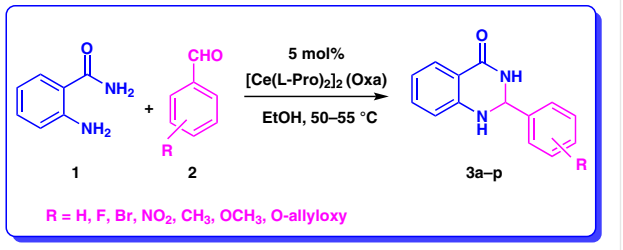


Figure 1 Bioactive scaffolds featuring a quinazolin-4(1*H*)-one moiety

above and, herein, we report a novel an efficient green approach to 2-aryl-2,3-dihydroquinazolin-4(1*H*)-ones through a one-pot protocol using anthranilamide, and several aldehydes in the presence of $[Ce(L\text{-}Pro)_2]_2$ (Oxa) as heterogeneous catalyst, and EtOH as a green solvent at 50–55 °C (Scheme 1).

We analyzed $[Ce(L\text{-}Pro)_2]_2$ (Oxa) as an efficient and accessible catalyst for the preparation of 2-aryl-2,3-dihydroquinazolin-4(1*H*)-one derivatives under mild conditions. The catalyst comprises mild Lewis acid properties,[20] is chemically stable, and does not dissociate at high temperatures.

At the beginning of the study, we conducted a model reaction between anthranilamide and benzaldehyde with neither catalyst nor solvent, at room temperature, and found that the corresponding product was formed in low yield after several hours of reaction. Hence, we conducted the reaction in the presence of both $[Ce(L\text{-}Pro)_2]_2$ (Oxa) as catalyst and solvent by using the same starting materials, and the reaction was found to proceed sluggishly at room temperature with the desired product being formed in around 62% in 6 hours. To improve the reaction yield, the reaction temperature was increased from r.t. to 50–55 °C with the same starting materials, and $[Ce(L\text{-}Pro)_2]_2$ (Oxa) was used as catalyst in EtOH as green solvent. Under these conditions we obtained the desired product in 89% yield within 4–4.5 hours. In addition, we carried out the present protocol using different catalyst loadings (1.0, 2.5, 5 mol% etc). Thus, 5 mol% catalyst loading was found to form 2-phenyl-2,3-dihydroquinazolin-4(1*H*)-one with optimal conversion. The yield was unaffected when the catalyst loading was increased to 20 mol%.



Scheme 1 Synthesis of 2-aryl-2,3-dihydroquinazolin-4(1*H*)-ones

The reaction was then performed using several solvents including CH_2Cl_2, $CHCl_3$, tetrahydrofuran (THF), H_2O and EtOH; we observed that EtOH was a suitable solvent for the preparation of 2-phenyl-2,3-dihydroquinazolin-4(1*H*)-one (**3k**) as shown in Table 1. Subsequent reactions were carried out using similar reaction conditions.

Starting benzaldehydes possessing either electron-donating groups such as -Me, -OMe, -OH, and -allyloxy, or electron withdrawing groups such as $-NO_2$, -F, -Cl, and -Br gave excellent yields (Table 2).

Upon completion of the reaction, the reaction mass was cooled to r.t., and the catalyst was filtered off and cleaned with diethyl ether before drying at 80 °C for 2 h. The recovered catalyst was reused with the same substrates, and the yields and the catalytic activity were monitored; the results are shown in Figure 2. In all these reactions the catalyst could be recycled and reused with little loss of its catalytic activity (Figure 2).

Table 1 Screening of Solvents for the Synthesis of 2-Phenyl-2,3-dihydroquinazolin-4(1*H*)-one (**3k**; Table 2, entry 11)[a]

Entry	Solvent	*T* (°C)	Yield (%)[b]
1	–	–	12[c]
2	CH_2Cl_2	r.t.	54
3	$CHCl_3$	r.t.	56
4	ACN	50	63
5	H_2O	70	67
6	EtOH	r.t.	70
7	EtOH	50–55	89

[a] Reaction conditions: Anthranilamide (1.0 mmol), benzaldehyde (1.0 mmol), $[Ce(L\text{-}Pro)_2]_2$ (Oxa) (5 mol%).
[b] Isolated yield.
[c] In the absence of catalyst and solvent.

Table 2 Synthesis of 2-Aryl-2,3-dihydroquinazolin-4(1*H*)-ones using $[Ce(L\text{-}Pro)_2]_2$ (Oxa)[a]

Entry	Aldehyde	Product	Yield (%)[b]
1	4-methylbenzaldehyde	**3a**	87
2	4-methoxybenzaldehyde	**3b**	88
3	4-hydroxybenzaldehyde	**3c**	87
4	2,5-dimethylbenzaldehyde	**3d**	86
5	3,4-dihydroxybenzaldehyde	**3e**	81
6	4-fluorobenzaldehyde	**3f**	89
7	4-chlorobenzaldehyde	**3g**	89
8	4-bromobenzaldehyde	**3h**	88
9	4-nitrobenzaldehyde	**3i**	96
10	3-nitrobenzaldehyde	**3j**	90
11	benzaldehyde	**3k**	89, 62[c]
12	1-naphthaldehyde	**3l**	84
13	anthracene-9-carbaldehyde	**3m**	84
14	3-phenylpropanal	**3n**	72
15	thiophene-2-carbaldehyde	**3o**	89
16	4-(allyloxy)benzaldehyde	**3p**	87

[a] Reaction conditions: Anthranilamide (1.0 mmol), benzaldehyde (1.0 mmol), $[Ce(L\text{-}Pro)_2]_2$ (Oxa) (5 mol%).
[b] Isolated yield.
[c] In the presence of catalyst and solvent at r.t.

We proposed a mechanism for the formation of 2-aryl-2,3-dihydroquinazolin-4(1*H*)-one derivatives as shown in Figure 3. Based on the experimental results, the mechanism of the reaction is thought to proceeded through condensation of aldehyde with anthranilamide, which subsequently forms an imine intermediate that cyclizes to afford the corresponding 2-aryl-2,3-dihydroquinazolin-4(1*H*)-one as the desired product, as shown in Figure 3.[21,14] SEM images of native (A) and used catalyst (B) are shown in Figure 4; their analysis confirmed that the morphology of the fresh catalyst and the catalyst after the reaction are similar. IR (C) and X-ray diffraction pattern (D) of the catalyst are shown in Figure 5.

All the products were compared with authentic samples and characterized by ^{1}H, ^{13}C NMR, IR and mass spectroscopic analyses.[8,21] All spectral data matched the corresponding data in the literature.

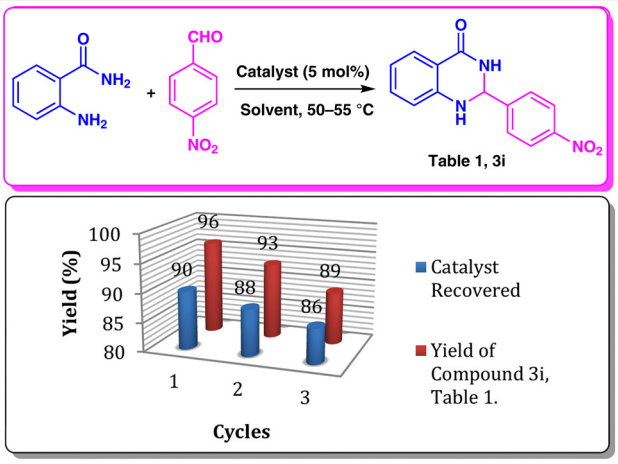


Figure 2 Recyclability of the catalyst

All chemical reagents and solvents were used without specific treatment. The respective reactions were monitored using thin-layer chromatography (TLC) MACHEREY-NAGEL (SIL G/UV$_{254}$). The compounds were purified by recrystallization. ^{1}H and ^{13}C NMR spectra were re-

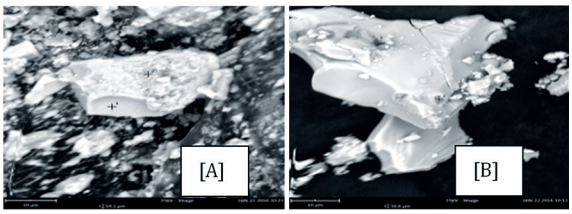


Figure 4 SEM images of native and used catalyst

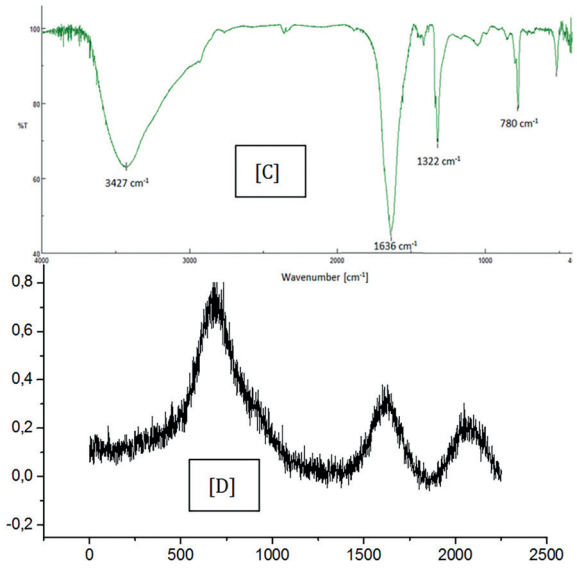


Figure 5 Catalyst IR (C) and X-ray diffraction pattern (D)

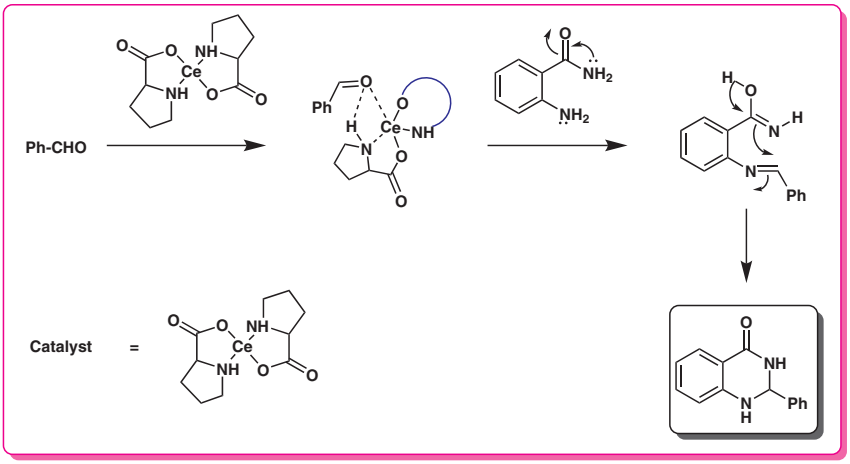


Figure 3 Proposed mechanistic pathway for the synthesis of 2,3-diphenyl-2,3-dihydroquinazolin-4(1*H*)-one mediated by [Ce(L-Pro)$_2$]$_2$ (Oxa)[21]

corded in $CDCl_3$ with a Bruker spectrometer (300 MHz, and 75 MHz, respectively). The IR spectra were recorded with a FT/IR 4100 type A spectrometer (Jasco).

[Ce(L-Pro)$_2$]$_2$(Oxa)[22]

L-Proline (2.7 mmol) was dissolved in methanol (15 ml), an aqueous solution of sodium hydroxide (2.7 mmol in 1 mL) was added at room temperature, and the mixture was stirred for 10 minutes. Then cerium (III) chloride (1.4 mmol) was added to it, and the reaction mixture was stirred for 45 minutes. Then a few drops of sodium oxalate solution (0.1 g mL^{-1}) were added to it. It was used as a precipitating agent. The semi-solid was centrifuged, washed with methanol, and dried overnight at 40 °C to obtain a pale yellow semi-solid.

2-(*p*-Tolyl)-2,3-dihydroquinazolin-4(1*H*)-one; Typical Procedure[8,21]

[Ce(L-Pro)$_2$]$_2$ (Oxa) was dissolved in EtOH (10 mL), and anthranilamide (1.0 mmol) and 4-methyl benzaldehyde (1.0 mmol) were added. The reaction mixture was heated at 50–55 °C until completion of the reaction as indicated by TLC. The reaction mixture was cooled to r.t., and the catalyst was separated from the reaction mixture by filtration. The solvent was evaporated under reduced pressure and the crude compound was purified by column chromatography (EtOAc/hexane, 3:7).

Yield: 207 mg (87%); yellow solid; mp 223–225 °C.

IR (KBr): 3310, 3192, 3060, 2924, 2855, 1908, 1662, 1607, 1509 cm^{-1}.

^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.93 (d, *J* = 7.2 Hz, 1 H), 7.46 (d, *J* = 8.3 Hz, 2 H), 7.34–7.23 (m, 3 H), 6.89 (t, *J* = 7.2 Hz, 1 H), 6.65 (d, *J* = 7.2 Hz, 1 H), 5.86 (s, 1 H), 5.76 (br. s, 1 H), 4.34 (br. s, 1 H), 2.39 (s, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 167.15, 133.99, 129.73, 128.69, 127.29, 119.61, 114.53, 68.84, 29.57.

MS (ESI): *m*/*z* = 239 [M + H]$^+$.

2-(4-Methoxyphenyl)-2,3-dihydroquinazolin-4(1*H*)-one (3b)[8]

Yield: 223 mg (88%); light-yellow solid; mp 182–184 °C.

IR (KBr): 3448, 3315, 3183, 2923, 1676 cm^{-1}.

^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.91 (d, *J* = 7.2 Hz, 1 H), 7.45 (d, *J* = 8.3 Hz, 2 H), 7.34–7.23 (m, 3 H), 6.88 (t, *J* = 7.2 Hz, 1 H), 6.63 (d, *J* = 7.2 Hz, 1 H), 5.84 (s, 1 H), 5.75 (br. s, 1 H), 4.39 (br. s, 1 H), 3.89 (s, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 167.14, 133.97, 129.70, 128.67, 127.24, 119.60, 114.51, 68.83, 56.52.

MS (ESI): *m*/*z* = 255 [M + H]$^+$.

2-(4-Hydroxyphenyl)-2,3-dihydroquinazolin-4(1*H*)-one (3c)[8]

Yield: 208 mg (87%); white solid; mp 278–280 °C.

IR (KBr): 3302, 3187, 3068, 2932, 1668, 1612, 1509, 1486 cm^{-1}.

^{1}H NMR (300 MHz, $CDCl_3$): δ = 9.33 (br. s, 1 H), 7.75 (d, *J* = 7.7 Hz, 1 H), 7.37 (d, *J* = 7.1 Hz, 3 H), 7.22 (t, *J* = 7.3 Hz, 1 H), 6.83–6.69 (m, 4 H), 6.28 (br. s, 1 H), 5.72 (s, 1 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 160.98, 156.32, 146.66, 132.71, 128.07, 118.90, 115.65, 113.89, 65.59.

MS (ESI): *m*/*z* = 241 [M + H]$^+$.

2-(2,5-Dimethylphenyl)-2,3-dihydroquinazolin-4(1*H*)-one (3d)[8]

Yield: 216 mg (86%); yellow solid; mp 222–224 °C.

IR (KBr): 3312, 3192, 3061, 2924, 2855, 1908, 1662, 1607, 1509 cm^{-1}.

^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.89 (d, *J* = 6.9 Hz, 1 H), 7.50 (s, 1 H), 7.30 (t, *J* = 6.9 Hz, 1 H), 7.06 (s, 3 H), 6.85 (t, *J* = 6.9 Hz, 1 H), 6.69 (d, *J* = 7.9 Hz, 1 H), 6.20 (br. s, 1 H), 6.07 (s, 1 H), 2.37 (s, 3 H), 2.30 (s, 3 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 163.17, 147.12, 135.70, 133.42, 131.60, 129.05, 127.65, 126.69, 126.00, 125.14, 124.63, 115.84, 113.34, 113.04, 63.55, 19.25, 16.84.

MS (ESI): *m*/*z* = 253 [M + H]$^+$.

2-(3,4-Dihydroxyphenyl)-2,3-dihydroquinazolin-4(1*H*)-one (3e)[8,21]

Yield: 207 mg (81%); light-yellow solid; mp 288–290 °C.

^{1}H NMR (300 MHz, $CDCl_3$): δ = 8.83 (br. s, 1 H), 7.79–7.56 (m, 2 H), 7.40 (t, *J* = 7.5 Hz, 1 H), 7.20 (t, *J* = 7.1 Hz, 1 H),7.03 (s, 1 H), 6.88 (d, *J* = 8.3 Hz, 1 H), 6.81–6.65 (m, 2 H), 5.64 (s, 1 H), 3.58 (br. s, 2 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 162.66, 161.08, 150.89, 147.84, 147.43, 146.84, 146.29, 143.95, 143.64, 143.36, 132.97, 131.90, 130.57, 125.76, 124.97, 124.39, 124.21, 121.77, 118.63, 118.23, 116.38, 115.60, 113.80, 113.50, 113.35, 112.86, 112.46, 64.97.

MS (ESI): *m*/*z* = 257 [M + H]$^+$.

2-(4-Fluorophenyl)-2,3-dihydroquinazolin-4(1*H*)-one (3f)[8,21]

Yield: 215 mg (89%); yellow solid; mp 202–204 °C.

IR (KBr): 3300, 3183, 3066, 2929, 1658, 1610, 1508, 1482 cm^{-1}.

^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.94 (d, *J* = 8.3 Hz, 1 H), 7.62–7.57 (m, 2 H), 7.37–7.32 (m, 1 H), 7.14 (t, *J* = 8.3 Hz, 2 H), 6.92 (t, *J* = 8.3 Hz, 1 H), 6.66 (d, *J* = 8.3 Hz, 1 H), 5.91 (s, 1 H), 5.78 (br. s, 1 H), 4.34 (br. s, 1 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 161.87, 158.69, 146.93, 132.64, 128.29, 128.18, 126.86, 117.83, 114.52, 114.24, 113.75, 66.39.

MS (ESI): *m*/*z* = 243 [M + H]$^+$.

2-(4-Chlorophenyl)-2,3-dihydroquinazolin-4(1*H*)-one (3g)[8]

Yield: 229 mg (89%); light-yellow solid; mp 196–198 °C.

^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.89 (d, *J* = 7.1 Hz, 1 H), 7.56 (d, *J* = 8.0 Hz, 2 H), 7.44 (d, *J* = 8.9 Hz, 2 H), 7.30 (t, *J* = 7.1 Hz, 1 H), 6.85 (t, *J* = 7.1 Hz, 1 H), 6.61 (d, *J* = 7.1 Hz, 1 H), 5.82 (s, 1 H), 5.72 (br. s, 1 H), 4.30 (br. s, 1 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 163.07, 146.44, 139.03, 133.12, 132.21, 130.34, 130.08, 128.30, 127.76, 126.36, 121.04, 116.50, 113.73, 113.36, 65.61.

MS (ESI): *m*/*z* = 259 [M + H]$^+$.

2-(4-Bromophenyl)-2,3-dihydroquinazolin-4(1*H*)-one (3h)[8]

Yield: 265 mg (88%); yellow solid; mp 200–202 °C.

IR (KBr): 3306, 3188, 3060, 2927, 1658, 1607 cm^{-1}.

^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.90 (d, *J* = 7.1 Hz, 1 H), 7.54 (d, *J* = 8.0 Hz, 2 H), 7.43 (d, *J* = 8.9 Hz, 2 H), 7.31 (t, *J* = 7.1 Hz, 1 H), 6.88 (t, *J* = 7.1 Hz, 1 H), 6.63 (d, *J* = 7.1 Hz, 1 H), 5.84 (s, 1 H), 5.74 (br. s, 1 H), 4.31 (br. s, 1 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 163.09, 146.45, 139.05, 133.13, 132.22, 130.37, 130.09, 128.31, 127.77, 126.38, 121.06, 116.51, 113.74, 113.38, 65.60.

MS (ESI): *m*/*z* = 303 [M + H]$^+$.

2-(4-Nitrophenyl)-2,3-dihydroquinazolin-4(1*H*)-one (3i)[8]

Yield: 258 mg (96%); yellow solid; mp 204–206 °C.

IR (KBr): 3278, 3174, 3032, 2922, 2855, 1647, 1608, 1520, 1461 cm^{-1}.

^1H NMR (300 MHz, $CDCl_3$): δ = 8.31 (d, *J* = 8.3 Hz, 1 H), 7.96 (m, 1 H), 7.83–7.79 (m, 2 H), 7.41–7.35 (m, 1 H), 7.27 (s, 1 H), 6.98–6.92 (m, 1 H), 6.70 (d, *J* = 7.5 Hz, 1 H), 6.15 (br. s, 1 H), 6.05 (s, 1 H), 4.43 (br. s, 1 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 162.96, 147.60, 146.50, 145.97, 132.46, 126.78, 126.49, 122.25, 116.70, 113.70, 64.89.

MS (ESI): *m*/*z* = 270 $[M + H]^+$.

2-(3-Nitrophenyl)-2,3-dihydroquinazolin-4(1*H*)-one (3j)[8]

Yield: 242 mg (90%); yellow solid; mp 210–212 °C.

IR (KBr): 3293, 3190, 3072, 2924, 2854, 1654, 1608, 1533 cm^{-1}.

^1H NMR (300 MHz, $CDCl_3$): δ = 8.47 (s, 1 H), 8.30 (d, *J* = 7.9 Hz, 1 H), 7.97 (t, *J* = 7.9 Hz, 2 H),7.66 (t, *J* = 7.9 Hz, 1 H), 7.37 (t, *J* = 6.9 Hz, 1 H), 6.95 (t, *J* = 6.9 Hz, 1 H), 6.70 (d, *J* = 7.9 Hz, 1 H), 6.05 (s, 1 H), 5.91 (br. s, 1 H), 4.42 (br. s, 1 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 163.23, 146.98, 146.12, 142.74, 132.59, 132.13, 128.50, 126.66, 122.24, 120.94, 116.99, 114.02, 113.69, 65.08.

MS (ESI): *m*/*z* = 270 $[M + H]^+$.

2-Phenyl-2,3-dihydroquinazolin-4(1*H*)-one (3k)[8]

Yield: 201 mg (90%); light-yellow solid; mp 225–227 °C.

IR (KBr): 3302, 3184, 3061, 2924, 1658, 1612 cm^{-1}.

^1H NMR (300 MHz, $CDCl_3$): δ = 7.94 (d, *J* = 7.5 Hz, 1 H), 7.61–7.58 (m, 2 H), 7.47–7.44 (m, 2 H), 7.37–7.31 (m, 2 H), 6.91 (t, *J* = 6.7 Hz, 1 H), 6.66 (d, *J* = 7.5 Hz, 1 H), 5.91 (s, 1 H), 5.78 (br. s, 1 H), 4.39 (br. s, 1 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 164.76, 147.20, 138.50, 134.03, 130.15, 129.11, 128.71, 127.38, 119.68, 114.56, 69.06.

MS (ESI): *m*/*z* = 225 $[M + H]^+$.

2-(Naphthalen-1-yl)-2,3-dihydroquinazolin-4(1*H*)-one (3l)[8]

Yield: 230 mg (84%); yellow solid; mp 208–210 °C.

IR (KBr): 3311, 3190, 3061, 2926, 2845, 1910, 1664, 1604, 1506 cm^{-1}.

^1H NMR (300 MHz, $CDCl_3$): δ = 8.11–8.08 (m, 2 H), 8.01 (d, *J* = 1.5 Hz, 1 H), 7.47 (t, *J* = 8.3 Hz, 1 H), 7.37–7.25 (m, 3 H), 7.19–7.17 (m, 3 H), 6.88 (t, *J* = 8.3 Hz, 1 H), 6.61 (d, *J* = 8.3 Hz, 1 H), 6.10 (s, 1 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 163.11, 145.26, 133.86, 130.13, 128.96, 128.71, 128.42, 126.94, 126.74, 119.61, 114.83, 74.62.

MS (ESI): *m*/*z* = 275 $[M + H]^+$.

2-(Anthracen-9-yl)-2,3-dihydroquinazolin-4(1*H*)-one (3m)[8]

Yield: 272 mg (84%); yellow solid; mp 235–237 °C.

IR (KBr): 3443, 2922, 2852, 1717, 1461, 1375, 1274 cm^{-1}.

^1H NMR (300 MHz, $CDCl_3$): δ = 8.54 (s, 1 H), 8.07–8.01 (m, 3 H), 7.51–7.34 (m, 6 H), 7.24 (m, 2 H), 6.96 (t, *J* = 6.7 Hz, 1 H), 6.67 (d, *J* = 8.3 Hz, 1 H), 6.22 (s, 1 H), 4.63 (s, 1 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 162.20, 130.86, 129.25, 125.34, 114.71, 63.89.

MS (ESI): *m*/*z* = 325 $[M + H]^+$.

2-Phenethyl-2,3-dihydroquinazolin-4(1*H*)-one (3n)[8]

Yield: 181 mg (72%); light-yellow solid; mp 209–211 °C.

IR (KBr): 3445, 3315, 3185, 2934, 1673, 1594, 1473, 1235, 1130, 1013, 750 cm^{-1}.

^1H NMR (300 MHz, $CDCl_3$): δ = 7.87 (d, *J* = 8.1 Hz, 1 H), 7.37–7.21 (m, 4 H), 6.85 (t, *J* = 7.2 Hz, 1 H), 6.68–6.52 (m, 3 H), 5.75 (br. s, 1 H), 4.91 (t, *J* = 5.4 Hz, 1 H), 4.12 (br. s, 1 H), 2.89–2.75 (m, 2 H), 2.14–2.10 (q, *J* = 7.2 Hz, 2 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 170.33, 163.57, 147.73, 147.08, 140.02, 132.84, 131.84, 130.75, 127.43, 126.92, 126.26, 124.42, 116.06, 115.78, 114.30, 113.79, 113.23, 63.00, 35.58, 28.36, 27.99.

MS (ESI): *m*/*z* = 253 $[M + H]^+$.

2-(Thiophen-2-yl)-2,3-dihydroquinazolin-4(1*H*)-one (3o)[8]

Yield: 204 mg (89%); light-yellow solid; mp 210–212 °C.

IR (KBr): 3448, 2923, 2853, 1763, 1651, 1457, 1376 cm^{-1}.

^1H NMR (300 MHz, $CDCl_3$): δ = 7.93 (d, *J* = 7.5 Hz, 1 H), 7.41–7.32 (m, 2 H), 7.22 (d, *J* = 3.7 Hz, 1 H), 7.02 (t, *J* = 4.5 Hz, 1 H), 6.92 (t, *J* = 7.5 Hz, 1 H), 6.70 (d, *J* = 8.3 Hz, 1 H), 6.20 (s, 1 H), 6.13 (br. s, 1 H), 4.56 (br. s, 1 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 161.95, 132.33, 126.49, 125.26, 124.89, 124.76, 116.93, 113.72, 102.96, 62.28.

MS (ESI): *m*/*z* = 231 $[M + H]^+$.

Anal. Calcd. for $C_{12}H_{10}N_2OS$: C, 62.59; H, 4.38; N, 12.16. Found: C, 62.51; H, 4.24; N, 12.12.

2-[4-(Allyloxy)phenyl]-2,3-dihydroquinazolin-4(1*H*)-one (3p)[8]

Yield: 243 mg (87%); light-yellow solid; mp 220–222 °C.

IR (KBr): 3299, 3187, 3061, 1651, 1611, 1509, 1484 cm^{-1}.

^1H NMR (300 MHz, $CDCl_3$): δ = 7.93 (d, *J* = 6.8 Hz, 1 H), 7.49 (d, *J* = 8.0 Hz, 2 H), 7.32 (t, *J* = 8.0 Hz, 1 H), 6.95 (d, *J* = 8.0 Hz, 2 H), 6.89 (t, *J* = 8.0 Hz, 1 H), 6.65 (d, *J* = 8.0 Hz, 1 H), 6.09–6.01 (m, 1 H), 5.84 (s, 1 H), 5.71 (br. s, 1 H), 5.40 (d, *J* = 17.1 Hz, 1 H), 5.30 (d, *J* = 9.1 Hz, 1 H), 4.57 (d, *J* = 5.7 Hz, 2 H), 4.33 (br. s, 1 H).

^{13}C NMR (50 MHz, $CDCl_3$): δ = 161.90, 160.92, 147.36, 133.98, 132.87, 131.76, 129.14, 127.86, 127.15, 125.75, 125.49, 125.27, 119.73, 117.23, 116.88, 114.12, 113.91, 68.03, 67.88, 67.13.

MS (ESI): *m*/*z* = 281 $[M + H]^+$.

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Supporting Information

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