

Benign and Efficient Synthesis of 2-Substituted 4(3*H*)-Quinazolinones Mediated by Iron(III) Chloride Hexahydrate in Refluxing Water

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Condensation of aldehydes with anthranilamide in refluxing water using iron(III) chloride hexahydrate as an oxidiant afforded 2-substituted 4(3*H*)-quinazolinones in good yields (77–93%). This method provides several advantages such as being environmentally friendly, having a simple work-up procedure, and affording high yields.

Owing to the increasing environmental concerns, chemical industries have been prompted to minimize the use of toxic and hazardous solvents in chemical manufacture. Technologies that pollute the environment must be replaced by benign alternatives. Organic chemists have been requested to develop clean, economical, and environmentally safer methodologies. Water as the reaction medium is generally considered as a cheap, safe, and environmentally benign alternative to unnatural solvents.[1] Furthermore, due to low solubility of common organic compounds in water, products can often be easily purified by simple filtration or extraction.[2] Nevertheless water has not yet become a widely accepted solvent for synthetic chemists. This may be due to concerns about the poor solubility of organic compounds in water. Recently, aqueous synthesis of heterocyclic compounds, such as benzimidazoles,[3] benzopyran derivatives,[4] and dihydropyrimidinones,[5] has received great interest owing to their broad range of potential biological and pharmaceutical applications.

2-Substituted 4(3*H*)-quinazolinones are of great interest due to the remarkable diversity of pharmacological uses.[6] Various approaches toward the synthesis of different 4(3*H*)-quinazolinone derivatives in organic solvents have been explored.[6–10] Nevertheless, some of these methods suffer from multi-step and tedious procedures, costly reagents, long reaction times, and often low yields. Among these methods 2-substituted 4(3*H*)-quinazolinones have been synthesized from anthranilamides and aldehyde using $NaHSO_3$,[8] DDQ,[9] or $CuCl_2$.[10] Iron(III) chloride hexahydrate ($FeCl_3 \cdot 6H_2O$) is a non-toxic, cheap, readily available, and water-miscible oxidant, which has not been utilized in the synthesis of 4(3*H*)-quinazolinones. To continue our work involving the use of water as a reaction medium,[11] herein, we report the synthesis of 2-substituted 4(3*H*)-quinazolinones by condensation of anthranilamide and aldehydes in refluxing water mediated by $FeCl_3 \cdot 6H_2O$.

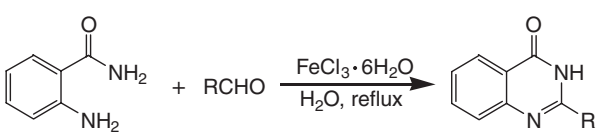
Results and Discussion

In order to determine if $CuCl_2$-promoted condensation of anthranilamide (**1**) and aldehydes[10] could occur in water, an environmentally benign solvent, we examined the one-pot reaction of compound **1**, 4-chlorobenzaldehyde (**2a**), and $CuCl_2 \cdot 2H_2O$ (1:1:2) in refluxing water. Only a trace amount of 2-(4-chlorophenyl)-4(3*H*)-quinazolinone (**3a**) was obtained after the reaction mixture had refluxed in water for 1 h. However, $FeCl_3 \cdot 6H_2O$ was found to promote the reaction efficiently and gave compound **3a** in 92% yield under the same conditions.

We then extended our studies on various substrates. The reaction of compound **1** (1 equiv) with aldehydes (**2a**–**2p**, 1 equiv) in the presence of $FeCl_3 \cdot 6H_2O$ (2 equiv) in refluxing water for 1 h afforded 2-substituted 4(3*H*)-quinazolinones **3a**–**3p** (Scheme 1). Benzaldehyde, substituted benzaldehydes containing electron-donating or electron-withdrawing groups as well as cinnamaldehyde (**2o**) and 2-furaldehyde (**2p**) were employed in this reaction. The work-up procedure is very straightforward, that is, the products were isolated and purified by simple filtration and recrystallization. Our protocol avoids the use of organic solvents during reaction process, making it superior to the previous methods.[8,10] Several aliphatic aldehydes (**2q**–**2t**) were also examined and were found to give the expected products in excellent yields. The work-up procedure was different and required extraction with methylene dichloride, because the products are soluble in water. The yields and melting points of 2-substituted 4(3*H*)-quinazolinones are summarized in Table 1.

The reaction proceeded to give products **3a**–**3t** in good to excellent yields (77–93%). As shown in Table 1, the electronic property of the substituents on the aromatic ring did not have an obvious effect on the yields under the current reaction conditions. A possible reason for the good performance of all of the substrates is the good solubility of anthranilamide and $FeCl_3 \cdot 6H_2O$ in refluxing water.

All of the 2-substituted 4(3*H*)-quinazolinones have been



1 **2** **3**

Scheme 1.

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Table 1. Yields and Melting Points of 2-Substituted Compound 4(3*H*)-Quinazolinones **3a–3t** Synthesized by the Reaction of **1** with Aldehydes **2a–2t** in the Presence of $FeCl_3 \cdot 6H_2O$ in Refluxing Water for 1 h

Entry	R	Product	Yield/%	Mp/°C (literature)
1	4-ClC_6H_4	**3a**	92	>300 (>300[9b])
2	4-BrC_6H_4	**3b**	87	294–295
3	2,4-$Cl_2C_6H_3$	**3c**	86	252–253
4	4-NCC_6H_4	**3d**	85	>300
5	3-$NO_2C_6H_4$	**3e**	88	>300 (>300[9b])
6	4-$NO_2C_6H_4$	**3f**	90	>300
7	2-$NO_2C_6H_4$	**3g**	85	224–226
8	C_6H_5	**3h**	90	237–238 (238[9b])
9	4-$CH_3C_6H_4$	**3i**	93	238–239 (239[9b])
10	4-$CH_3OC_6H_4$	**3j**	90	244–245 (245[9b])
11	3-CH_3O-4-HOC_6H_3	**3k**	85	266–267
12	4-$(CH_3)_2NC_6H_4$	**3l**	89	240–242
13	3,4-$(CH_2O_2)C_6H_3$	**3m**	80	277–278 (278[9b])
14	4-HOC_6H_4	**3n**	77	>300
15	–CH=CH–C_6H_5	**3o**	86	249–251 (223–226[7a])
16	2-Furyl	**3p**	80	219–221 (218–220[10])
17	H[a)]	**3q**	92	215–216 (215[12])
18	CH_3[b)]	**3r**	90	239–241 (240–240.5[13])
19	CH_3CH_2[c)]	**3s**	90	231–233 (233[14])
20	$CH_3CH_2CH_2$[c)]	**3t**	92	200–202 (200–201[15])

a) Paraformaldehyde was used. b) Paraldehyde was used. c) 1.5 equiv was employed due to low boiling point.

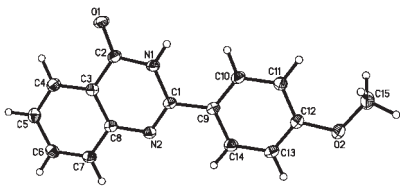


Fig. 1. ORTEP diagram of compound **3j**. Selected bond length (Å): O1–C2 1.234(2), N1–C2 1.373(2), N1–C1 1.382(2), N2–C1 1.297(2), N2–C8 1.388(2), C1–C9 1.482(2), C2–C3 1.448(2), C3–C8 1.405(2).

characterized by ^{1}H NMR, ^{13}C NMR, and IR spectra, and the known compounds were confirmed by comparison of their spectral data and melting points with those reported in the literature.[7a,9a,10,12–15] Using compound **3f** as an example, in the IR spectrum, there is a strong carbonyl absorption at 1683 cm^{-1}. In the ^{1}H NMR spectrum, aromatic protons appeared at δ 7.59 (1H, t, $J = 7.5$ Hz), 7.80 (1H, d, $J = 8.0$ Hz), 7.89 (1H, t, $J = 7.6$ Hz), 8.19 (1H, d, $J = 8.1$ Hz), 8.38 (2H, d, $J = 9.1$ Hz), and 8.43 (2H, d, $J = 9.1$ Hz). The NH proton does not appear probably due to a fast exchange with water present in DMSO-d_6. In the ^{13}C NMR spectrum of compound **3f**, besides the peaks for the aromatic carbons, there are two peaks at 162.1 and 150.8 ppm for the carbons of the amide and *C*=N groups, respectively. The quinazolinone skeleton of products **3a–3t** was further established by the single crystal structure (Fig. 1) of 2-(4-methoxyphenyl)-4(3*H*)-quinazolinone (**3j**) which was grown from a mixture of DMF and water. The double bond character of C1–N2 is confirmed by its length (1.297 Å). The quinazolinone moiety is nearly coplanar, and the dihedral angle between the 4-methoxyphenyl ring and the pyrimidone ring is only 4.34°.

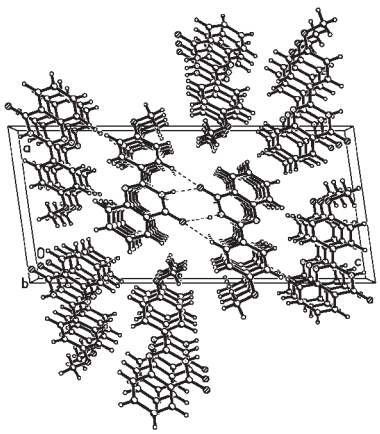
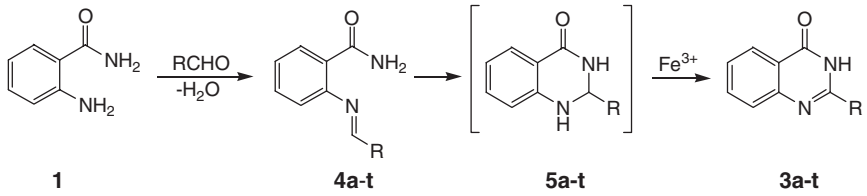


Fig. 2. The packing arrangement in a unit cell of compound **3j**.

The packing arrangement in the unit cell is shown in Fig. 2. Intermolecular hydrogen bonds occur between adjacent molecules involving a N1–H1···O1 (2.853 Å) hydrogen bond and a C10–H10···O1 (3.182 Å) hydrogen bond. Hydrogen-bonded layers are stacked upon one another by translation and held together by van der Waals attractions.

A possible mechanism for the formation of 2-substituted 4(3*H*)-quinazolinone derivatives **3a–3t** is shown in Scheme 2. Reaction of compound **1** with aldehydes **2a–2t** results in Schiff bases **4a–4t**. Intramolecular cyclization of the Schiff bases affords intermediates **5a–5t**, which undergo oxidation with $FeCl_3 \cdot 6H_2O$ to afford products **3a–3t**. The oxidation of compounds **5a–5t** to products **3a–3t** requires two equiv of $FeCl_3 \cdot 6H_2O$ for completion, and $FeCl_3$ is converted to $FeCl_2$. Schiff bases **4a–4t** were isolated in the early stage of the reactions, which supports their role in the reaction mechanism.



Scheme 2.

Table 2. Conditions and Yields of Compound **3a** for the Reaction of Compound **1** with Compound **2a** in Various Solvents for 1 h

Entry	Oxidant	Media	Temp/°C	Yield/%
1	$CuCl_2 \cdot 2H_2O$	H_2O	100	trace
2	$FeCl_3 \cdot 6H_2O$	H_2O	100	92
3	$FeCl_3 \cdot 6H_2O$	DMF	100	36
4	$FeCl_3 \cdot 6H_2O$	DMSO	100	31
5	$FeCl_3 \cdot 6H_2O$	EtOH	80	22

To compare with the result in water, organic solvents such as DMF, DMSO, and EtOH as the reaction media were also examined. The reaction conditions and yields for the reaction of compound **1** with compound **2a** and $CuCl_2 \cdot 2H_2O/FeCl_3 \cdot 6H_2O$ (1:1:2) in water and organic solvents for 1 h are listed in Table 2. As seen from Table 2, water exhibited obvious advantages over organic solvents and gave much higher product yields. It should be noted that other reagents such as $CuSO_4 \cdot 5H_2O$, $CeCl_3 \cdot 7H_2O$, $SnCl_4 \cdot 5H_2O$, $NiCl_2 \cdot 6H_2O$, $KMnO_4$, $Pb(OAc)_2 \cdot 3H_2O$, $BiCl_3$, $CdCl_2 \cdot 2.5H_2O$, $ZnCl_2$, and $NaNO_2$ were ineffective, demonstrating the unique ability of $FeCl_3 \cdot 6H_2O$ in this condensation.

In addition, the aqueous filtrate could be reused for the next batch reaction. As an example, compound **3a** was obtained without any decrease of yield with the addition of compounds **1** and **2a** and $FeCl_3 \cdot 6H_2O$ in a molar ratio of 1:1:2 in all five subsequent runs (Fig. 3). In fact, there was a slight increase in the yield in the subsequent runs, probably due to the accumulation of excess unreacted starting materials in the aqueous filtrate.

In conclusion, we have developed a rapid, efficient, and environmentally friendly method for the synthesis of 2-substituted 4(3*H*)-quinazolinones by condensation of various aldehydes with anthranilamide in refluxing water using $FeCl_3 \cdot 6H_2O$ as an oxidant. The current protocol provides several advantages such as simple work-up procedure, good yields, and environmental friendly solvent.

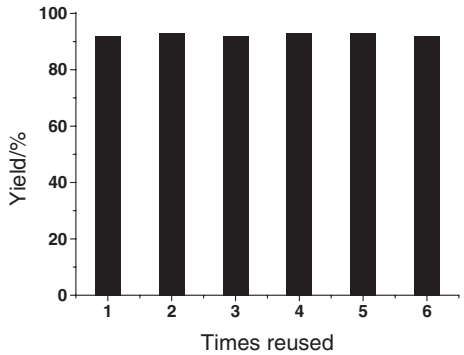


Fig. 3. Yields of compound **3a** in reused water phase.

Experimental

General. ^{1}H NMR spectra were recorded on a Bruker Avance-300 (300 MHz) spectrometer, and chemical shifts (δ) are reported in parts per million relative to tetramethylsilane. Coupling constants (J) are reported in Hz. ^{13}C NMR spectra were recorded on a Bruker Avance-300 (75 MHz) spectrometer with complete proton decoupling, and chemical shifts are reported in parts per million relative to the solvent resonance as the internal standard (DMSO-d_6, δ 39.52 ppm). IR spectra were taken on a Bruker Vector-22 spectrometer in KBr pellets and are reported in cm^{-1}. Elemental analyses were performed by using a Vario ELIII elemental analyzer (for CHN). High-resolution mass spectra (HRMS) were obtained on a Micromass GCT mass spectrometer in a positive EI mode. X-ray crystallographic analysis was performed with a Rigaku Mercury CCD area detector (graphite monochromator, Mo Kα radiation $\lambda = 0.71073$ Å). Melting points were determined on an XT-4 apparatus (Beijing Tech Instrument Co., China). Analytical TLC and column chromatography were performed on silica gel GF254, and silica gel H60, respectively.

General Procedure for the Synthesis of Compounds 3a–3t. A mixture of an aldehyde (**2a**–**2r**, 1 mmol; **2s**–**2t**, 1.5 mmol), anthranilamide (**1**, 136 mg, 1 mmol), and $FeCl_3 \cdot 6H_2O$ (540 mg, 2 mmol) in refluxing water (10 mL) was stirred for 1 h. The reaction mixture was cooled to room temperature and filtered to give the crude product, which was purified by recrystallization from DMF and water. For the synthesis of compounds **3q**–**3t**, the work-up procedure was different. After cooling to room temperature, the reaction mixture was neutralized with sodium acetate and then extracted with methylene dichloride (10 mL × 3). The extracted solutions were dried over anhydrous magnesium sulfate. The crude product was obtained after evaporation in vacuo and purified by recrystallization from ethyl acetate. Crystals of compound **3j** were obtained from a mixture of DMF and water at room temperature.

2-(4-Bromophenyl)-4(3*H*)-quinazolinone (3b): IR (KBr) ν 1675 (C=O), 1603, 1560, 1482, 1309, 1150, 1066, 1010, 941, 772, 727, 550 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6) δ 7.54 (t, 1H, J = 7.6 Hz, Ph), 7.75 (d, 1H, J = 7.5 Hz, Ph), 7.77 (d, 2H, J = 8.7 Hz, Ph), 7.85 (t, 1H, J = 7.4 Hz, Ph), 8.13 (d, 2H, J = 8.7 Hz, Ph), 8.16 (d, 1H, J = 8.0 Hz, Ph); ^{13}C NMR (75 MHz, DMSO-d_6) δ 121.0, 125.2, 125.8, 126.7, 127.5, 129.8 (2C), 131.6 (2C), 131.9, 134.6, 148.5, 151.4, 162.1; Anal. Calcd for $C_{14}H_9BrN_2O$: C, 55.84; H, 3.01; N, 9.30%. Found: C, 55.66; H, 3.12; N, 9.41%; HRMS (+EI) calcd for (M^+): 299.9898, found: 299.9896.

2-(2,4-Dichlorophenyl)-4(3*H*)-quinazolinone (3c): IR (KBr) ν 1674 (C=O), 1606, 1470, 1302, 1151, 1110, 955, 871, 806, 769, 687 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6) δ 7.59 (t, 1H, J = 7.2 Hz, Ph), 7.64–7.68 (m, 2H, Ph), 7.72 (d, 1H, J = 8.3 Hz, Ph), 7.83 (s, 1H, Ph), 7.87 (t, 1H, J = 7.7 Hz, Ph), 8.18 (d, 1H, J = 8.0 Hz, Ph); ^{13}C NMR (75 MHz, DMSO-d_6) δ 121.3, 125.8, 127.2, 127.5, 130.4, 130.6, 131.3, 131.4, 131.7, 134.6, 135.3, 148.4, 150.9,

161.3; Anal. Calcd for $C_{14}H_8Cl_2N_2O$: C, 57.76; H, 2.77; N, 9.62%. Found: C, 57.59; H, 2.74; N, 9.54%; HRMS (+EI) calcd for (M^+): 290.0014, found: 290.0015.

2-(4-Cyanophenyl)-4(3*H*)-quinazolinone (3d): IR (KBr) ν 2225 (C≡N), 1681 (C=O), 1603, 1558, 1468, 1311, 1151, 946, 843, 775, 554 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6) δ 7.58 (t, 1H, J = 7.5 Hz, Ph), 7.79 (d, 1H, J = 8.1 Hz, Ph), 7.88 (t, 1H, J = 7.6 Hz, Ph), 8.05 (d, 2H, J = 8.2 Hz, Ph), 8.18 (d, 1H, J = 8.2 Hz, Ph), 8.34 (d, 2H, J = 8.2 Hz, Ph); ^{13}C NMR (75 MHz, DMSO-d_6) δ 113.5, 118.3, 121.1, 125.8, 127.2, 127.7, 128.6 (2C), 132.5 (2C), 134.7, 136.8, 148.3, 150.9, 162.0; Anal. Calcd for $C_{15}H_9N_3O$: C, 72.87; H, 3.67; N, 16.99%. Found: C, 72.95; H, 3.74; N, 16.86%; HRMS (+EI) calcd for (M^+): 247.0746, found: 247.0752.

2-(4-Nitrophenyl)-4(3*H*)-quinazolinone (3f): IR (KBr) ν 1682 (C=O), 1609, 1590, 1523, 1469, 1348, 1151, 1109, 946, 859, 772, 707, 556 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6) δ 7.59 (t, 1H, J = 7.5 Hz, Ph), 7.80 (d, 1H, J = 8.0 Hz, Ph), 7.89 (t, 1H, J = 7.6 Hz, Ph), 8.19 (d, 1H, J = 8.1 Hz, Ph), 8.38 (d, 2H, J = 9.1 Hz, Ph), 8.43 (d, 2H, J = 9.1 Hz, Ph); ^{13}C NMR (75 MHz, DMSO-d_6) δ 121.2, 123.6 (2C), 125.9, 127.3, 127.8, 129.3 (2C), 134.8, 138.6, 148.3, 149.0, 150.8, 162.1; Anal. Calcd for $C_{14}H_9N_3O_3$: C, 62.92; H, 3.39; N, 15.72%. Found: C, 62.79; H, 3.57; N, 15.88%; HRMS (+EI) calcd for (M^+): 267.0644, found: 267.0639.

2-(2-Nitrophenyl)-4(3*H*)-quinazolinone (3g): IR (KBr) ν 1664 (C=O), 1603, 1574, 1526, 1465, 1355, 1315, 1255, 1148, 943, 848, 776, 760, 716, 622, 543 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6) δ 7.59 (t, 1H, J = 7.5 Hz, Ph), 7.66 (d, 1H, J = 8.1 Hz, Ph), 7.81–7.96 (m, 4H, Ph), 8.21 (t, 2H, J = 9.1 Hz, Ph); ^{13}C NMR (75 MHz, DMSO-d_6) δ 121.2, 124.5, 125.9, 127.1, 127.4, 129.2, 131.4, 131.5, 133.9, 134.7, 147.5, 148.5, 151.6, 161.5; Anal. Calcd for $C_{14}H_9N_3O_3$: C, 62.92; H, 3.39; N, 15.72%. Found: C, 62.80; H, 3.46; N, 15.80%; HRMS (+EI) calcd for (M^+): 267.0644, found: 267.0641.

2-(4-Hydroxyphenyl-3-methoxy)-4(3*H*)-quinazolinone (3k): IR (KBr) ν 1664 (C=O), 1578, 1527, 1483, 1288, 1248, 1120, 1027, 1018, 866, 769 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6) δ 3.90 (s, 3H, CH_3), 6.93 (d, 1H, J = 8.3 Hz, Ph), 7.49 (t, 1H, J = 7.4 Hz, Ph), 7.71 (d, 1H, J = 8.0 Hz, Ph), 7.75 (d, 1H, J = 8.3 Hz, Ph), 7.82 (s, 1H, Ph), 7.85 (t, 1H, J = 7.2 Hz, Ph), 8.13 (d, 1H, J = 7.8 Hz, Ph), 9.82 (s, 1H, OH); ^{13}C NMR (75 MHz, DMSO-d_6) δ 55.8, 111.3, 115.4, 120.6, 121.5, 123.4, 125.8, 125.9, 127.2, 134.5, 147.5, 149.0, 149.9, 162.3; Anal. Calcd for $C_{15}H_{12}N_2O_3$: C, 67.16; H, 4.51; N, 10.44%. Found: C, 67.23; H, 4.65; N, 10.30%; HRMS (+EI) calcd for (M^+): 268.0848, found: 268.0839.

2-(4-Dimethylaminophenyl)-4(3*H*)-quinazolinone (3l): IR (KBr) ν 1665 (C=O), 1607, 1590, 1535, 1487, 1370, 1292, 1208, 823, 768 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6) δ 3.04 (s, 6H, 2 × CH_3), 6.82 (d, 2H, J = 9.0 Hz, Ph), 7.46 (t, 1H, J = 7.7 Hz, Ph), 7.68 (d, 1H, J = 8.2 Hz, Ph), 7.81 (t, 1H, J = 7.6 Hz, Ph), 8.10 (d, 2H, J = 9.0 Hz, Ph), 8.11 (d, 1H, J = 8.0 Hz, Ph); ^{13}C NMR (75 MHz, DMSO-d_6) δ 39.6, 111.3 (2C), 118.7, 120.3, 125.6, 125.8, 127.0, 128.9 (2C), 134.6, 145.3, 149.3, 152.3, 162.4; Anal. Calcd for $C_{16}H_{15}N_3O$: C, 72.43; H, 5.70; N, 15.84%. Found: C, 72.57; H, 5.81; N, 15.73%; HRMS (+EI) calcd for (M^+): 265.1215, found: 265.1218.

2-(4-Hydroxyphenyl)-4(3*H*)-quinazolinone (3n): IR (KBr) ν 1668 (C=O), 1601, 1559, 1524, 1489, 1451, 1377, 1344, 1288, 1237, 1183, 948, 846, 765, 638, 612, 542, 526 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6) δ 6.91 (d, 2H, J = 8.7 Hz, Ph), 7.48 (t, 1H, J = 7.5 Hz, Ph), 7.69 (d, 1H, J = 7.9 Hz, Ph), 7.82 (t, 1H, J = 7.6 Hz, Ph), 8.09 (d, 2H, J = 8.7 Hz, Ph), 8.13 (d, 1H, J = 8.0 Hz, Ph), 10.22 (s, 1H, OH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 115.4 (2C), 120.6, 123.2, 125.8, 125.9, 127.2, 129.6 (2C), 134.5, 149.1, 152.1, 160.6, 162.3; Anal. Calcd for $C_{14}H_{10}N_2O_2$: C, 70.58; H, 4.23; N, 11.76%. Found: C, 70.46; H, 4.17; N, 11.85%; HRMS (+EI) calcd for (M^+): 238.0742, found: 238.0750.

2-Styryl-4(3*H*)-quinazolinone (3o): IR (KBr) ν 1669 (C=O), 1608, 1582, 1447, 1295, 1145, 968, 832, 767, 695 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6) δ 7.02 (d, 1H, J = 16.2 Hz, H_{vinyl}), 7.40–7.54 (m, 4H, Ph), 7.67–7.70 (m, 3H, Ph), 7.83 (t, 1H, J = 7.7 Hz, Ph), 7.98 (d, 1H, J = 16.2 Hz, H_{vinyl}), 8.12 (d, 1H, J = 7.8 Hz, Ph); ^{13}C NMR (75 MHz, DMSO-d_6) δ 121.1 (2C), 125.9, 126.2, 127.1, 127.6, 129.1 (2C), 129.8 (2C), 134.5, 135.0, 138.3, 149.0, 151.4, 161.7; Anal. Calcd for $C_{16}H_{12}N_2O$: C, 77.40; H, 4.87; N, 11.28%. Found: C, 77.33; H, 4.92; N, 11.24%; HRMS (+EI) calcd for (M^+): 248.0950, found: 248.0943.

2-(4-Furyl)-4(3*H*)-quinazolinone (3p): IR (KBr) ν 1663 (C=O), 1602, 1553, 1457, 1310, 965, 770, 753 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6) δ 6.73 (dd, 1H, J = 3.4, 1.3 Hz), 7.49 (t, 1H, J = 7.5 Hz, Ph), 7.59 (d, 1H, J = 3.4 Hz), 7.68 (d, 1H, J = 8.1 Hz, Ph), 7.81 (t, 1H, J = 7.6 Hz, Ph), 7.97 (d, 1H, J = 1.3 Hz), 8.11 (d, 1H, J = 7.8 Hz, Ph); ^{13}C NMR (75 MHz, DMSO-d_6) δ 112.5, 114.5, 121.1, 125.9, 126.4, 127.2, 134.6, 144.0, 146.1, 146.5, 148.6, 161.5; Anal. Calcd for $C_{12}H_8N_2O_2$: C, 67.92; H, 3.80; N, 13.20%. Found: C, 67.84; H, 3.78; N, 13.21%; HRMS (+EI) calcd for (M^+): 212.0586, found: 212.0592.

X-ray Crystallographic Data for 3j. $C_{15}H_{12}N_2O_2$, colorless, crystal dimensions = 0.51 × 0.30 × 0.10 mm^3, monoclinic, space group $P2_1/n$, a = 10.4202(14), b = 5.0199(5), c = 22.802(3) Å, β = 98.102(4)°, V = 1180.9(3) $Å^3$, M_r = 252.27, Z = 4, D_{calcd} = 1.419 g cm^{-3}, λ = 0.71070 Å, μ (Mo Kα) = 0.096 mm^{-1}, $F(000)$ = 880, S = 1.156, R_1 = 0.0532, wR_2 = 0.1264. Crystallographic data for the structure reported have been deposited in the Cambridge Crystallographic Data Centre (CCDC-249927). Crystallographic data have been deposited with Cambridge Crystallographic Data Centre. Copies of the data can be obtained free of charge via http://www.ccdc.cam.ac.uk/conts/retrieving.html (or from the Cambridge Crystallographic Data Centre, 12, Union Road, Cambridge, CB2 1EZ, UK; Fax: +44 1223 336033; e-mail: deposit@ccdc.cam.ac.uk).

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