

Lijiu Gao, Honglou Ji, Liangce Rong,\* Hongxia Han, Yanhui Shi, and Shujiang Tu

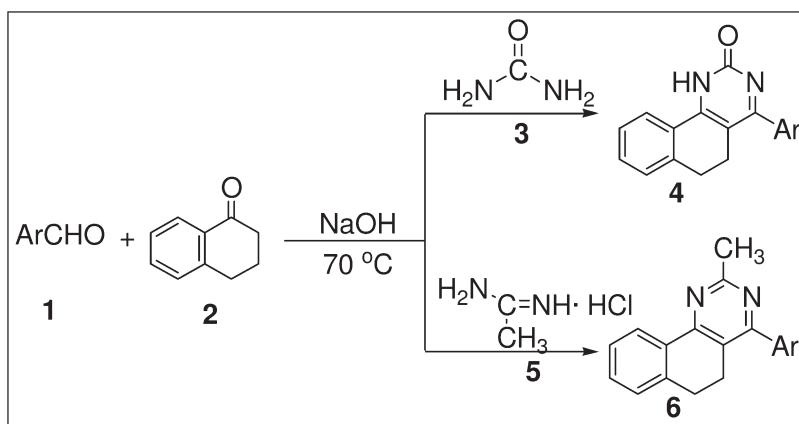
College of Chemistry and Chemical Engineering, Xuzhou Normal University, Key Laboratory of Biotechnology for Medicinal Plant, Xuzhou 221116, Jiangsu, People's Republic of China

\*E-mail: lcrong2005@yahoo.com

Received July 27, 2009

DOI 10.1002/jhet.318

Published online 2 March 2010 in Wiley InterScience (www.interscience.wiley.com).



An efficient and convenient method for the preparation of 5,6-dihydrobenzo[*h*]quinazoline derivatives by the multicomponent reactions of aromatic aldehydes, 3,4-dihydronaphthalen-1(2*H*)-one and urea or acetamidine hydrochloride, in the presence of sodium hydroxide under solvent-free conditions was reported. This method has the advantages of excellent yields, mild reaction conditions, easy work-up, and environmentally friendly procedure.

*J. Heterocyclic Chem.*, **47**, 358 (2010).

## INTRODUCTION

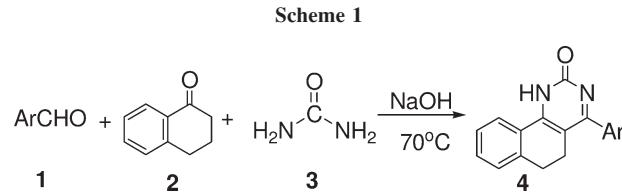
The quinazoline skeleton is a very important and useful scaffold in medicinal chemistry: it can be found as a pharmacophore in a wide variety of biologically active compounds, such as antitumors [1], antimicrobials [2], antivirals [3,4]. Benzoquinazoline, the important containing quinazoline skeleton system derivatives, is often found in different natural alkaloids, and these compounds also display specific biological activities, and often used as asdiuretic, anticancer, anticonvulsant, and antihypertensive agents [5–8].

Recently, there was an increasing emphasis on developing new environmentally safer chemical transformations by lessening/removing the toxic waste, where by-products from the chemical processes were avoided or minimized making them ecologically more acceptable. It is highly desirable to develop eco-friendly methods for producing organic fine chemicals. One of the major problems encountered in various chemical processes is the use of organic solvents. Hence, the organic transformations under solvent-free conditions are attracting increasing attentions [9–11]. Herein, we would like to report an efficient and facile method to synthesize 5,6-

dihydrobenzo[*h*]quinazoline derivatives under solvent-free conditions.

## RESULTS AND DISCUSSION

The synthesis process could be depicted as follows: at first, we try to prepare 4-aryl-5,6-dihydrobenzo[*h*]quinazolin-2(1*H*)-one derivatives under solvent-free conditions (Scheme 1). The aromatic aldehydes **1** (1 mmol), 3,4-dihydronaphthalen-1(2*H*)-one **2** (1 mmol) and urea **3** (1.5 mmol) were chosen as starting materials, and the reactants were blent enough in a mortar in presence of NaOH (0.1 g) as catalyst, then the mixture was introduced into a round flask and reacted under 70°C. To our delight, the reaction could be finished about 30 min and the 4-aryl-5,6-dihydrobenzo[*h*]quinazolin-2(1*H*)-one



**Table 1**

The results of synthesis of 4-aryl-5,6-dihydrobenzo[*h*]quinazolin-2(1*H*)-one.

Entry	Ar	Product	Yields (%)
1	C <sub>6</sub> H <sub>5</sub>	<b>4a</b>	80
2	4-CH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	<b>4b</b>	85
3	3,4-(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>3</sub>	<b>4c</b>	83
4	3,4-(CH <sub>3</sub> O) <sub>2</sub> C <sub>6</sub> H <sub>3</sub>	<b>4d</b>	87
5	4-FC <sub>6</sub> H <sub>4</sub>	<b>4e</b>	90
6	4-BrC <sub>6</sub> H <sub>4</sub>	<b>4f</b>	88
7	2-ClC <sub>6</sub> H <sub>4</sub>	<b>4g</b>	80
8	4-ClC <sub>6</sub> H <sub>4</sub>	<b>4h</b>	91
9	2,4-Cl <sub>2</sub> C <sub>6</sub> H <sub>3</sub>	<b>4i</b>	82
10	3,4-Cl <sub>2</sub> C <sub>6</sub> H <sub>3</sub>	<b>4j</b>	80

derivatives could be gained with excellent yields. The result of reaction is shown in Table 1. From Table 1 we could see the reaction was carried out smoothly and a series of 4-aryl-5,6-dihydrobenzo[*h*]quinazolin-2(1*H*)-one derivatives were obtained ignoring the properties of substitute groups on the aromatic aldehydes. So, we could say that substitute groups on the aromatic aldehydes do not affect this reaction. In addition, in this reaction the catalyst NaOH was necessary.

To extend this reaction to prepare more benzo[*h*]quinazoline derivatives, we replaced urea by acetamidine hydrochloride to react with aromatic aldehydes **1** and 3,4-dihydronaphthalen-1(2*H*)-one **2** under similar condition (Scheme 2), and we found that other benzo[*h*]quinazoline derivatives, 4-aryl-5,6-dihydro-2-methylbenzo[*h*]quinazoline could be gained with good yields. The result of reaction was listed in Table 2. In this reaction, we thought that two functions were played by NaOH, one it was used as catalyst to promote the reaction, and the another it reacted with acetamidine hydrochloride to release acetamidine.

The structures of **4** and **6** were characterized by <sup>1</sup>H NMR, IR, and HRMS spectra, and the structures of **6d** [12] was additionally confirmed by X-ray diffraction analysis. The crystal structure of is shown in Figure 1.

In conclusion, we have developed an efficient and facile process to synthesize a variety of 4-aryl-5,6-dihydrobenzo[*h*]quinazolin-2(1*H*)-one and 4-aryl-5,6-dihydro-2-methylbenzo[*h*]quinazoline derivatives *via* one-pot reaction of different aromatic aldehydes, 3,4-dihy-

**Table 2**

The results of synthesis of 4-aryl-5,6-dihydro-2-methylbenzo[*h*]quinazoline.

Entry	Ar <sup>1</sup>	Product	Yields (%)
1	4-CH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	<b>6a</b>	78
2	4-CH <sub>3</sub> OC <sub>6</sub> H <sub>4</sub>	<b>6b</b>	80
3	3,4-(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>3</sub>	<b>6c</b>	76
4	4-FC <sub>6</sub> H <sub>4</sub>	<b>6d</b>	85
5	3-ClC <sub>6</sub> H <sub>4</sub>	<b>6e</b>	79
6	4-ClC <sub>6</sub> H <sub>4</sub>	<b>6f</b>	89
7	3,4-Cl <sub>2</sub> C <sub>6</sub> H <sub>3</sub>	<b>6g</b>	88

dronaphthalen-1(2*H*)-one, and urea or acetamidine hydrochloride under solvent-free conditions. The mild reaction conditions, short reaction times, good to high yields, low cost, easy preparation, easy handling, and reusability of catalyst are the advantages of this method.

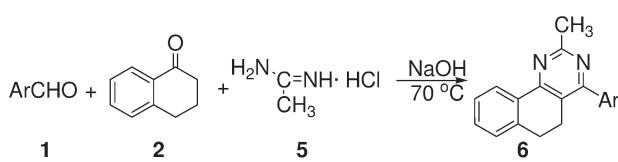
## EXPERIMENTAL

Melting points were determined on XT-5 microscopic melting-point apparatus and were uncorrected. IR spectra were recorded on a FT Bruker Tensor 27 spectrometer. <sup>1</sup>H NMR spectra were obtained from solution in DMSO-d<sub>6</sub> with Me<sub>4</sub>Si as internal standard using a Bruker-400 spectrometer. X-ray diffractions were recorded on a Siemens P4 or Simart-1000 diffractometer. HRMS spectra were obtained with a Bruker micrOTOF-Q 134 instrument.

**General procedure for the synthesis of 5,6-dihydrobenzo[*h*]quinazoline derivatives.** The mixture of aromatic aldehydes **1** (1 mmol), 3,4-dihydronaphthalen-1(2*H*)-one **2** (1 mmol), urea **3** (1.5 mmol) or acetamidine hydrochloride **5** (1.5 mmol), and NaOH (0.1 g) was put in a reaction flask, and the reagents were reacted at 70°C about 30 min. When the reactions were completed, the reaction mixture was poured into water (0.5% HCl), and then washed with water thoroughly. The product was filtered, dried, and recrystallized from 95% ethanol.

**5,6-Dihydro-4-phenylbenzo[*h*]quinazolin-2(1*H*)-one (4a).** m.p. 251–253°C; IR (KBr, n, cm<sup>-1</sup>): 3327, 3232, 3089, 3019, 2943, 2890, 2830, 1689, 1550, 1488, 1454, 1431, 1366, 1342, 1319, 1298, 1279, 1262, 1228, 1191, 1180, 1162, 1122, 1072, 1046, 1026, 981, 943, 895, 826, 770, 723, 700, 656, 638, 608, 595 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, DMSO-d<sub>6</sub>) ( $\delta$ , ppm): 2.59 (2H, t, J = 7.6 Hz, J = 7.6 Hz, CH<sub>2</sub>), 2.69 (2H, t, J = 7.2 Hz, J = 7.2 Hz, CH<sub>2</sub>), 7.15 (1H, d, J = 6.4 Hz, ArH), 7.20 (2H, t, J = 5.2 Hz, J = 5.4 Hz, ArH), 7.32–7.38 (4H, m, ArH), 7.58 (1H, d, J = 6.8 Hz, ArH), 8.57 (1H, br, ArH), 11.95 (1H, s, NH); HRMS *m/z* calculated for C<sub>18</sub>H<sub>14</sub>N<sub>2</sub>O [M+H]<sup>+</sup>: 275.1184, found: 275.1185.

**5,6-Dihydro-4-p-tolylbenzo[*h*]quinazolin-2(1*H*)-one (4b).** m.p. 243–244°C; IR (KBr, n, cm<sup>-1</sup>): 3462, 3275, 3062, 3000, 2909, 2859, 1667, 1595, 1509, 1465, 1427, 1375, 1323, 1231, 1150, 1091, 1064, 824, 759, 733 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, DMSO-d<sub>6</sub>) ( $\delta$ , ppm): 2.39 (3H, s, CH<sub>3</sub>), 2.67 (2H, t, J = 6.0 Hz, J = 7.6 Hz, CH<sub>2</sub>), 2.80 (2H, t, J = 6.4 Hz, J = 7.2 Hz, CH<sub>2</sub>), 7.37

**Scheme 2**

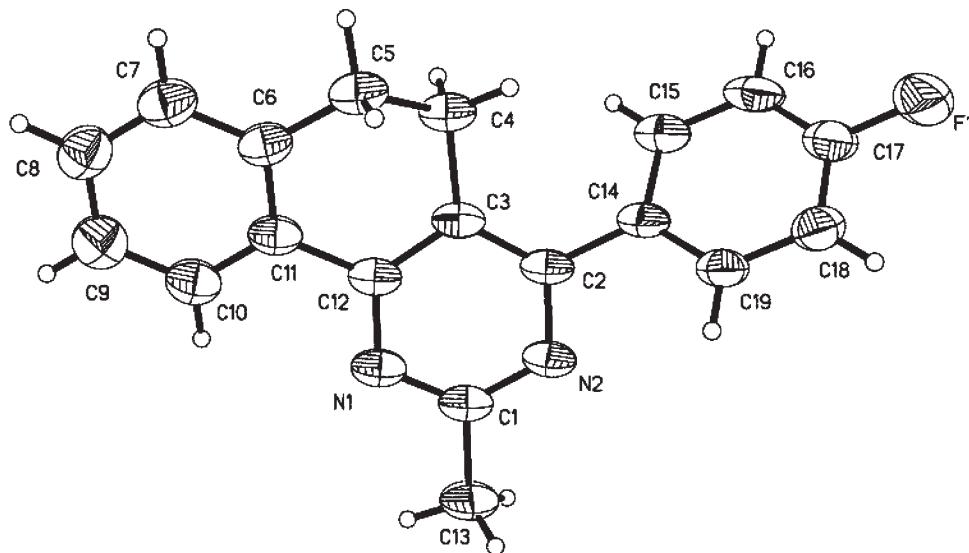


Figure 1. Structure of compound 6d.

(3H, t,  $J = 6.8$  Hz,  $J = 7.6$  Hz, ArH), 7.41 (1H, t,  $J = 7.2$  Hz,  $J = 7.2$  Hz, ArH), 7.49 (3H, t,  $J = 5.6$  Hz,  $J = 7.6$  Hz, ArH), 8.18 (1H,  $J = 7.6$  Hz, ArH), 11.84 (1H, s, NH); HRMS  $m/z$  calculated for  $C_{19}H_{16}N_2O$  [M+H]: 289.1341, found: 289.1342.

**5,6-Dihydro-4-(3,4-dimethylphenyl)benzo[h]quinazolin-2(1H)-one (4c).** m.p. 277–279°C; IR (KBr, v,  $\text{cm}^{-1}$ ): 3330, 3229, 3090, 3001, 2931, 2886, 2831, 1687, 1488, 1454, 1384, 1362, 1314, 1297, 1278, 1261, 1230, 1204, 1178, 1158, 1124, 1090, 1047, 1026, 998, 942, 891, 773, 735, 725, 639, 608  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ) ( $\delta$ , ppm): 2.19 (3H, s,  $\text{CH}_3$ ), 2.20 (3H, s,  $\text{CH}_3$ ), 2.58 (2H, t,  $J = 7.2$  Hz,  $J = 8.0$  Hz,  $\text{CH}_2$ ), 2.68 (2H, t,  $J = 7.2$  Hz,  $J = 8.0$  Hz,  $\text{CH}_2$ ), 7.03 (1H, d,  $J = 7.6$  Hz, ArH), 7.09 (1H, d,  $J = 8.6$  Hz, ArH), 7.16–7.21 (3H, m, ArH), 7.57 (1H, d,  $J = 6.8$  Hz, ArH), 8.50 (1H, s, ArH), 11.82 (1H, s, NH); HRMS  $m/z$  calculated for  $C_{20}H_{18}N_2O$  [M+H]: 303.1497, found: 303.1496.

**5,6-Dihydro-4-(3,4-dimethoxyphenyl)benzo[h]quinazolin-2(1H)-one (4d).** m.p. 240–241°C; IR (KBr, v,  $\text{cm}^{-1}$ ): 3465, 3213, 2938, 2836, 1634, 1538, 1510, 1424, 1372, 1261, 1143, 1024, 853, 780, 765, 613  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ) ( $\delta$ , ppm): 2.75 (2H, t,  $J = 2.8$  Hz,  $J = 4.0$  Hz,  $\text{CH}_2$ ), 2.80 (2H, t,  $J = 2.8$  Hz,  $J = 4.0$  Hz,  $\text{CH}_2$ ), 3.82 (3H, s,  $\text{OCH}_3$ ), 3.83 (3H, s,  $\text{OCH}_3$ ), 7.09 (1H, d,  $J = 8.4$  Hz, ArH), 7.14 (1H, d,  $J = 8.4$  Hz, ArH), 7.19 (1H, d,  $J = 1.6$  Hz, ArH), 7.33 (1H, d,  $J = 7.2$  Hz, ArH), 7.40 (1H, t,  $J = 7.2$  Hz,  $J = 7.2$  Hz, ArH), 7.47 (1H, t,  $J = 7.2$  Hz,  $J = 7.2$  Hz, ArH), 8.17 (1H, d,  $J = 7.6$  Hz, ArH), 11.81 (1H, s, NH); HRMS  $m/z$  calculated for  $C_{20}H_{18}N_2O_3$  [M+H]: 335.1396, found: 335.1393.

**5,6-Dihydro-4-(4-fluorophenyl)benzo[h]quinazolin-2(1H)-one (4e).** m.p. 270–273°C; IR (KBr, v,  $\text{cm}^{-1}$ ): 3334, 3068, 3009, 2944, 2901, 2835, 1628, 1587, 1506, 1467, 1426, 1376, 1228, 1146, 1062, 844, 762  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ) ( $\delta$ , ppm): 2.69 (2H, t,  $J = 6.4$  Hz,  $J = 6.4$  Hz,  $\text{CH}_2$ ), 2.80 (2H, t,  $J = 6.4$  Hz,  $J = 6.4$  Hz,  $\text{CH}_2$ ), 7.33–7.43 (4H, m, ArH), 7.48 (1H, d,  $J = 7.2$  Hz, ArH), 7.66 (2H, t,  $J = 5.6$  Hz,  $J = 7.2$  Hz, ArH), 8.18 (1H, d,  $J = 7.2$  Hz, ArH), 11.89 (1H,

s, NH); HRMS  $m/z$  calculated for  $C_{18}H_{13}FN_2O$  [M+H]: 293.1090, found: 293.1089.

**4-(4-Bromophenyl)-5,6-dihydrobenzo[h]quinazolin-2(1H)-one (4f).** m.p. 252–256°C; IR (KBr, v,  $\text{cm}^{-1}$ ): 3305, 3087, 2936, 2819, 1643, 1465, 1429, 1372, 1182, 1010, 836, 738  $\text{cm}^{-1}$ ; 2.68 (2H, t,  $J = 6.4$  Hz,  $J = 6.4$  Hz,  $\text{CH}_2$ ), 2.81 (2H, t,  $J = 6.4$  Hz,  $J = 6.4$  Hz,  $\text{CH}_2$ ), 7.34 (1H, d,  $J = 7.8$  Hz, ArH), 7.41 (1H, t,  $J = 7.2$  Hz,  $J = 7.2$  Hz, ArH), 7.49 (1H, t,  $J = 7.2$  Hz,  $J = 7.2$  Hz, ArH), 7.55 (2H, d,  $J = 7.8$  Hz, ArH), 7.74 (2H, d,  $J = 8.0$  Hz, ArH), 8.18 (1H, d,  $J = 7.2$  Hz, ArH), 1.91 (1H, s, NH); HRMS  $m/z$  calculated for  $C_{18}H_{13}BrN_2O$  [M+H]: 353.0290, found: 353.0278.

**4-(2-Chlorophenyl)-5,6-dihydrobenzo[h]quinazolin-2(1H)-one (4g).** m.p. > 290°C; IR (KBr, v,  $\text{cm}^{-1}$ ): 3322, 3238, 3103, 2945, 2893, 2833, 1683, 1596, 1483, 1328, 1276, 1162, 1046, 822, 756  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ) ( $\delta$ , ppm): 2.57 (2H, t,  $J = 6.4$  Hz,  $J = 6.4$  Hz,  $\text{CH}_2$ ), 2.69 (2H, t,  $J = 6.4$  Hz,  $J = 6.4$  Hz,  $\text{CH}_2$ ), 7.15 (1H, s, ArH), 7.27 (2H, s, ArH), 7.36 (2H, d,  $J = 3.2$  Hz, ArH), 7.42 (1H, s, ArH), 7.99 (2H, d,  $J = 6.4$  Hz, ArH), 11.89 (1H, s, NH); HRMS  $m/z$  calculated for  $C_{18}H_{13}ClN_2O$  [M+H]: 309.0795, found: 309.0778.

**4-(4-Chlorophenyl)-5,6-dihydrobenzo[h]quinazolin-2(1H)-one (4h).** m.p. 287–289°C; IR (KBr, v,  $\text{cm}^{-1}$ ): 3312, 3071, 3021, 2967, 2848, 1638, 1464, 1403, 1372, 1231, 1146, 1089, 1059, 888, 737  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ) ( $\delta$ , ppm): 2.70 (2H, t,  $J = 6.0$  Hz,  $J = 6.4$  Hz,  $\text{CH}_2$ ), 2.82 (2H, t,  $J = 6.4$  Hz,  $J = 7.6$  Hz,  $\text{CH}_2$ ), 7.34 (1H, d,  $J = 7.2$  Hz, ArH), 7.41 (1H, t,  $J = 7.2$  Hz,  $J = 7.6$  Hz, ArH), 7.49 (1H, t,  $J = 7.2$  Hz,  $J = 7.2$  Hz, ArH), 7.62 (4H, dd,  $J = 8.8$  Hz,  $J = 8.8$  Hz, ArH), 8.18 (1H, d,  $J = 7.6$  Hz, ArH), 11.90 (1H, s, NH); HRMS  $m/z$  calculated for  $C_{18}H_{13}ClN_2O$  [M+H]: 309.0795, found: 309.0791.

**4-(2,4-Dichlorophenyl)-5,6-dihydrobenzo[h]quinazolin-2(1H)-one (4i).** m.p. 286–288°C; IR (KBr, v,  $\text{cm}^{-1}$ ): 3411, 3043, 2935, 2836, 1635, 1466, 1429, 1376, 1316, 1150, 1100, 1046, 847, 762  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ) ( $\delta$ , ppm): 2.40 (2H, t,  $J = 6.8$  Hz,  $J = 6.8$  Hz,  $\text{CH}_2$ ), 2.83 (2H, t,  $J = 6.8$  Hz,  $J = 6.8$  Hz,

$\text{CH}_2$ ), 7.34 (1H, d,  $J = 7.2$  Hz, ArH), 7.42 (1H, t,  $J = 7.2$  Hz,  $J = 7.6$  Hz, ArH), 7.50 (1H, t,  $J = 6.4$  Hz,  $J = 7.6$  Hz, ArH), 7.58–7.64 (2H, m, ArH), 7.80 (1H, br, ArH), 8.20 (1H, d,  $J = 7.6$  Hz, ArH), 12.02 (1H, s, NH); HRMS  $m/z$  calculated for  $\text{C}_{18}\text{H}_{12}\text{Cl}_2\text{N}_2\text{O}$  [M+H]: 343.0405, found: 343.0413.

**4-(3,4-Dichlorophenyl)-5,6-dihydrobenzo[*h*]quinazolin-2(1*H*)-one (4j).** m.p. > 290°C; IR (KBr, v,  $\text{cm}^{-1}$ ): 3378, 3069, 2894, 2843, 2737, 1743, 1600, 1469, 1393, 1199, 1065, 873, 738  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ) ( $\delta$ , ppm): 2.72 (2H, t,  $J = 7.6$  Hz,  $J = 8.8$  Hz,  $\text{CH}_2$ ), 2.82 (2H, t,  $J = 7.6$  Hz,  $J = 5.6$  Hz,  $\text{CH}_2$ ), 7.35 (1H, d,  $J = 7.6$  Hz, ArH), 7.42 (1H, t,  $J = 7.2$  Hz,  $J = 7.6$  Hz, ArH), 7.50 (1H, t,  $J = 6.8$  Hz,  $J = 7.2$  Hz, ArH), 7.61 (1H, d,  $J = 7.2$  Hz, ArH), 7.81 (1H, t,  $J = 4.0$  Hz,  $J = 4.0$  Hz, ArH), 7.79 (1H, s, ArH), 8.18 (1H, d,  $J = 7.6$  Hz, ArH), 11.93 (1H, s, NH); HRMS  $m/z$  calculated for  $\text{C}_{18}\text{H}_{12}\text{Cl}_2\text{N}_2\text{O}$  [M+H]: 343.0405, found: 343.0406.

**5,6-Dihydro-2-methyl-4-p-tolylbenzo[*h*]quinazoline (6a).** m.p. 104–105°C; IR (KBr, v,  $\text{cm}^{-1}$ ): 3029, 2941, 2896, 2834, 1605, 1585, 1539, 1429, 1410, 1376, 1318, 1227, 1185, 1157, 1115, 1017, 894, 837, 805, 756, 725, 651, 571  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ) ( $\delta$ , ppm): 2.39 (3H, s,  $\text{CH}_3$ ), 2.67 (3H, s,  $\text{CH}_3$ ), 2.81 (2H, t,  $J = 6.4$  Hz,  $J = 7.6$  Hz,  $\text{CH}_2$ ), 2.94 (2H, t,  $J = 8.0$  Hz,  $J = 6.4$  Hz,  $\text{CH}_2$ ), 7.32 (3H, d,  $J = 8$  Hz, ArH), 7.38–7.46 (2H, m, ArH), 7.52–7.54 (2H, d,  $J = 8.0$  Hz, ArH), 8.27 (1H, d,  $J = 6.8$  Hz, ArH); HRMS  $m/z$  calculated for  $\text{C}_{20}\text{H}_{18}\text{N}_2$  [M+H]: 287.1548, found: 287.1549.

**5,6-Dihydro-4-(4-methoxyphenyl)-2-methylbenzo[*h*]quinazoline (6b).** m.p. 98–99°C; IR (KBr, v,  $\text{cm}^{-1}$ ): 3040, 2980, 2934, 2843, 1606, 1579, 1540, 1508, 1441, 1419, 1375, 1302, 1248, 1174, 1112, 1027, 847, 772, 759, 750, 729, 587  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ) ( $\delta$ , ppm): 2.67 (3H, s,  $\text{CH}_3$ ), 2.82 (2H, t,  $J = 6.4$  Hz,  $J = 8.0$  Hz,  $\text{CH}_2$ ), 2.98 (2H, t,  $J = 7.6$  Hz,  $J = 6.4$  Hz,  $\text{CH}_2$ ), 3.83 (3H, s,  $\text{OCH}_3$ ), 7.07 (2H, d,  $J = 8.8$  Hz, ArH), 7.33 (1H, d,  $J = 7.2$  Hz, ArH), 7.38–7.47 (2H, m, ArH), 7.62 (2H, d,  $J = 8.4$  Hz, ArH), 8.26 (1H, d,  $J = 7.2$  Hz, ArH); HRMS  $m/z$  calculated for  $\text{C}_{20}\text{H}_{18}\text{N}_3\text{O}$  [M+H]: 303.1497, found: 303.1494.

**5,6-Dihydro-4-(3,4-dimethoxyphenyl)-2-methylbenzo[*h*]quinazoline (6c).** m.p. 161–163°C; IR (KBr, v,  $\text{cm}^{-1}$ ): 3074, 2959, 2936, 2837, 1603, 1541, 1513, 1464, 1442, 1407, 1386, 1318, 1257, 1173, 1138, 1102, 877, 806, 762, 729, 680, 609  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ) ( $\delta$ , ppm): 2.68 (3H, s,  $\text{CH}_3$ ), 2.83 (2H, t,  $J = 6.4$  Hz,  $J = 7.6$  Hz,  $\text{CH}_2$ ), 2.99 (2H, t,  $J = 7.6$  Hz,  $J = 6.4$  Hz,  $\text{CH}_2$ ), 3.81 (3H, s,  $\text{OCH}_3$ ), 3.84 (3H, s,  $\text{OCH}_3$ ), 7.08 (1H, d,  $J = 8.0$  Hz, ArH), 7.20 (1H, d,  $J = 8.4$  Hz, ArH), 7.24 (1H, s, ArH), 7.33 (1H, d,  $J = 6.8$  Hz, ArH), 7.39–7.47 (2H, m, ArH), 8.26 (1H, d,  $J = 7.6$  Hz, ArH); HRMS  $m/z$  calculated for  $\text{C}_{21}\text{H}_{20}\text{N}_2\text{O}_2$  [M+H]: 333.1603, found: 333.1602.

**4-(4-Fluorophenyl)-5,6-dihydro-2-methylbenzo[*h*]quinazoline (6d).** m.p. 127–128°C; IR (KBr, v,  $\text{cm}^{-1}$ ): 3058, 2962, 2935, 2840, 1604, 1542, 1413, 1377, 1322, 1295, 1179, 1222, 1156, 1099, 1012, 895, 847, 812, 759, 749, 728, 665, 576  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ) ( $\delta$ , ppm): 2.68 (3H, s,  $\text{CH}_3$ ), 2.84 (2H, t,  $J = 6.4$  Hz,  $J = 7.6$  Hz,  $\text{CH}_2$ ), 2.95 (2H, t,  $J = 6.4$  Hz,  $J = 7.6$  Hz,  $\text{CH}_2$ ), 7.32–7.38 (3H, m, ArH), 7.39–7.46 (2H, m, ArH), 7.69–7.73 (2H, dd,  $J = 5.6$  Hz,  $J = 5.6$  Hz, ArH), 8.28 (1H, d,  $J = 7.2$  Hz, ArH); HRMS  $m/z$  calculated for  $\text{C}_{19}\text{H}_{15}\text{FN}_2$  [M+H]: 291.1298, found: 291.1296.

**4-(3-Chlorophenyl)-5,6-dihydro-2-methylbenzo[*h*]quinazoline (6e).** m.p. 97–98°C; IR (KBr, v,  $\text{cm}^{-1}$ ): 3020, 2939, 2898,

2839, 1604, 1585, 1572, 1478, 1441, 1369, 1321, 1228, 1184, 1079, 930, 892, 786, 762, 738, 722, 697, 657, 626  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ) ( $\delta$ , ppm): 2.69 (3H, s,  $\text{CH}_3$ ), 2.84 (2H, t,  $J = 6.4$  Hz,  $J = 7.2$  Hz,  $\text{CH}_2$ ), 2.94 (2H, t,  $J = 8.0$  Hz,  $J = 6.4$  Hz,  $\text{CH}_2$ ), 7.33 (1H, d,  $J = 7.2$  Hz, ArH), 7.40–7.48 (2H, m, ArH), 7.53–7.61 (3H, m, ArH), 7.69 (1H, s, ArH), 8.28 (1H, d,  $J = 7.2$  Hz, ArH); HRMS  $m/z$  calculated for  $\text{C}_{19}\text{H}_{15}\text{ClN}_2$  [M+H]: 307.1002, found: 307.1000.

**4-(4-Chlorophenyl)-5,6-dihydro-2-methylbenzo[*h*]quinazoline (6f).** m.p. 120–121°C; IR (KBr, v,  $\text{cm}^{-1}$ ): 3044, 2940, 2901, 2837, 1595, 1587, 1574, 1541, 1490, 1431, 1413, 1373, 1317, 1275, 1225, 1183, 1157, 1110, 1091, 1036, 1012, 956, 919, 893, 874, 849, 830, 761, 732, 710, 647  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 Hz, DMSO- $d_6$ ) ( $\delta$ , ppm): 2.51 (3H, s,  $\text{CH}_3$ ), 2.84 (2H, t,  $J = 6.4$  Hz,  $J = 7.2$  Hz,  $\text{CH}_2$ ), 2.99 (2H, t,  $J = 6.4$  Hz,  $J = 7.2$  Hz,  $\text{CH}_2$ ), 7.34 (1H, d,  $J = 7.2$  Hz, ArH), 7.40–7.45 (2H, m, ArH), 7.60 (2H, d,  $J = 8.4$  Hz, ArH), 7.68 (2H, d,  $J = 8.4$  Hz, ArH), 8.28 (1H, d,  $J = 7.6$  Hz, ArH); HRMS  $m/z$  calculated for  $\text{C}_{19}\text{H}_{15}\text{ClN}_2$  [M+H]: 307.1002, found: 307.1001.

**4-(3,4-Dichlorophenyl)-5,6-dihydro-2-methylbenzo[*h*]quinazoline (6g).** m.p. 98–101°C; IR (KBr, v,  $\text{cm}^{-1}$ ): 3084, 2939, 2899, 2836, 1603, 1586, 1540, 1470, 1431, 1411, 1363, 1315, 1222, 1183, 1133, 1021, 933, 905, 839, 763, 742, 728, 677, 641  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ) ( $\delta$ , ppm): 2.69 (3H, s,  $\text{CH}_3$ ), 2.84 (2H, t,  $J = 6.4$  Hz,  $J = 7.8$  Hz,  $\text{CH}_2$ ), 2.95 (2H, t,  $J = 7.6$  Hz,  $J = 6.0$  Hz,  $\text{CH}_2$ ), 7.33 (1H, d,  $J = 7.2$  Hz, ArH), 7.39 ~ 7.48 (2H, m, ArH), 7.63 (1H, dd,  $J = 2.0$  Hz,  $J = 1.6$  Hz, ArH), 7.79 (1H, d,  $J = 8.4$  Hz, ArH), 7.89 (1H, d,  $J = 2.0$  Hz, ArH), 8.27 (1H, d,  $J = 7.2$  Hz, ArH); HRMS  $m/z$  calculated for  $\text{C}_{19}\text{H}_{14}\text{N}_2\text{Cl}_2$  [M+H]: 341.0612, found: 341.0613.

**Acknowledgments.** This work was supported by the Natural Science Foundation of Jiangsu Education Department (No. 08KJB150017), PeiYu Foundation of Xuzhou Normal University (07PYL06), and the Qing Lan Project (No. 08QLT001).

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