An Efficient Synthesis of 5,6-Dihydrobenzo[*h*]quinazoline Derivatives under Solvent-Free Conditions

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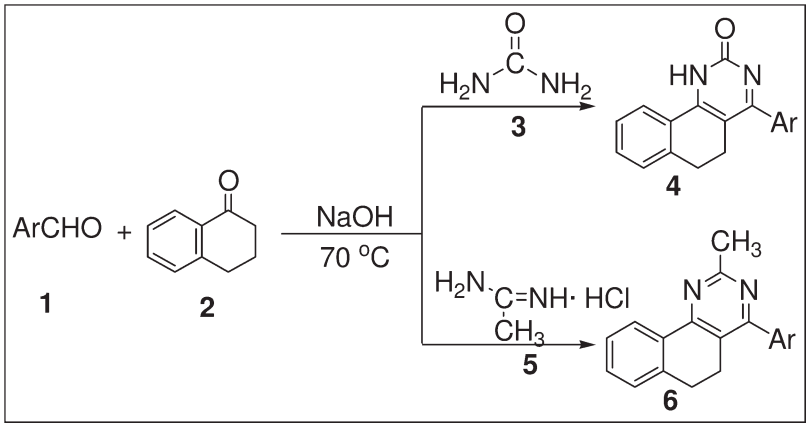

An efficient and convenient method for the preparation of 5,6-dihydrobenzo[*h*]quinazoline derivatives by the multicomponent reactions of aromatic aldehydes, 3,4-dihydronaphthalen-1(2*H*)-one and urea or acetamidine hydrochloride, in the presence of sodium hydroxide under solvent-free conditions was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

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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

Scheme 1

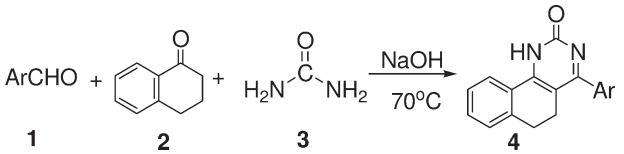


Table 1

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

Entry	Ar	Product	Yields (%)
1	C_6H_5	**4a**	80
2	4-$CH_3C_6H_4$	**4b**	85
3	3,4-$(CH_3)_2C_6H_3$	**4c**	83
4	3,4-$(CH_3O)_2C_6H_3$	**4d**	87
5	4-FC_6H_4	**4e**	90
6	4-BrC_6H_4	**4f**	88
7	2-ClC_6H_4	**4g**	80
8	4-ClC_6H_4	**4h**	91
9	2,4-$Cl_2C_6H_3$	**4i**	82
10	3,4-$Cl_2C_6H_3$	**4j**	80

Table 2

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

Entry	Ar^1	Product	Yields (%)
1	4-$CH_3C_6H_4$	**6a**	78
2	4-$CH_3OC_6H_4$	**6b**	80
3	3,4-$(CH_3O)_2C_6H_3$	**6c**	76
4	4-FC_6H_4	**6d**	85
5	3-ClC_6H_4	**6e**	79
6	4-ClC_6H_4	**6f**	89
7	3,4-$Cl_2C_6H_3$	**6g**	88

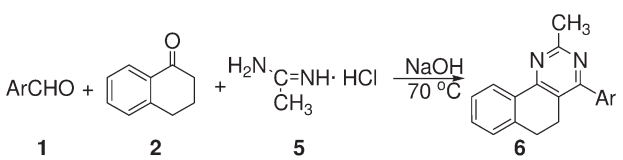
derivatives could be gained with excellent yields. The result of reaction is shown in Table 1. From Table 1 we could see the reaction was carried out smoothly and a series of 4-aryl-5,6-dihydrobenzo[*h*]quinazolin-2(1*H*)-one derivatives were obtained ignoring the properties of substitute groups on the 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 ^{1}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-dihydronaphthalen-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.

Scheme 2



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. ^{1}H NMR spectra were obtained from solution in DMSO-d_6 with Me_4Si 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(1H)-one (4a). m.p. 251–253°C; IR (KBr, n, cm^{-1}): 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^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 2.59 (2H, t, J = 7.6 Hz, J = 7.6 Hz, CH_2), 2.69 (2H, t, J = 7.2 Hz, J = 7.2 Hz, CH_2), 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 $C_{18}H_{14}N_2O$ [M+H]: 275.1184, found: 275.1185.

5,6-Dihydro-4-p-tolylbenzo[h]quinazolin-2(1H)-one (4b). m.p. 243–244°C; IR (KBr, n, cm^{-1}): 3462, 3275, 3062, 3000, 2909, 2859, 1667, 1595, 1509, 1465, 1427, 1375, 1323, 1231, 1150, 1091, 1064, 824, 759, 733 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 2.39 (3H, s, CH_3), 2.67 (2H, t, J = 6.0 Hz, J = 7.6 Hz, CH_2), 2.80 (2H, t, J = 6.4 Hz, J = 7.2 Hz, CH_2), 7.37

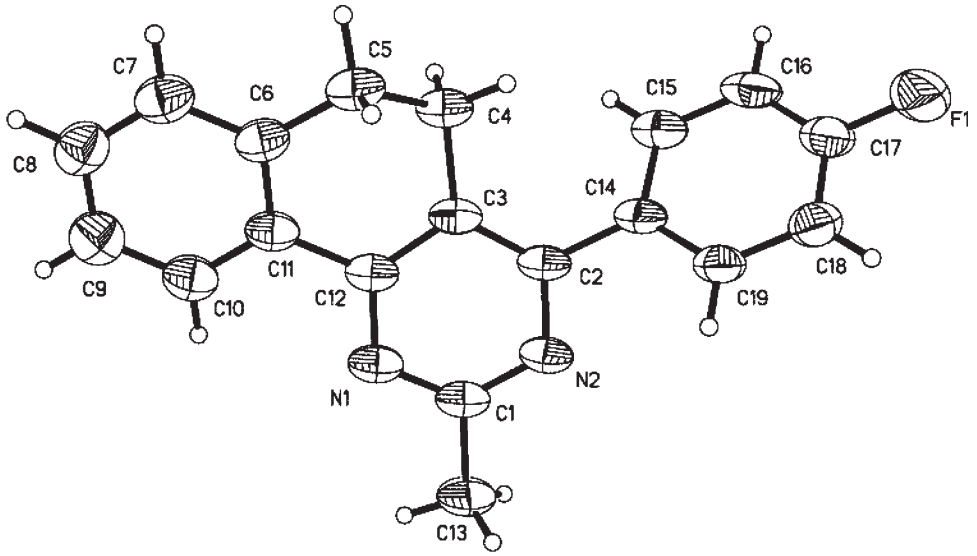


Figure 1. Structure of compound **6d**.

(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, ν, 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 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 2.19 (3H, s, CH_3), 2.20 (3H, s, CH_3), 2.58 (2H, t, J = 7.2 Hz, J = 8.0 Hz, CH_2), 2.68 (2H, t, J = 7.2 Hz, J = 8.0 Hz, 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, ν, cm^{-1}): 3465, 3213, 2938, 2836, 1634, 1538, 1510, 1424, 1372, 1261, 1143, 1024, 853, 780, 765, 613 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 2.75 (2H, t, J = 2.8 Hz, J = 4.0 Hz, CH_2), 2.80 (2H, t, J = 2.8 Hz, J = 4.0 Hz, CH_2), 3.82 (3H, s, OCH_3), 3.83 (3H, s, 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, ν, cm^{-1}): 3334, 3068, 3009, 2944, 2901, 2835, 1628, 1587, 1506, 1467, 1426, 1376, 1228, 1146, 1062, 844, 762 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 2.69 (2H, t, J = 6.4 Hz, J = 6.4 Hz, CH_2), 2.80 (2H, t, J = 6.4 Hz, J = 6.4 Hz, 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, ν, cm^{-1}): 3305, 3087, 2936, 2819, 1643, 1465, 1429, 1372, 1182, 1010,836,738 cm^{-1}; 2.68 (2H, t, J = 6.4 Hz, J = 6.4 Hz, CH_2), 2.81 (2H, t, J = 6.4 Hz, J = 6.4 Hz, 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, ν, cm^{-1}): 3322, 3238, 3103, 2945, 2893, 2833, 1683, 1596, 1483, 1328, 1276, 1162, 1046, 822, 756 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 2.57 (2H, t, J = 6.4 Hz, J = 6.4 Hz, CH_2), 2.69 (2H, t, J = 6.4 Hz, J = 6.4 Hz, 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, ν, cm^{-1}): 3312, 3071, 3021, 2967, 2848, 1638, 1464, 1403, 1372, 1231, 1146, 1089, 1059, 888, 737 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 2.70 (2H, t, J = 6.0 Hz, J = 6.4 Hz, CH_2), 2.82 (2H, t, J = 6.4 Hz, J = 7.6 Hz, 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, ν, cm^{-1}): 3411, 3043, 2935, 2836, 1635, 1466, 1429, 1376, 1316, 1150, 1100, 1046, 847, 762 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 2.40 (2H, t, J = 6.8 Hz, J = 6.8 Hz, CH_2), 2.83 (2H, t, J = 6.8 Hz, J = 6.8 Hz,

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 $C_{18}H_{12}Cl_2N_2O$ [M+H]: 343.0405, found: 343.0413.

4-(3,4-Dichlorophenyl)-5,6-dihydrobenzo[h]quinazolin-2(1H)-one (4j). m.p. > 290°C; IR (KBr, ν, cm^{-1}): 3378, 3069, 2894, 2843, 2737, 1743, 1600, 1469, 1393, 1199, 1065, 873, 738 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) (δ, ppm): 2.72 (2H, t, J = 7.6 Hz, J = 8.8 Hz, CH_2), 2.82 (2H, t, J = 7.6 Hz, J = 5.6 Hz, 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 $C_{18}H_{12}Cl_2N_2O$ [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, ν, 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 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) (δ, ppm): 2.39 (3H, s, CH_3), 2.67(3H, s, CH_3), 2.81 (2H, t, J = 6.4 Hz, J = 7.6 Hz, CH_2), 2.94 (2H, t, J = 8.0 Hz, J = 6.4 Hz, 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 $C_{20}H_{18}N_2$ [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, ν, cm^{-1}): 3040, 2980, 2934, 2843, 1606, 1579, 1540, 1508, 1441, 1419, 1375, 1302, 1248, 1174, 1112, 1027, 847, 772, 759, 750, 729, 587 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) (δ, ppm): 2.67 (3H, s, CH_3), 2.82 (2H, t, J = 6.4 Hz, J = 8.0 Hz, CH_2), 2.98 (2H, t, J = 7.6 Hz, J =6.4 Hz, CH_2), 3.83 (3H, s, 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 $C_{20}H_{18}N_3O$ [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, ν, cm^{-1}): 3074, 2959, 2936, 2837, 1603, 1541, 1513, 1464, 1442, 1407, 1386, 1318, 1257, 1173, 1138, 1102, 877, 806, 762, 729, 680, 609 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) (δ, ppm): 2.68 (3H, s, CH_3), 2.83 (2H, t, J = 6.4 Hz, J = 7.6 Hz, CH_2), 2.99 (2H, t, J = 7.6 Hz, J = 6.4 Hz, CH_2), 3.81 (3H, s, OCH_3), 3.84 (3H, s, 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 $C_{21}H_{20}N_2O_2$ [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, ν, 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 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) (δ, ppm): 2.68 (3H, s, CH_3), 2.84 (2H, t, J = 6.4 Hz, J =7.6 Hz, CH_2), 2.95 (2H, t, J = 6.4 Hz, J =7.6 Hz, 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 $C_{19}H_{15}FN_2$ [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, ν, 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 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) (δ, ppm): 2.69 (3H, s, CH_3), 2.84 (2H, t, J = 6.4 Hz, J = 7.2 Hz, CH_2), 2.94 (2H, t, J = 8.0 Hz, J = 6.4 Hz, 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 $C_{19}H_{15}ClN_2$ [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, ν, 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 cm^{-1}; ^1H NMR (400 Hz, DMSO-d_6), (d, ppm): 2.51 (3H, s, CH_3), 2.84 (2H, t, J = 6.4 Hz, J = 7.2 Hz, CH_2), 2.99 (2H, t, J = 6.4 Hz, J = 7.2 Hz, 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 $C_{19}H_{15}ClN_2$ [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, ν, 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 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) (δ, ppm): 2.69 (3H, s, CH_3), 2.84 (2H, t, J = 6.4 Hz, J = 7.8 Hz, CH_2), 2.95 (2H, t, J = 7.6 Hz, J = 6.0 Hz, 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 $C_{19}H_{14}N_2Cl_2$ [M+H]: 341.0612, found: 341.0613.

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[12] X-ray crystallography for **5:** Empirical formula $C_{19}H_{15}FN_2$, $F_w = 290.33$, $T = 298(2)$ K, monoclinic, space group *p 2(1)/c*, $a = 12.6454$ (13) Å, $b = 15.214$ (2) Å, $c = 7.7432$ (10) Å, $\alpha = 90^{\circ}$, $\beta = 90.952(2)^{\circ}$, $\gamma = 90^{\circ\circ}$, $V = 1489.5$ (3) Å^3, $Z = 4$, $D_c = 1.295$ Mg/m^3, λ (MoKα) = 0.71073 Å, $\mu = 0.086$ mm^{-1}, $F(000) = 608$. $1.61^{\circ} < \theta < 25.00^{\circ}$, $R = 0.0536$ $R_w = 0.1424$. $S = 1.118$, largest diff. peak and hole: 0.141 and -0.146 e. Å^{-3}.