ISSN 1070-3632, Russian Journal of General Chemistry, 2015, Vol. 85, No. 8, pp. 1959–1964. © Pleiades Publishing, Ltd., 2015.

3,3′-(Butane-1,4-diyl)bis(1,2-dimethyl-1*H*-imidazol-3-ium) Dibromide [BDBIm]Br—An Efficient Reusable Ionic Liquid for the Microwave-Assisted Synthesis of Quinazolinones[1]

M. Nikpassand[a], L. Zare Fekri[b], K. F. Sina[a], S. Z. Abed[a], and O. Marvi[b]

[a] *Department of Chemistry, Rasht Branch, Islamic Azad University, Rasht, Iran*
email: nikpassand@iaurasht.ac.ir

[b] *Department of Chemistry, Payame Noor University, PO Box 19395-3697 Tehran, Iran*

Received April 28, 2015

Abstract—A series of 2-substituted quinazolin-4(1*H*)-ones have been synthesized in excellent yields and short reaction times by one-pot reaction of isatoic anhydride with ammonium acetate and aromatic aldehydes under microwave irradiation. The reaction was efficiently promoted by 0.5 mmol of 3,3′-(butane-1,4-diyl)bis(1,2-dimethyl-1*H*-imidazol-3-ium) dibromide [BDBIm]Br, and the catalyst could be recovered easily and reused without appreciable loss of reactivity.

Keywords: quinazolinone, ionic liquid, microwave

DOI: 10.1134/S1070363215080265

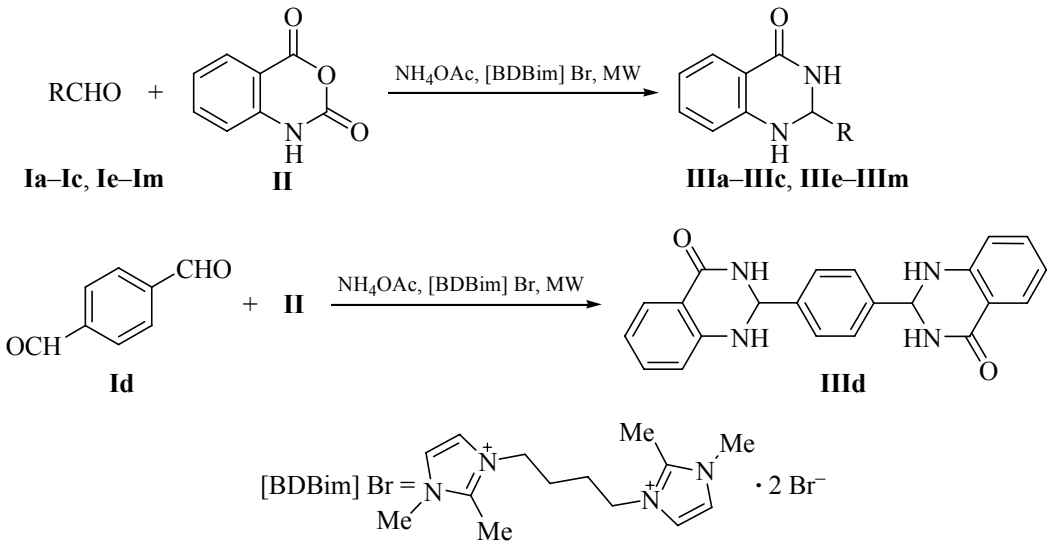
Dihydroquinazolinone derivatives constitute an important class of fused heterocyclic compounds that display a wide range of biological, pharmacological, and medicinal properties, including antitumor, antibiotic, antipyretic, analgesic, antihypertensive, diuretic, antihistamine, antidepressant, and vasodilator activity [1].

A number of synthetic methods to prepare these compounds have been described in the past few years. A typical procedure for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones involves the condensation of anthranilamide with an aldehyde or ketone using *p*-toluenesulfonic acids as catalyst under severe conditions [2]. In 2002, Su reported a method to prepare 2,3-dihydroquinazolin-4(1*H*)-ones by reductive cyclization of *o*-nitrobenzamide or *o*-azido-benzamide with aldehydes and ketones using metallic samarium in the presence of iodine or SmI_2 [3]. A recent report described the preparation of 2,3-dihydroquinazolin-4(1*H*)-ones by the reductive desulfurization of 2-thioxo-3*H*-quinazolin-4-ones with nickel boride in dry methanol [4]. Shi reported the synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones by the novel reductive cyclization of *o*-nitrobenzamides and orthoformate, aldehydes, or ketones with the aid of a low-valent titanium reagent [5]. Recently, Kurth reported a one-pot conversion of *N*-aryl-2-nitrobenzamides to 2,3-dihydroquinazolin-4(1*H*)-ones using $SnCl_2$ [6]. Nowadays, new one-pot syntheses of these compounds using *p*-toluenesulfonic acid, silica sulfuric acid, alum, Montmorillonite K-10, [bmim]BF_4, $Al(H_2PO_4)_3$, zinc perfluorooctanoate, gallium triflate, and Amberlyst-15 under microwaves [7, 8] were reported.

These methods suffer from disadvantages such as strongly acidic conditions, long reaction time, high temperature, poor selectivity, expensive reagents, toxicity, and need for excess amounts of reagents. Catalysts help synthetic chemists to avoid using strong acids or bases and other corrosive media and replace hazardous or expensive reagents by safer and economic ones. The use of ionic liquids as reaction medium and catalyst can offer a solution to solvent emission and catalyst recycle problems. Ionic liquids are advantageous due to negligible vapor pressure, non-flammability, immiscibility with nonpolar solvents, reasonable thermal and chemical stability, and recyclability [9–11]. They dissolve many organic and inorganic substrates and are tunable to specific chemical tasks [12]. Recently, ionic liquids have been successfully employed as solvents with catalytic activity for a variety of reactions [13].

[1] The text was submitted by the authors in English.

Scheme 1.



R = 3-$O_2NC_6H_4$ (**a**), 2-ClC_6H_4 (**b**), naphthalen-1-yl (**c**), 4-BrC_6H_4 (**e**), 4-ClC_6H_4 (**f**), 4-MeC_6H_4 (**g**), 4-$MeOC_6H_4$ (**h**), 4-HOC_6H_4 (**i**), 4-$Me_2NC_6H_4$ (**j**), Ph (**k**), furan-2-yl (**l**), pyridin-2-yl (**m**).

In continuation of our studies on the synthesis of pharmaceutical compounds [14], herein we describe a fast, convenient, green, and simple method for the synthesis of quinazolinones by reaction of aromatic aldehydes with isatoic anhydride and ammonium acetate in the presence of a ionic liquid, 3,3′-(butane-1,4-diyl)bis(1,2-dimethyl-1*H*-imidazol-3-ium) dibromide [BDBIm]Br, under microwave irradiation (Scheme 1). To examine the generality and scope of the described method, a wide range of functional groups in the aldehydes were investigated; the results are summarized in Table 1. It is seen that electron-withdrawing substituents on the aromatic ring of the aldehyde, such as nitro group, give higher yield in shorter reaction time than do electron-donating groups like methyl and methoxy. Recycling experiments showed that the catalyst can be recycled up to four times without significant loss of activity (Table 1).

Table 1. Microwave-assisted [BDBIm]Br-catalyzed synthesis of quinazolinones

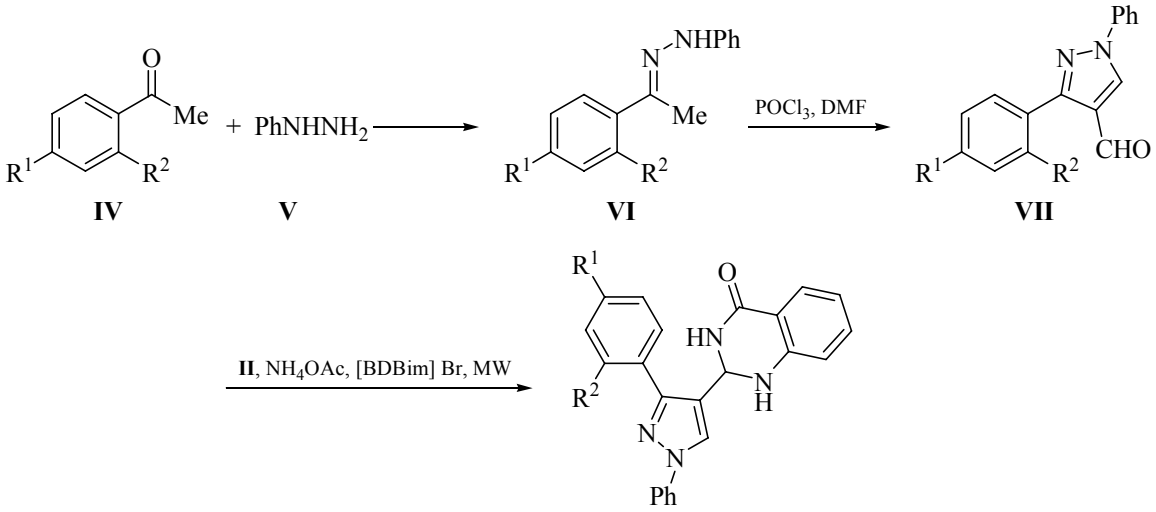
Comp. no.	Reaction time, min	Yield[a] in the first/ fourth cycle, %	Reference
IIIa	1	97/94	[8, 15]
IIIb	4	95/92	[15]
IIIc	3	99/96	–
IIId	5	97/95	–
IIIe	1	97/93	–
IIIf	1	98/97	[8, 15, 16]
IIIg	6	89/86	[8, 15, 16]
IIIh	6	87/85	[8, 15, 16]
IIIi	5	88/86	–
IIIj	10	89/87	[8, 15]
IIIk	8	86/84	[8, 15]
IIIl	6	97/95	[8]
IIIm	10	89/85	[8]

[a] Compounds **IIIa**, **IIIb**, **IIIf**–**IIIh**, and **IIIk**–**IIIm** are known, and their spectra and physical data have been reported previously.

In continuation of our efforts to synthesize heterocyclic pharmaceutical compounds, we focused on the synthesis of pyrazoles linked to quinazolinone using the developed procedure (Scheme 2, Table 2). Pyrazolyl-substituted quinazolinones **IIIn**–**IIIr** were thus obtained in high yields.

In summary, we have developed a new efficient, convenient, and simple procedure for the synthesis of quinazolinones via three-component condensation of aromatic aldehydes with isatoic anhydride and ammonium acetate. The developed procedure is advantageous due to short reaction time, mild reaction conditions, high yield, and experimental simplicity. The catalyst is nontoxic and reusable, which makes the process economic, recyclable, and environmentally benign.

Scheme 2.



R^1 = MeO, R^2 = H (**n**), R^1 = HO, R^2 = H (**o**), R^1 = Cl, R^2 = H (**p**), R^1 = H, R^2 = HO (**q**), R^1 = R^2 = H (**r**).

EXPERIMENTAL

Chemicals were purchased from Merck and Fluka and were used without additional purification. The melting points were measured on an Electrothermal 9100 apparatus and are uncorrected. The ^{1}H and ^{13}C NMR spectra were obtained on a Bruker Avance DRX 500 spectrometer (500 and 125 MHz, respectively) in DMSO-d_6 as solvent with TMS as internal standard. The IR spectra were recorded in KBr on a Shimadzu FT-IR-8400S spectrometer. The elemental analyses were obtained on a Carlo Erba EA1110 CHNO-S analyzer. All microwave-assisted reactions were performed using a Discover single- mode cavity microwave synthesizer (CEM Corp.).

3,3′-(Butane-1,4-diyl)bis(1,2-dimethyl-1*H*-imidazole-3-ium) dibromide [BDBIm]Br. A mixture of 10 mmol 1,4-dibromobutane and 20 mmol of 1,2-dimethyl-1*H*-imidazole was irradiated in a microwave furnace (180 W) for 3 × 2 min at 100°C. After completion of the reaction, the mixture was washed with diethyl ether (3 × 10 mL), and the extract was evaporated under reduced pressure. IR spectrum, ν, cm^{-1}: 3075, 2909, 1610, 1520, 1486 (C=C$_{arom}$), 1424. ^{1}H NMR spectrum, δ, ppm: 1.45 t (4H), 2.28 s (6H), 2.72 s (6H), 3.01 t (4H), 6.65 d (1H, *J* = 7.5 Hz), 6.72 d (1H, *J* = 7.5 Hz). ^{13}C NMR spectrum, δ_C, ppm: 23.2, 41.0, 52.2, 56.1, 127.9, 129.0.

2-Aryl-2,3-dihydroquinazolin-4(1*H*)-ones IIIa–IIIr *(general procedure).* A mixture of 1 mmol of the corresponding aldehyde, 1 mmol of isatoic anhydride, 1 mmol of ammonium acetate, and 0.5 mmol of [BDBIm] Br was irradiated in a microwave furnace (300 W) for a required time (Tables 1, 2). The progress of the reaction was monitored by TLC (EtOAc–petroleum ether, 1 : 4). After completion of the reaction, the mixture was extracted with $CHCl_3/H_2O$, the organic phase was separated and evaporated, and the residue was purified by recrystallization. The aqueous phase was evaporated also to regenerate [BDBIm]Br which can be reused.

2-(3-Nitrophenyl)-2,3-dihydroquinazolin-4(1*H*)-one (IIIa). Yield 97%, mp 190–192°C. IR spectrum, ν, cm^{-1}: 3358 (N–H), 3192, 3067, 2918, 1645 (C=O), 1614, 1500 (C=C$_{arom}$), 1389, 1327. ^{1}H NMR spectrum, δ, ppm: 6.10 s (1H, 2-H), 6.67 t (1H, *J* = 7.4 Hz), 6.72 d (1H, *J* = 8.1 Hz), 6.97 br.s (1H), 7.21 d (1H, *J* =

Table 2. Microwave induced [BDBIm]Br synthesis of pyrazole linked quinazolinones

Compound no.	Time, min	Yield,[a] %
IIIn	3	97
IIIo	3	92
IIIp	5	98
IIIq	7	91
IIIr	5	89

[a] Isolated yield.

7.0 Hz), 7.34–7.37 m (2H), 7.43–7.46 m (1H), 7.62 d (1H, *J* = 7.0 Hz), 8.16 br.s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 64.1 (C^2), 115.0, 115.13, 127.8, 127.9, 129.2, 130.0, 130.7, 132.3, 133.9, 138.3 (2C), 148.11, 164.1 (C=O). Found, %: C 62.36; H 4.19; N 15.52. $C_{14}H_{11}N_3O_3$. Calculated, %: C 62.45; H 4.12; N 15.61.

2-(2-Chlorophenyl)-2,3-dihydroquinazolin-4(1*H*)-one (IIIb). Yield 95%, mp 205–206°C. IR spectrum, ν, cm^{-1}: 3202 (N–H), 3190 (N–H), 3070, 2920, 1652 (C=O), 1608, 1529 (C=C$_{arom}$). ^{1}H NMR spectrum, δ, ppm: 5.96 s (1H, 2-H), 6.71 br.s (1H), 6.79 br.s (1H), 7.28 br.s (1H), 7.37 br.s (1H), 7.62 br.s (1H), 7.71 br.s (1H), 7.95 br.s (1H), 8.21 br.s (1H), 8.37 br.s (1H, NH), 8.56 br.s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 62.5 (C^2), 115.1, 115.8, 118.0, 127.2, 127.6, 130.7, 131.3, 132.0, 134.0, 137.5, 140.2, 149.7, 163.8 (C=O). Found, %: C 65.06; H 4.19; N 10.88. $C_{14}H_{11}ClN_2O$. Calculate, %: C 65.00; H 4.29; N 10.83.

2-(Naphtalen-1-yl)-2,3-dihydroquinazolin-4(1*H*)-one (IIIc). Yield 99%, mp 111–113°C. IR spectrum, ν, cm^{-1}: 3312 (N–H), 3187 (N–H), 3045, 2939, 1656 (C=O), 1605, 1512 (C=C$_{arom}$), 1370, 1321. ^{1}H NMR spectrum, δ, ppm: 5.89 s (1H, 2-H), 6.63 t (1H, *J* = 7.4 Hz), 6.72 d (1H, *J* = 8.1 Hz), 7.13 br.s (1H), 7.22 t (1H, *J* = 7.0 Hz), 7.48–7.50 m (2H), 7.58 d (1H, *J* = 7.1 Hz), 7.64 d (1H, *J* = 8.6 Hz), 7.87–7.97 m (3H), 8.30 br.s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 63.7 (C^2), 116.4, 116.9, 118.9, 123.5, 124.0, 125.0, 127.5, 129.5, 130.3, 130.6, 134.9, 135.6, 136.3, 137.0, 138.4, 146.2, 165.2 (C=O). Found, %: C 78.74; H 5.20; N 10.14. $C_{18}H_{14}N_2O$. Calculated, %: C 78.81; H 5.14; N 10.21.

2,2′-(Benzene-1,4-diyl)bis[2,3-dihydroquinazolin-4(1*H*)-one] (IIId). Yield 97%, mp 122–123°C. IR spectrum, ν, cm^{-1}: 3747 (N–H), 3446 (N–H), 3068, 2926, 1653 (C=O), 1610, 1506 (C=C$_{arom}$), 1383. ^{1}H NMR spectrum, δ, ppm: 5.69 s (1H, 2-H), 6.60 t (2H, *J* = 7.4 Hz), 6.69 d (2H, *J* = 8.0 Hz), 7.08 br.s (2H), 7.20 t (2H, *J* = 7.9 Hz), 7.43 s (2H), 7.66 d (1H, *J* = 7.1 Hz), 8.32 br.s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 62.7 (C^2), 115.8, 117.2, 127.8, 130.6, 132.2 (2C), 133.9, 138.3 (2C), 150.9, 166.3 (C=O). Found, %: C 71.39; H 4.89; N 15.09. $C_{22}H_{18}N_4O_2$. Calculated, %: C 71.34; H 4.90; N 15.13.

2-(4-Bromophenyl)-2,3-dihydroquinazolin-4(1*H*)-one (IIIe). Yield 97%, mp 171–173°C. IR spectrum, ν, cm^{-1}: 3301 (N–H), 3197 (N–H), 3067, 2912, 1651 (C=O), 1605, 1504 (C=C$_{arom}$). ^{1}H NMR spectrum, δ, ppm: 5.75 s (1H, 2-H), 6.68 t (1H, *J* = 7.2 Hz), 6.73 d (1H, *J* = 7.6 Hz), 7.14 s (1H), 7.24 t (1H, *J* = 6.8 Hz), 7.43 d (2H, *J* = 7.6 Hz), 7.58 m (2H), 8.34 s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 66.2 (C^2), 114.9, 115.3, 117.7, 122.0, 129.5, 131.6, 133.8, 141.5 (2C), 148.0, 163.9 (C=O). Found, %: C 55.36; H 3.57; N 9.22. $C_{14}H_{11}BrN_2O$. Calculated, %: C 55.47; H 3.66; N 9.24.

2-(4-Chlorophenyl)-2,3-dihydroquinazolin-4(1*H*)-one (IIIf). Yield 98%, mp 200–202°C. IR spectrum, ν, cm^{-1}: 3744 (N–H), 3304 (N–H), 3063, 2933, 1655 (C=O), 1610, 1508 (C=C$_{arom}$), 1385. ^{1}H NMR spectrum, δ, ppm: 6.35 s (1H, 2-H), 6.64 d (1H, *J* = 8.0 Hz), 6.79 t (1H, *J* = 7.5 Hz), 7.23–7.27 m (2H), 7.36 d (2H, *J* = 8.2 Hz), 7.52 d (2H, *J* = 8.2 Hz), 8.48 br.s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 66.1 (C^2), 114.9, 115.3, 117.7, 127.8, 128.7, 129.1, 129.2, 133.8, 148.1, 163.9, 188.9 (C=O). Found, %: C 65.06; H 4.24; N 10.93. $C_{14}H_{11}ClN_2O$. Calculated, %: C 65.00; H 4.29; N 10.83.

2-(4-Methylphenyl)-2,3-dihydroquinazolin-4(1*H*)-one (IIIg). Yield 89%, mp 217–218°C. IR spectrum, ν, cm^{-1}: 3310 (N–H), 3194 (N–H), 3061, 2933, 1661 (C=O), 1608, 1508 (C=C$_{arom}$), 1292. ^{1}H NMR spectrum, δ, ppm: 2.3 s (CH_3), 6.01 s (1H, 2-H), 7.09 t (1H, *J* = 7.2 Hz), 7.18 d (1H, *J* = 7.6 Hz), 7.53 t (2H, *J* = 6.8 Hz), 7.70 d (2H, *J* = 7.6 Hz), 7.83 t (1H, *J* = 7.2 Hz), 8.14 d (1H, *J* = 8.0 Hz). ^{13}C NMR spectrum, δ_C, ppm: 41.01 (CH_3), 64.8 (C^2), 115.8, 116.6, 127.0, 128.3, 129.9, 130.9, 132.7, 133.0, 139.8, 149.0, 165.6 (C=O). Found, %: C 75.67; H 5.90; N 11.76. $C_{15}H_{14}N_2O$. Calculated, %: C 75.61; H 5.92; N 11.76.

2-(4-Mehoxyphenyl)-2,3-dihydroquinazolin-4(1*H*)-one (IIIh). Yield 87%, mp 184–185°C. IR spectrum, ν, cm^{-1}: 3406 (N–H), 3191 (N–H), 3072, 1645 (C=O), 1514, 1458 (C=C$_{arom}$), 746. ^{1}H NMR spectrum, δ, ppm: 3.51 s (3H, OCH_3), 6.01 s (1H, 2-H), 6.68 t (1H, *J* = 7.2 Hz), 6.79–6.83 m (2H), 6.86 d (1H, *J* = 8.0 Hz), 7.14 t (1H, *J* = 7.2 Hz), 7.23 t (1H, *J* = 7.2 Hz), 7.34 d (1H, *J* = 7.2 Hz), 7.62 d (1H, *J* = 7.2 Hz), 7.96 s (1H, NH), 9.88 s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 53.2 (OCH_3), 64.3 (C^2), 115.6, 116.1, 118.6, 127.5, 127.9, 128.8, 130.4, 130.7, 138.5, 148.2, 166.0 (C=O). Found, %: C 69.96; H 5.11; N 11.52. $C_{14}H_{12}N_2O_2$. Calculated, %: C 69.99; H 5.03; N 11.66.

2-(4-Hydroxyphenyl)-2,3-dihydroquinazolin-4(1*H*)-one (IIIi). Yield 88%, mp 201–202°C. IR spectrum, ν, cm^{-1}: 3416 (N–H), 3267 (N–H), 3055, 1653 (C=O), 1547, 1467 (C=C$_{arom}$), 754. ^{1}H NMR spectrum, δ,

ppm: 6.23 s (1H, 2-H), 6.74 t (1H, *J* = 7.5 Hz), 6.81–6.84 m (2H), 6.89 d (1H, *J* = 7.5 Hz), 7.18 t (1H, *J* = 8.0 Hz), 7.45 t (1H, *J* =7.6 Hz), 7.76 d (1H, *J* = 7.6 Hz), 8.16 s (1H, NH), 9.88 s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 64.3 (C^2), 113.6, 115.1, 119.1, 124.3, 127.9, 128.8, 133.9, 136.8, 137.2, 147.3, 166.0 (C=O). Found, %: C 69.96; H 5.11; N 11.52. $C_{14}H_{12}N_2O_2$. Calculated, %: C 69.99; H 5.03; N 11.66.

2-[4-(Dimethylamino)phenyl]-2,3-dihydroquinazolin-4(1*H*)-one (IIIj). Yield 89%, mp 209–210°C. IR spectrum, ν, cm^{-1}: 3312 (N–H), 3195 (N–H), 3065, 1663 (C=O), 1610, 1520 (C=C$_{arom}$). ^{1}H NMR spectrum, δ, ppm: 3.35 s (6H, NCH_3), 5.63 s (1H, 2-H), 6.66 t (1H, *J* = 7.7 Hz), 6.70–6.74 m (2H), 6.92 s (1H), 7.23 t (1H, *J* = 6.8 Hz), 7.29 d (2H, *J* = 8.4 Hz), 7.59 d (1H, *J* = 8.6 Hz), 8.07 s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 34.3 (NCH_3), 64.6 (C^2), 115.2, 115.5, 117.3, 126.5, 130.2, 130.8, 132.0, 133.0, 138.4, 149.8, 166.0 (C=O). Found, %: C 71.78; H 6.51; N 15.76. $C_{16}H_{17}N_3O$. Calculated, %: C 71.89; H 6.41; N 15.72.

2-[3-(4-Methoxyphenyl)-1-phenyl-1*H*-pyrazol-4-yl]-2,3-dihydroquinazolin-4(1*H*)-one (IIIn). Yield 85%, mp 223–225°C. IR spectrum, ν, cm^{-1}: 3299 (N–H), 3213 (N–H), 3068 (C–H$_{arom}$), 2918 (C–H$_{aliph}$), 1730 (C=O), 1494, 1610 (C=C$_{arom}$), 1379 (C–N). ^{1}H NMR spectrum, δ, ppm: 2.51 s (3H, OCH_3), 5.89 s (1H, 2-H), 6.77 t (2H, *J* = 8.0 Hz), 7.07 s (1H), 7.28 d (2H, *J* = 8.0 Hz), 7.31 t (2H, *J* = 7.4 Hz), 7.54 t (2H, *J* = 8.0 Hz), 7.67–7.71 m (3H), 7.96 d (1H, *J* = 8.0 Hz), 8.30 s (1H, NH), 8.89 s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 54.4 (OCH_3), 63.2 (C^2), 115.3, 115.9, 117.1, 120.5, 122.5, 128.8, 130.0, 131.5, 133.2, 133.8, 134.0, 134.6, 138.2, 139.3, 147.3, 150.3, 155.8, 166.3 (C=O). Found, %: C 72.64; H 5.12; N 14.06. $C_{24}H_{20}N_4O_2$. Calculated, %: C 72.71; H 5.08; N 14.13.

2-[3-(4-Hydroxyphenyl)-1-phenyl-1*H*-pyrazol-4-yl]-2,3-dihydroquinazolin-4(1*H*)-one (IIIo). Yield 79%, mp 235–237°C. IR spectrum, ν, cm^{-1}: 3483 (O–H), 3369 (N–H), 1610 (C=O), 1560, 1494 (C=C$_{arom}$), 1380 (C–N), 1230 (C–O). ^{1}H NMR spectrum, δ, ppm: 5.86 s (1H, 2-H), 6.75 t (2H, *J* = 8.4 Hz), 6.85 d (2H, *J* = 8.8 Hz), 7.05 s (1H), 7.28–7.36 m (2H), 7.54 t (2H, *J* = 7.6 Hz), 7.60 d.t (2H, *J* = 8.8, 2.8 Hz), 7.64 d.d (1H, *J* = 8.4, 1.6 Hz), 7.97 d.d (2H, *J* = 7.2, 1.2 Hz), 8.29 s (1H, NH), 8.85 s (1H, NH), 9.67 s (1H, OH). ^{13}C NMR spectrum, δ-$_C$, ppm: 64.5 (C^2), 115.2, 115.8, 117.0, 119.5, 121.9, 127.9, 130.1, 132.0, 133.0, 133.3, 134.8, 136.0, 138.2, 138.8, 148.0, 151.3, 155.0, 166.5 (C=O). Found, %: C 72.18; H 4.81; N 14.68. $C_{23}H_{18}N_4O_2$. Calculated, %: C 72.24; H 4.74; N 14.65.

2-[3-(4-Chlorophenyl)-1-phenyl-1*H*-pyrazol-4-yl]-2,3-dihydroquinazolin-4(1*H*)-one (IIIp). Yield 85%, mp 201–202°C. IR spectrum, ν, cm^{-1}: 3294 (N–H), 3222 (N–H), 3074 (C–H$_{arom}$), 1652 (C=O), 1610, 1546, 1490 (C=C$_{arom}$), 1375 (C–N). ^{1}H NMR spectrum, δ, ppm: 5.93 s (1H, 2-H), 6.76–6.79 m (2H), 7.08 s (1H), 7.30 t (1H, *J* = 7.20 Hz), 7.37 t (1H, *J* = 7.6 Hz), 7.53–7.57 m (4H), 7.68 d (1H, *J* = 6.8 Hz), 7.86 d (2H, *J* = 8.2 Hz), 7.96 d (2H, *J* = 8.2 Hz), 8.32 s (1H, NH), 8.92 s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 63.9 (C^2), 115.0, 116.6, 117.3, 118.4, 120.3, 128.0, 128.9, 132.2, 133.6, 133.9, 135.7, 136.4, 137.7, 138.9, 143.3, 150.2, 153.9, 165.3 (C=O). Found, %: C 68.95; H 4.20; N 14.04. $C_{23}H_{17}ClN_4O$. Calculated, %: C 68.91; H 4.27; N 13.98.

2-[3-(2-Hydroxyphenyl)-1-phenyl-1*H*-pyrazol-4-yl)-2,3-dihydroquinazolin-4(1*H*)-one (IIIq). Yield 85%, mp 236–238°C. IR spectrum, ν, cm^{-1}: 3511 (O–H), 3379 (N–H), 2925 (C–H$_{aliph}$), 1681 (C=O), 1612, 1558, 1504 (C=C$_{arom}$), 1382 (C–N), 1247 (C–O). ^{1}H NMR, δ, ppm: 5.77 d (1H, *J* = 2.8 Hz, 2-H), 6.80 t (2H, *J* = 7.6 Hz), 6.91 s (1H), 6.93 d.t (1H, *J* = 7.2, 0.8 Hz), 6.97 d (1H, *J* = 8.0 Hz), 7.26–7.30 m (2H), 7.34 t (1H, *J* = 7.2 Hz), 7.49 d.d (1H, *J* = 7.6, 1.6 Hz), 7.51 t (2H, *J* = 7.6 Hz), 7.65 d.d (1H, *J* = 8.0, 1.6 Hz), 7.93 d (1H, *J* = 1.2 Hz), 7.50 d (2H, *J* = 0.8 Hz), 8.85 s (1H, NH), 10.06 s (1H, OH). ^{13}C NMR spectrum, δ_C, ppm: 63.9 (C^2), 114.8, 115.2, 116.9, 118.8, 120.1, 121.2, 123.0, 128.0, 130.4, 131.6, 133.4, 134.0, 134.3, 135.0, 137.2, 138.0, 147.3, 151.8, 154.6, 165.6 (C=O). Found, %: C 72.28; H 4.83; N 14.63. $C_{23}H_{18}N_4O_2$. Calculated, %: C 72.24; H 4.74; N 14.65.

2-(1,3-Diphenyl-1*H*-pyrazol-4-yl)-2,3-dihydroquinazolin-4(1*H*)-one (IIIr). Yield 87%, mp 238–240°C. IR spectrum, ν, cm^{-1}: 3257 (N–H), 3184 (N–H), 3062 (C–H$_{arom}$), 2925 (C–H$_{aliph}$), 1658 (C=O), 1614, 1550, 1502 (C=C$_{arom}$), 1375 (C–N). ^{1}H NMR spectrum, δ, ppm: 5.91 s (1H, 2-H), 6.75–6.80 m (2H), 7.09 s (1H), 7.28 t (1H, *J* = 8.4 Hz), 7.38 t (1H, *J* = 7.6 Hz), 7.42 t (1H, *J* = 7.2 Hz), 7.49 t (2H, *J* = 7.2 Hz), 7.55 t (2H, *J* = 7.6 Hz), 7.68 d (1H, *J* = 7.2 Hz), 7.81 d (2H, *J* = 7.2 Hz), 7.98 d (2H, *J* = 7.6 Hz), 8.33 s (1H, NH), 8.92 s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 64.3 (C^2), 115.3, 115.6, 117.6, 118.0, 121.3, 125.5, 127.2, 128.1, 130.2, 132.9, 133.7, 135.5, 136.9, 137.9, 138.0, 143.3, 150.3, 165.7 (C=O). Found, %: C 75.42; H 4.92; N 15.24. $C_{23}H_{18}N_4O$. Calculated, %: C 75.39; H 4.95; N 15.29.

ACKNOWLEDGMENTS

Financial support by Rasht Branch, Islamic Azad University (grant no. 4.5830) is gratefully acknowledged.

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