

Brønsted acidic ionic liquids catalyze the preparation of 2,3-dihydroquinazolin-4(1*H*)-one derivatives

Hamid Reza Shaterian · Morteza Aghakhanizadeh

Received: 20 November 2012 / Accepted: 23 January 2013
© Springer Science+Business Media Dordrecht 2013

Abstract A simple and facile method for the green synthesis of 2,3-dihydroquinazolin-4(1*H*)-one derivatives by direct three-component cyclo-condensation of isatoic anhydride, ammonium acetate (or primary amines), and arylaldehydes using different Brønsted acidic ionic liquids, for example 2-pyrrolidonium hydrogensulfate ([hnmp][HSO₄]), *N*-methyl-2-pyrrolidonium hydrogensulfate ([NMP][HSO₄]), and *N*-methyl-2-pyrrolidonium dihydrogenphosphate ([NMP][H₂PO₄]), as reusable catalysts under solvent-free conditions is described. In addition, reaction of anthranilamide and arylaldehydes for synthesis of 2,3-dihydroquinazolin-4(1*H*)-one derivatives was investigated.

Keywords 2,3-Hydroquinazolin-4(1*H*)-one · Anthranilamide · Ionic liquid · Combinatorial chemistry · Primary amine

Introduction

Multi-component reactions (MCRs) are important in combinatorial chemistry because of the ability to synthesize target compounds with greater efficiency and atom economy by generating structural complexity in a single step from three or more reactants [1]. Moreover, MCRs have the advantage of simplicity and synthetic efficiency over conventional chemical reactions [1].

Heterocyclic compounds are important because of their chemical, biological, and technical significance [2]. Heterocyclic compounds include many natural products, for example vitamins, hormones, antibiotics, alkaloids, pharmaceuticals, herbicides, and dyes [3].

H. R. Shaterian (✉) · M. Aghakhanizadeh
Department of Chemistry, Faculty of Sciences, University of Sistan and Baluchestan,
PO Box 98135-674, Zahedan, Iran
e-mail: hrshaterian@chem.usb.ac.ir

2,3-Dihydroquinazolinone derivatives have been reported to have a wide range of pharmacological activity, for example anti-inflammatory and analgesic [4]. In addition, these compounds as chemical intermediates can easily be oxidized to their corresponding quinazolin-4(3*H*)-ones [5], which are also important biologically active heterocyclic compounds [6]. Therefore, a variety of methods have been developed for synthesis of quinazolinone structures, for example cyclization of *o*-acylaminobenzamides [7], amidation of 2-aminobenzonitrile followed by oxidative ring closure [8], and solid-phase synthesis of 2-aryl-amino-substituted quinazolinones [9].

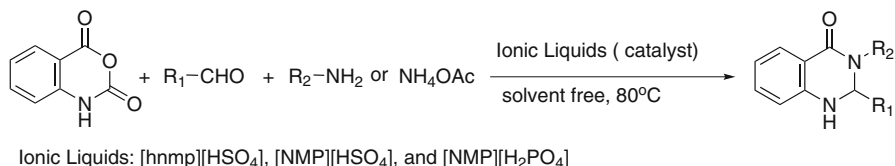
One of the most important aspects of green chemistry is the use of ionic liquids (ILs) as more environmentally benign solvents and catalysts. ILs have several advantages, for example control of product distribution, enhanced rate and/or reactivity, ease of product recovery, catalyst immobilization, and reusability [10].

Quinazolinones have been prepared by three-component reaction of isatoic anhydride, primary amine or ammonium acetate, and arylaldehydes in the presence of several catalysts, for example aluminium methanesulfonate ($\text{Al}(\text{MS})_3 \cdot 4\text{H}_2\text{O}$) [11], *p*-toluenesulfonic acid (*p*-TSA) [12], $\text{KAl}(\text{SO}_4)_2 \cdot 12\text{H}_2\text{O}$ (alum) [13], 1-methylimidazolium nitrate [Hmim][NO_3] [14], silica supported ferric chloride ($\text{SiO}_2\text{-FeCl}_3$) [15], silica-bonded *N*-propylsulfamic acid (SBNPSA) [16], acetic acid [17], copper benzenesulfonate ($\text{Cu}(\text{C}_6\text{H}_5\text{SO}_3)_2 \cdot 6\text{H}_2\text{O}$) [18], silica supported ferric chloride ($\text{SiO}_2\text{-FeCl}_3$) [15], gallium(III) triflate ($\text{Ga}(\text{OTf})_3$) [19], and strontium chloride ($\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$) [20]. They have also been prepared by condensation of anthranilamide and arylaldehydes by using CuCl_2 , [21] 1-butyl-3-methylimidazolium hexafluorophosphate ([Bmim][PF_6]) [22], tetrabutylammonium bromide (Bu_4NBr) [23], cerium(IV) ammonium nitrate (CAN) [24], zirconium (IV) chloride (ZrCl_4) [25], and $\text{H}_3\text{PW}_{12}\text{O}_{40}$ [26], as catalysts. However, some of these procedures have limitations, for example tedious process, long reaction times, high temperatures, harsh reaction conditions, expensive reagents, and low yields. Thus, the development of novel methods for synthesis of dihydroquinazolin-4(1*H*)-ones is of great importance because of the potential biological and pharmaceutical activity of these compounds.

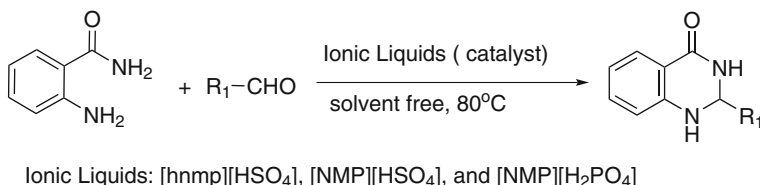
In continuation our research on new methods of organic synthesis [27–30], we developed the synthesis of 2,3-dihydroquinazolin-4(1*H*)-one derivatives by one-pot three-component condensation of isatoic anhydride, ammonium acetate (or primary amines), and arylaldehydes (Method A, Scheme 1) and by two-component reaction of anthranilamide and arylaldehydes (Method B, Scheme 2), using three Brønsted acidic ionic liquids (BAILs), 2-pyrrolidonium hydrogensulfate ([hmp][HSO_4]), *N*-methyl-2-pyrrolidonium hydrogensulfate ([NMP][HSO_4]), and *N*-methyl-2-pyrrolidonium dihydrogenphosphate ([NMP][H_2PO_4]) (Fig. 1), as catalysts.

Results and discussion

To achieve more efficient synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones, reaction of isatoic anhydride (1 mmol), ammonium acetate (1.2 mmol), and benzaldehyde (1 mmol) was selected as model system, under thermal solvent-free conditions, to find the optimum conditions. The preparation of 2-phenyl-2,3-dihydroquinazolin-4(1*H*)-



Scheme 1 Preparation of 2,3-dihydroquinazolin-4(1*H*)-one derivatives by three-component reaction of isatoic anhydride, primary amine or ammonium acetate, and arylaldehydes



Scheme 2 Preparation of 2,3-dihydroquinazolin-4(1*H*)-one derivatives by condensation of anthranilamide and arylaldehydes

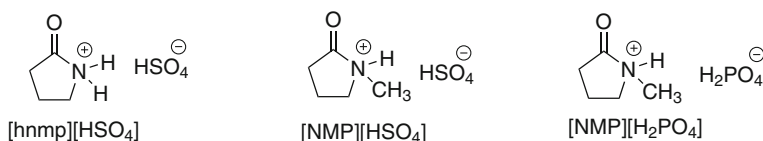


Fig. 1 The structures of [hnmp][HSO_4], [NMP][HSO_4], and [NMP][H_2PO_4]

one was studied at different reaction temperatures (25, 60, 80, 100, and 120°C) and with different amounts of acidic ionic liquids as catalyst (5, 10, 15, 20, 25, and 30 mol %) (Table 1). The best result was obtained by use of 5 mol % [hnmp][HSO_4], 5 mol % [NMP][HSO_4], or 5 mol % [NMP][H_2PO_4] at 80°C (Table 1).

By use of these optimized reaction conditions, the condensation reaction of isatoic anhydride, arylaldehydes and ammonium acetate as source of ammonia or primary amines proceeded well and afforded the desired products (Table 2) in good to excellent yields.

As shown in Table 2, arylaldehydes bearing either electron-donating or electron-withdrawing groups on the aromatic ring were investigated. The substituent on the phenyl ring did not make any difference in this reaction. We also used aliphatic aldehydes, for example propionaldehyde and butyraldehyde instead of benzaldehydes. Experimental observation showed us that all the starting reactants were intact after 12 h without formation of any by-products (Table 2, Entries 20, 21).

In continuation of our work, using these optimized reaction conditions, the scope and efficiency of the reaction were investigated for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-one derivatives by direct condensation of arylaldehydes (1 mmol), anthranilamide (1 mmol), with ILs (5 mol %) as catalysts under solvent-free conditions (Table 3).

Table 1 Optimization the amount of the acidic ionic liquids, 2-pyrrolidonium hydrogensulfate (IL₁), *N*-methyl-2-pyrrolidonium hydrogensulfate (IL₂), and *N*-methyl-2-pyrrolidonium dihydrogenphosphate (IL₃), as catalysts at different temperature in the reaction of isatoic anhydride (1 mmol), ammonium acetate (1.2 mmol), and benzaldehyde (1 mmol) under solvent-free conditions

Entry	Catalyst (mol %)	Temperature (°C)	Time (min)			Yield (%) ^a		
			IL ₁	IL ₂	IL ₃	IL ₁	IL ₂	IL ₃
1	–	100	180	180	180	22	28	19
2	5	100	9	8	9	91	89	87
3	10	100	10	9	11	93	87	90
4	15	100	11	10	13	92	90	88
5	20	100	11	10	14	88	90	85
6	25	100	25	17	19	78	75	80
7	30	100	30	21	23	80	82	75
8	5	25	180	180	180	–	–	–
9	5	60	45	43	50	57	60	70
10	5	80	11	10	13	89	87	85
11	5	100	9	8	9	91	89	87
12	5	120	7	6	8	90	92	90

^a Yields refer to isolated pure products

We also compared the results from preparation of 2,3-dihydroquinazolin-4(1*H*)-one derivatives by direct condensation of isatoic anhydride, primary amines or ammonium acetate, and arylaldehydes (Method A) and the reaction of anthranilamide and arylaldehydes (Method B) using ILs as catalysts under solvent-free conditions. Method A produces 2,3-dihydroquinazolin-4(1*H*)-ones in shorter reaction times and higher yields than method B, because isatoic anhydride is more reactive than anthranilamide. Loss of CO₂ from isatoic anhydride is an excellent driving force for the reaction and preparation of a more reactive intermediate for synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones in shorter reaction times and higher yields.

Next, we compared the results obtained by use of these ILs and other catalysts reported in the literature for preparation of 2,3-dihydroquinazolin-4(1*H*)-ones, for example Al(MS)₃·4H₂O [11], *p*-TSA [12], KAl(SO₄)₂·12H₂O [13], Cu(C₆H₅SO₃)₂·6H₂O, 18 SiO₂–FeCl [3, 15] Ga(OTf)₃ [3, 19] SrCl₂·6H₂O [20], CuCl [2, 21] [Bmim]PF₆, [22] Bu₄NBr [23], ZrCl₄, [25] and H₃PW₁₂O₄₀ [26] (Table 4). Table 4 clearly reveals that 2-pyrrolidonium hydrogensulfate, *N*-methyl-2-pyrrolidonium hydrogensulfate, and *N*-methyl-2-pyrrolidonium dihydrogenphosphate are effective catalysts in terms of reaction time and yield of product obtained compared with other reported catalysts.

On the basis of the literature [16] and our experimental results, a plausible mechanism of the three-component reaction is proposed in Scheme 3. In the presence of acidic ionic liquids as catalyst, first, the isatoic anhydride (**1**) is activated, forming intermediate **2**. The *N*-nucleophilic amine (**3**) then attacks on the carbonyl unit of **2**, producing intermediate **4**, which in turn affords the intermediate

Table 2 Preparation of 2,3-dihydroquinazolin-4(1*H*)-one derivatives (Method A)

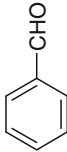
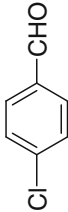
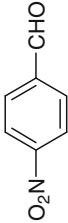
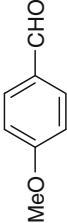
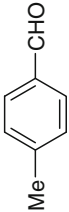
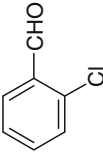
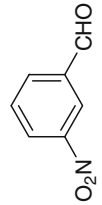
Entry	R	Amine	Time (min)			Yield (%) ^a			M.p.(°C)	Ref
			IL ₁	IL ₂	IL ₃	IL ₁	IL ₂	IL ₃		
1		NH ₄ OAc	11	10	13	89	87	85	223–225	[11]
2		NH ₄ OAc	11	9	10	89	90	89	210–212	[11]
3		NH ₄ OAc	19	17	18	85	92	88	199–201	[11]
4		NH ₄ OAc	14	15	17	88	87	87	181–183	[11]
5		NH ₄ OAc	12	11	13	80	88	90	220	[11]
6		NH ₄ OAc	12	10	10	84	81	92	203–205	[11]
7		NH ₄ OAc	8	10	9	89	89	90	190–192	[11]

Table 2 continued

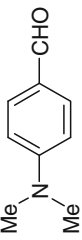
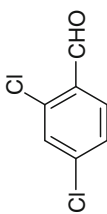
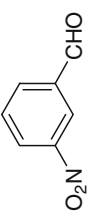
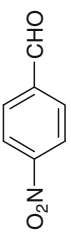
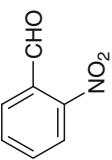
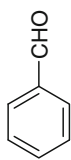
Entry	R	Amine	Time (min)			Yield (%) ^a			M.p.(°C)	Ref
			IL ₁	IL ₂	IL ₃	IL ₁	IL ₂	IL ₃		
8		NH ₄ OAc	16	16	19	70	78	69	205–207	[11]
9		NH ₄ OAc	32	30	35	85	83	80	166–168	[11]
10		EtNH ₂	48	35	44	90	85	84	174–176	[14]
11		EtNH ₂	25	23	37	84	89	89	161	[14]
12		EtNH ₂	32	28	41	88	81	85	156–157	[14]
13			31	28	32	80	78	80	209–211	[17]

Table 2 continued

Entry	R	Amine	Time (min)			Yield (%) ^a			M.p.(°C)	Ref
			IL ₁	IL ₂	IL ₃	IL ₁	IL ₂	IL ₃		
14			45	38	49	85	89	8	185–187	[17]
15			28	31	35	88	84	87	196–198	[17]
16			32	27	41	89	90	89	227–229	[17]
17			38	35	40	85	80	82	138–140	[17]
18			28	25	32	81	87	90	188–190	[17]
19			49	48	50	86	84	89	214–216	[12]
20		NH ₄ OAc	720	720	720	–	–	–	–	

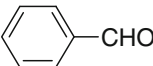
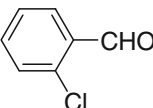
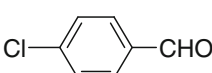
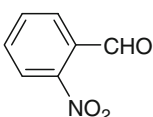
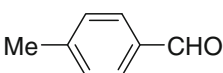
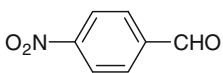
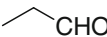
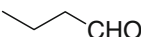
Table 2 continued

Entry	R	Amine	Time (min)		Yield (%) ^a			M.p.(°C)	Ref
			IL ₁	IL ₂	IL ₃	IL ₁	IL ₂	IL ₃	
21		NH ₄ OAc	720	720	720	–	–	–	–

^a Yields refer to isolated pure products

The molar ratio of the starting reactants was: isatoic anhydride (1 mmol), ammonium acetate (or primary amine) (1.2 mmol), benzaldehyde (1 mmol). The reaction was performed in an oil bath at 80 °C

Table 3 Preparation of 2,3-dihydroquinazolin-4(1*H*)-one derivatives (Method B)

Entry	Aldehyde	Time (min)			Yield (%) ^a			M.p. (°C)	
		IL ₁	IL ₂	IL ₃	IL ₁	IL ₂	IL ₃		
1		20	18	23	80	85	83	223–225	25
2		25	23	28	82	87	81	203–205	25
3		28	22	38	83	84	79	210–212	25
4		27	24	29	79	80	75	191–193	25
5		25	18	32	81	80	85	181–183	25
6		32	28	25	78	75	81	220	25
7		24	23	25	85	87	81	199–201	25
8		720	720	720	–	–	–	–	–
9		720	720	720	–	–	–	–	–

^a Yields refer to isolated pure products

The molar ratio of the starting reactants was: arylaldehyde (1 mmol) and anthranilamide (1 mmol). The reaction was performed in an oil bath at 80 °C

5 by decarboxylation and proton transfer. Subsequently, reaction of arylaldehyde activated by ILs with **5** proceeds, resulting in formation of imine intermediate **7**, which cyclizes to yield the final product.

In green organic synthesis, recovery of catalysts is very important. Thus, the reusability of ILs as catalysts was studied. After completion of the reaction, the reaction mixture was cooled to room temperature and 5 mL water was added. The ionic liquid dissolved in the water, and the solution was filtered to isolate the crude product. The separated product was washed twice with water (2 × 5 mL). To recover the ionic liquid, the water containing the IL was evaporated and the remaining viscous liquid was washed with CH₂Cl₂ (5 mL) and dried under reduced pressure. The recovered IL was tested to study its catalytic activity in the subsequent

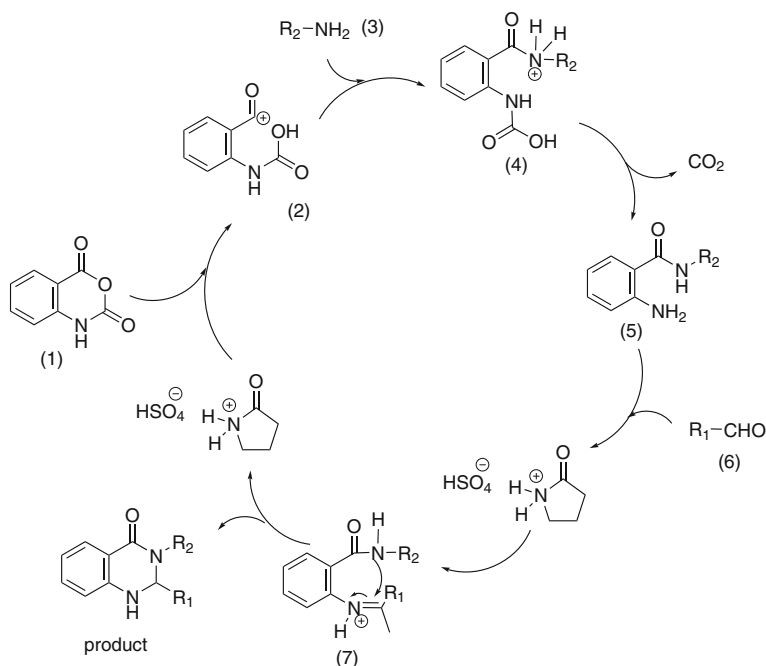
Table 4 Comparison of results obtained by use of 2-pyrrolidonium hydrogensulfate, *N*-methyl-2-pyrrolidonium hydrogensulfate, and *N*-methyl-2-pyrrolidonium dihydrogenphosphate with those obtained by use of other catalysts reported in the literature for synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones

Entry	Catalyst	Conditions	Time (min)	Yield (%)	Ref
1	Al(MS) ₃ ·4H ₂ O (5 mol %) ^a	EtOH/H ₂ O, reflux	120	88	[11]
2	<i>p</i> -TSA (50 mol %) ^a	H ₂ O, reflux	60	84	[12]
3	<i>p</i> -TSA (50 mol %) ^a	EtOH, reflux	240	75	[12]
4	KAl(SO ₄) ₂ ·12H ₂ O (40 mol %) ^a	EtOH, reflux	240	71	[13]
5	Cu(C ₆ H ₅ SO ₃) ₂ ·6H ₂ O (40 mol %) ^a	EtOH/H ₂ O, reflux	30	93	[18]
6	SiO ₂ -FeCl ₃ (0.005 g) ^a	Solvent-free, 80 °C	18	89	[15]
7	Ga(OTf) ₃ (1 mol %) ^a	EtOH, 70 °C	55	87	[19]
8	Ga(OTf) ₃ (1 mol %) ^a	DMSO, 85 °C	55	83	[19]
9	SrCl ₂ ·6H ₂ O (1 mol %) ^a	EtOH/H ₂ O, reflux	42	93	[20]
10	[hnmp][HSO ₄] (5 mol %) ^a	Solvent-free, 80 °C	11	89	This work
11	[NMP][HSO ₄] (5 mol %) ^a	Solvent-free, 80 °C	10	87	This work
12	[NMP][H ₂ PO ₄] (5 mol %) ^a	Solvent-free, 80 °C	13	85	This work
13	SiO ₂ -FeCl ₃ (0.005 g) ^b	Solvent-free, 80 °C	360	87	[15]
13	CuCl ₂ (100 mol %) ^b	EtOH, reflux, 70 °C	150	84	[21]
14	[Bmim]PF ₆ ^b	75 °C	35	89	[22]
15	Bu ₄ NBr (40 mol %) ^b	N ₂ atmosphere, 100 °C	90	82	[23]
16	ZrCl ₄ (2 mol %) ^b	EtOH, r.t.	25	95	[25]
17	H ₃ PW ₁₂ O ₄₀ (0.1 mol %) ^b	H ₂ O, r.t.	8	94	[26]
18	[hnmp][HSO ₄] (5 mol %) ^b	Solvent-free, 80 °C	20	80	This work
19	[NMP][HSO ₄] (5 mol %) ^b	Solvent-free, 80 °C	18	85	This work
20	[NMP][H ₂ PO ₄] (5 mol %) ^b	Solvent-free, 80 °C	23	83	This work

^a Based on the reaction of isatoic anhydride, ammonium acetate, and benzaldehyde^b Based on the reaction of anthranilamide and benzaldehyde

run without addition of fresh catalyst. ILs were tested for five runs. It was observed that the ILs as catalysts had very good reusability without substantial loss of their activity (Fig. 2).

In conclusion, we have described a rapid and highly efficient method for the green synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones by direct one-pot, three-component reaction of isatoic anhydride, ammonium acetate (or primary amines) and arylaldehydes (Method A) or anthranilamide and arylaldehydes (Method B) using Brønsted reusable acidic ionic liquids as catalysts under thermal solvent-free reaction conditions. With such successful results, this convenient and efficient procedure should provide a superior alternative to the existing methods because of its rapid and clean reactions and high yields. Furthermore, the simple work-up procedure makes these methods useful and important for synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones.



Scheme 3 The catalytic mechanism for preparation of the 2,3-dihydroquinazolin-4(1*H*)-one derivatives, with 2-pyrrolidonium hydrogensulfate as selected ionic liquid

Experimental

General

All reagents were purchased from Merck and Sigma–Aldrich and used without further purification. Ionic liquids, 2-pyrrolidonium hydrogensulfate ($[\text{hmp}][\text{HSO}_4]$)

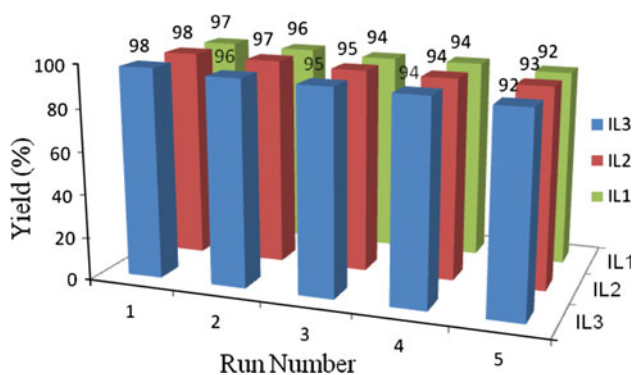


Fig. 2 Reusability of ionic liquids as catalysts

[31], *N*-methyl-2-pyrrolidonium hydrogensulfate ([NMP][HSO₄]) [32], and *N*-methyl-2-pyrrolidonium dihydrogenphosphate ([NMP][H₂PO₄]) [33], were prepared in accordance with reported procedures. All yields refer to isolated products after purification. NMR spectra were recorded on a Bruker Avance DPX 500 MHz instrument, in DMSO-*d*₆ as solvent, relative to TMS (0.00 ppm). IR spectra were recorded on a Jasco FT-IR 460 plus spectrophotometer. Melting points were determined in open capillaries with a Buchi 510 melting point apparatus. TLC was performed on Polygram SILG/UV 254 silica gel plates.

General procedure for synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones by three-component reaction of isatoic anhydride, primary amine or ammonium acetate, and arylaldehydes (Method A)

A stirred mixture of isatoic anhydride (10 mmol), ammonium acetate (or primary amine) (12 mmol), arylaldehyde (10 mmol), and 2-pyrrolidonium hydrogensulfate (0.09 g, 5 mol %, 0.5 mmol) or *N*-methyl-2-pyrrolidonium hydrogensulfate (0.098 g, 5 mol %, 0.5 mmol) or *N*-methyl-2-pyrrolidonium dihydrogenphosphate (0.098 g, 5 mol %, 0.5 mmol) was reacted in an oil bath at 80 °C for the appropriate time (Table 2). After completion of the reaction (indicated by TLC) the mixture was cooled to room temperature and 5 mL water was added. The ionic liquid dissolved in the water, and the mixture was filtered for separation of the crude product. The separated product was washed twice with water (2 × 5 mL) then purified by recrystallization from ethanol. To recover the ionic liquids, after isolation of the insoluble products, the water was evaporated and the remaining viscous liquid was washed with CH₂Cl₂ (5 mL) and dried under reduced pressure.

General procedure for synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones by reaction of anthranilamide and arylaldehydes (Method B)

A mixture of arylaldehyde (10 mmol), anthranilamide (10 mmol), and 2-pyrrolidonium hydrogensulfate (0.09 g, 5 mol %, 0.05 mmol) or *N*-methyl-2-pyrrolidonium hydrogensulfate (0.098 g, 5 mol %, 0.5 mmol) or *N*-methyl-2-pyrrolidonium dihydrogenphosphate (0.098 g, 5 mol %, 0.5 mmol) was reacted in an oil bath at 80 °C for the appropriate time (Table 3). After completion of the reaction (indicated by TLC) the mixture was cooled to room temperature and 5 mL water was added. The ionic liquid dissolved in the water, and the mixture was filtered for separation of the crude product. The separated product was washed twice with water (2 × 5 mL) then purified by recrystallization from ethanol. To recover the ionic liquids, after isolation of the insoluble products, water was evaporated and the remaining viscous liquid was washed with CH₂Cl₂ (5 mL) and dried under reduced pressure.

All the products are known. NMR and IR spectroscopic data for two of the compounds are given below.

2-(2-Chlorophenyl)-3-phenyl-2,3-dihydroquinazolin-4(1H)-one (Table 2, Entry 17)

M.p. 138–140 °C; ^1H NMR (500 MHz, $\text{DMSO}-d_6$): δ = 6.60 (d, J = 2.7 Hz, 1H), 6.74–6.79 (m, 2H), 7.17–7.21 (m, 3H), 7.27–7.31 (m, 5H), 7.37–7.39 (m, 1H), 7.46 (d, J = 2.5 Hz, 1H), 7.58–7.60 (m, 1H), 7.77 (dd, J = 1.3, 6.4 Hz, 1H), ppm; ^{13}C NMR (125 MHz, $\text{DMSO}-d_6$): δ = 70.2, 114.6, 114.8, 117.6, 126.6, 126.9, 127.4, 127.8, 128.2, 128.6, 129.8, 130.3, 131.3, 133.8, 136.9, 140.1, 146.0, 162.2 ppm; IR (KBr, cm^{-1}): 758, 1452, 1491, 1606, 1637, 2361, 3070, 3309.

2-(4-Nitrophenyl)-2,3-dihydroquinazolin-4(1H)-one (Table 3, Entry 7)

M.p. 199–201 °C; ^1H NMR (500 MHz, $\text{DMSO}-d_6$): δ = 5.93 (1H, s, CH), 6.71 (1H, t, J = 7.4, H Ar), 6.79 (1H, d, J = 8.1, H Ar), 7.29 (1H, t, J = 7.4, H Ar), 7.33 (1H, s, NH), 7.64 (1H, d, J = 7.6, H Ar), 7.76 (2H, d, J = 8.6, H Ar), 8.27 (2H, d, J = 8.6, H Ar), 8.53 (1H, s, CONH) ppm; ^{13}C NMR (125 MHz, $\text{DMSO}-d_6$): δ = 65.2, 114.5, 114.8, 117.4, 123.5, 127.3, 127.9, 133.4, 147.2, 147.4, 149.2, 163.2 ppm; IR (KBr, cm^{-1}): 753, 859, 1012, 1162, 1348, 1486, 1521, 1613, 1661, 3291, 3363.

Acknowledgments We are grateful to the University of Sistan and Baluchestan Research Council for the partial support of this research.

References

1. J. Zhu, H. Bienaym, *Multicomponent reaction* (Wiley, Weinheim, 2005)
2. J.A. Jouler, K. Mills, *Heterocyclic chemistry* (Wiley, United Kingdom, 2010)
3. H. Veisi, R. Ghorbani-Vaghei, *Tetrahedron* **66**, 7445–7463 (2010)
4. Y.S. Sadanandam, K.R.M. Reddy, A.B. Rao, *Eur. J. Med. Chem.* **22**, 169–173 (1987)
5. M. Bakavoli, O. Sabzevari, M. Rahimizadeh, *Chin. Chem. Lett.* **18**, 1466–1468 (2007)
6. J.B. Jiang, D.P. Hesson, B.A. Dusak, D.L. Dexter, G.J. Kang, E. Hamel, *J. Med. Chem.* **33**, 1721–1728 (1990)
7. W.L.F. Armarego, *Adv. Heterocycl. Chem.* **24**, 1 (1979)
8. V. Segarra, M.I. Crespo, F. Pujol, J. Belata, T. Domenech, M. Miralpeix, J.M. Palacios, A. Castro, A. Martinez, *Bioorg. Med. Chem. Lett.* **8**, 505–510 (1998)
9. Y. Yu, J.M. Ostresh, R.A. Houghten, *J. Org. Chem.* **67**, 5831–5834 (2002)
10. A.M. Inamuddin, *Green solvents I: properties and applications in chemistry* (Springer, United Kingdom, 2012)
11. Z. Song, L. Liu, Y. Wang, X. Sun, *Res. Chem. Intermed.* **38**, 1091–1099 (2012)
12. M. Baghbanzadeh, P. Salehi, M. Dabiri, G. Kozehgary, *Synthesis* **2**, 344–348 (2006)
13. M. Dabiri, P. Salehi, S. Otokesh, M. Baghbanzadeh, G. Kozehgarya, A.A. Mohammadi, *Tetrahedron Lett.* **46**, 6123–6126 (2005)
14. I. Mohammadpoor-Baltork, A.R. Khosropour, M. Moghadam, S. Tangestaninejad, V. Mirkhani, M. Soltani, A. Mirjafari, *J. Heterocycl. Chem.* **48**, 1419–1427 (2011)
15. M. Ghashang, K. Azizi, H. Moulavi-Pordanjani, H.R. Shaterian, *Chin. J. Chem.* **29**, 1617–1623 (2011)
16. K. Niknam, N. Jafarpour, E. Niknam, *Chin. Chem. Lett.* **22**, 69–72 (2011)
17. Z. Karimi-Jaberi, R. Arjmandi, *Monatsh. Chem.* **142**, 631–635 (2011)
18. M. Wang, T.T. Zhang, Y. Liang, J.J. Gao, *Monatsh. Chem.* **143**, 835–839 (2012)
19. J. Chen, D. Wu, F. He, M. Liu, H. Wu, J. Ding, W. Su, *Tetrahedron Lett.* **49**, 3814–3818 (2008)
20. M. Wang, T.T. Zhang, Y. Liang, J.J. Gao, *Chin. Chem. Lett.* **22**, 1423–1426 (2011)
21. R.J. Abdel-Jalil, W. Voelter, M. Saeed, *Tetrahedron Lett.* **45**, 3475–3476 (2004)

22. J. Chen, W. Su, H. Wu, M. Liu, C. Jin, *Green Chem.* **9**, 972–975 (2007)
23. A. Davoodnia, S. Allameh, A.R. Fakhari, N. Tavakoli-Hoseini, *Chin. Chem. Lett.* **21**, 550–553 (2010)
24. M. Wang, J.J. Gao, Z.G. Song, L. Wang, *Chem. Heterocycl. Compd.* **47**, 851–855 (2011)
25. M. Abdollahi-Alibeik, E. Shabani, *Chin. Chem. Lett.* **22**, 1163–1166 (2011)
26. Y.X. Zong, Y. Zhao, W.C. Luo, X.H. Yu, J.K. Wang, Y. Pan, *Chin. Chem. Lett.* **21**, 778–781 (2010)
27. H.R. Shaterian, F. Khorami, A. Amirzadeh, R. Doostmohammadi, M. Ghashang, *J. Iran. Chem. Res.* **2**, 57–62 (2009)
28. H.R. Shaterian, M. Ghashang, M. Feyzi, *Appl. Catal. A* **345**, 128–133 (2008)
29. H.R. Shaterian, A. Hosseini, M. Ghashang, *Arkivoc* **ii**, 59–67 (2009)
30. H.R. Shaterian, F. Rigi, *Starch-Starke* **63**, 340–346 (2011)
31. B. Huang, Y. Wang, K. Zhang, Y. Fang, B. Zhou, *Chin. J. Catal.* **28**, 743–748 (2007)
32. C. Xie, H. Li, L. Li, S. Yu, F. Liu, *J. Hazard. Mater.* **151**, 847–850 (2008)
33. H. Guo, X. Li, J.-L. Wang, X.-H. Jin, X.-F. Lin, *Tetrahedron* **66**, 8300–8303 (2010)