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SYNTHESIS OF 2,3-DIHYDROQUINAZOLIN-4(1H)-ONES USING BRONSTED ACIDIC IONIC LIQUID

HESHMATOLLAH ALINEZHAD, MAHMOOD TAJBAKHSH and NEDA GHOBADI

Department of Organic Chemistry
Faculty of Chemistry
University of Mazandaran
Babolsar
Iran
e-mail: heshmat@umz.ac.ir

Abstract

2,3-Dihydroquinazolin-4(1H)-ones were efficiently synthesized via one-pot, three-component reaction of isatoic anhydride, primary amines or ammonium acetate, and different aromatic aldehydes in the presence of 3-methyl-1-sulphonic acid imidazolium hydrogen sulphate $\{[\mathrm{Msim}]\mathrm{HSO_4}\}$ as an efficient Brønsted acidic ionic liquid in aqueous media. The catalyst could be recovered and reused for at least four cycles without significant loss of activity.

1. Introduction

2,3-Dihydroquinazolin-4(1H)-one derivatives are an important class of fused heterocycles that display a wide range of biological, pharmacological, and medicinal properties [1], including antitumor, antibiotic, antipyretic, analgesic, antihypertonic, diuretic, antihistamine, antidepressant, and

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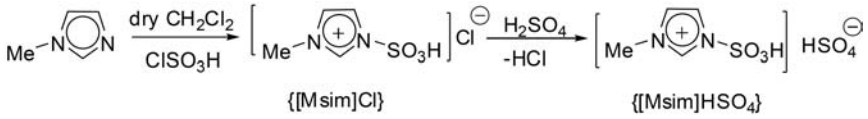
vasodilation activities [2]. Quinazolin-4(1H)-ones are valuable intermediates in synthetic organic chemistry and are easily oxidized with $KMnO_4$ to the corresponding quinazoline-4(3H)-ones that are useful as growth inhibitor against leukemia cells [3, 4].

Most conventional method for the preparation of 2,3-dihydroquinazolin-ones includes condensation of aryl, alkyl, and heteroaryl aldehydes with anthranilamide in the presence of *p*-toluenesulphonic acid as a catalyst [5]. Conventionally, these compounds have been synthesized by variety of procedures as stated in literature [6-9]. Reductive cyclization of *o*-nitrobenzamid or *o*-azidobenzamide was also reported for the synthesis of 2,3-dihydroquinazolin-ones [10]. Recently, condensation of isatoic anhydride, aldehydes, and ammonium acetate or primary amines using different reagents, namely, *p*-TsOH [11], amberlyst-15/microwave [12], montmorillonite K-10 [13], ionic liquid [14-16], and supported ionic liquid [17], silica sulphuric acid [18], $Zn(PFO)_2$ [19], $Al(H_2PO_4)_3$ [20], $KAI(SO_4)_2 \cdot 12H_2O$ (alum) [21], $Ga(OTf)_3$ [22], silica-bonded *N*-propyl-sulphamic acid [23], and ceric ammonium nitrate [24], have been reported in the literature.

Multicomponent one-pot reactions (MCRs) have received a great deal of interest because of their atom-economy and straight forward reaction design due to substantial minimization of waste, labor, time, and cost and easier access to diverse compound libraries [25-29]. MCRs lead to interesting heterocyclic scaffolds, which are particularly useful for the construction of various chemical libraries of privileged medicinal molecules [30-33].

Furthermore, ionic liquids (IL) have been widely used as eco-friendly solvents, catalysts, and reagents in green synthesis because of their unique properties, such as low volatility, non-flammability, high thermal stability, negligible vapour pressure, and ability to dissolve a wide range of materials [34, 35]. Brønsted acidic ionic liquids have been designed to replace solid acids and traditional mineral liquid acids like sulphuric acid and hydrochloric acid in chemical procedures [36-38].

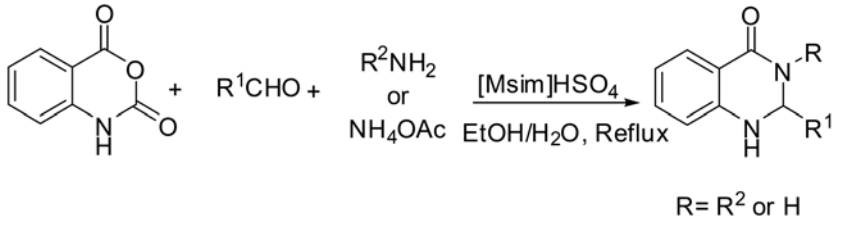
3-Methyl-1-sulphonic acid imidazolium hydrogen sulphate $\{[\text{Msim}]\text{HSO}_4\}$ is easily synthesized by addition of chlorosulphonic acid to 1-methylimidazole and subsequent treatment with H_2SO_4 (Scheme 1) as reported previously [39]. It has been used as an efficient catalyst for protection of alcohols [39] and synthesis of coumarins [40].



Scheme 1. The synthesis of 3-methyl-1-sulphonic acid imidazolium hydrogen sulphate ionic liquid.

Although many of the reported methods for the synthesis of quinazolinones are effective, but some of these methods associated with certain drawbacks, such as long reaction time, low yields, expensive and large amount of catalyst, high reaction temperature, and using microwave irradiation for accelerated synthesis.

In our efforts toward the development of environmentally methodologies [41, 42], herein, we report an efficient one-pot three-component reaction, involving primary amines or ammonium acetate, isatoic anhydride, and different aldehydes using catalytic amount of 3-methyl-1-sulphonic acid imidazolium hydrogen sulphate $\{[\text{Msim}]\text{HSO}_4\}$ as a recyclable and reuseable Brønsted acidic ionic liquid (Scheme 2).



Scheme 2. Synthesis of 2,3-dihydro-quinazolin-4(1H)-ones.

2. Experimental

2.1. General

All reagents were purchased from Merck and Aldrich and used without further purification. All yields refer to isolated products after purification. Products were characterized by comparison with authentic samples and by spectroscopy data (FTIR, ^{1}HNMR, and ^{13}CNMR spectra) and melting point. 1H-and 13C-NMR spectra were recorded on a Bruker DRX-400 AVANCE (400 and 100MHz for ^{1}H and ^{13}C, respectively) in DMSO-d_6 as the solvent. Chemical shifts are on the δ scale, relative to internal Me_4Si, and IR spectra were determined on a on a Bruker Tensor 27, using the KBr disk technique. Melting points were determined on an electro-thermal apparatus IA9200.

2.2. General procedure for the synthesis of quinazolinone using IL

To a mixture of isatoic anhydride (1.0m mol), aromatic aldehydes (1.0m mol), ammonium acetate or primary amines (1.0m mol) in 3mL EtOH/H_2O (2/1(v/v)) was added 3-methyl-1-sulphonic acid imidazolium hydrogen-sulphate (0.018g, 0.07m mol) and then reaction mixture was refluxed for appropriate time (Tables 2 and 3). After completion of the reaction, as indicated by TLC (EtOAc-cyclohexane, (3:7)), the reaction mixture was cooled to room temperature, filtered and recrystallized in ethanol to afford the pure product.

2.3. Spectral data

2.3.1. Spectral data of ionic liquid [Msim]HSO_4 synthesised

Viscous pale yellow oil: FT-IR (liquid film): 2924, 1713, 1586, 1375, 1168, 1063, 878cm^{-1}; ^{1}H–NMR (400 MHz, DMSO-d_6) : δ = 3.84 (s, 3H, CH_3), 7.61 (s, 1H), 7.66 (s, 1H), 9.01 (s, 1H), 12.24 (s, 1H), 14.22 (s, 1H); ^{13}C–NMR (DMSO-d_6) : δ = 35.8, 120.1, 123.6, 136.1.

2.3.2. Spectral data for selected products

2-(2-Nitrophenyl)-2, 3-dihydroquinazolin-4(1H)-one:

(Table 3, entry 9) Yellow powder, IR (KBr): 3375, 1684, 1570, 1523, 1350, 739 cm^{-1}; ^{1}H–NMR (400MHz, DMSO-d_6) : δ = 8.20 (1H), 7.94 (dd, 1H, J = 1.6, 8Hz), 7.80 (dd, 1H, J = 1.2, 8Hz), 7.68 (td, 1H, J = 0.8, 7.8Hz), 7.61 (dd, 1H, J = 1.6, 8Hz), 7.53 (td, 1H, J = 1.6, 7.2Hz), 7.25 (td, 1H, J = 1.6, 8Hz), 6.95 (1H), 6.94 (d, 1H, J = 15.6Hz), 6.79 (dd, 1H, J = 0.8, 8.2Hz), 6.69 (td, 1H, J = 0.8, 7.6Hz), 5.34(d, 1H, J = 6.4Hz); ^{13}C–NMR (400MHz, DMSO-d_6) : δ = 163.6, 148.4, 147.9, 134.3, 133.8, 133.7, 130.6, 129.6, 128.8, 127.8, 125.9, 124.7, 117.6, 115.3, 115, 65.5.

2-(4-Methoxyphenyl)-3-(phenyl)-2,3-dihydroquinazolin-4(1H)-one:

(Table 4, entry 8) White powder, IR (KBr): 3329, 2958, 1637, 1516, 781cm^{-1}; ^{1}H–NMR (400MHz, DMSO-d_6) : δ : 8.02 (dd, 1H, J = 1.6, 7.6Hz), 7.37-7.33 (m, 2H), 7.30-7.26 (m, 4H), 7.10-7.06 (m, 2H), 6.89 (t, 1H, J = 7.6Hz), 6.82-6.78 (m, 2H), 6.62 (d, 1H, J = 7.6Hz), 6.05 (s, 1H), 4.79 (s, 1H), 3.76 (s, 3H). ^{13}C–NMR (400MHz, DMSO-d_6) : δ = 163.2, 145.4, 139.9, 133.7, 133.2, 129.0, 128.7, 128.5, 126.8, 119.4, 116.6, 114.6, 114.2, 75.0, 55.3.

3. Results and Discussion

Condensation reaction of isatoic anhydride, benzaldehyde, and ammonium acetate as a model reaction was studied under different reaction condition for a 100% conversion to 2,3-dihydroquinazolin-4(1H)-ones (Table 2). As it is clear in this table, when the model reaction was carried out in the presence of 10mol% of $[Msim]HSO_4$ in nonpolar solvents, such as CH_2Cl_2, hexane, ethyl acetate, a low yield of the product was obtained (Table 2, entries 1-4). This could be due to insolubility of the isatoic anhydride in these solvents. However, this reaction in protic solvents such as H_2O, MeOH, and EtOH gave a high yield of the expected product (Table 2, entries 5-12). Although, H_2O was a good solvent for this reaction, but workup procedure was tedious. The model

reaction under solvent-free condition at different temperatures did not give suitable yield of the product (Table 2, entries 15 and 16). This reaction when carried out without catalyst in $EtOH/H_2O$ after 4 hours no product was formed, which implies the role of the catalyst in this reaction (Table 2, entries 13).

The catalytic activity of 3-methyl-1-sulphonic acid imidazolium chloride {[Msim]Cl} was also investigated under same reaction condition and gave a lower yields of the expected production longer reaction time (Table 2, entry 14) compare with $[Msim]HSO_4$ (Table 2, entry 10). After optimizing the reaction conditions (Table 2, entry 12), the generality and drawbacks of the method were investigated. The results are show in Tables 3 and 4. As it is clear in Table 3, three component reaction of different substituted aldehydes, isatoic anhydride, and ammonium acetate under optimizing conditions gave a good yield of the 2-subtituted 2,3-dihydroquinazolin-4(1H)-ones in short reaction times (entries 1-14). When anilines bearing either electron-withdrawing or electron-donating groups on the benzene ring used instead of ammonium acetate, slightly lower yields of the desired products obtained in longer reaction times (Table 4, entries 1-16).

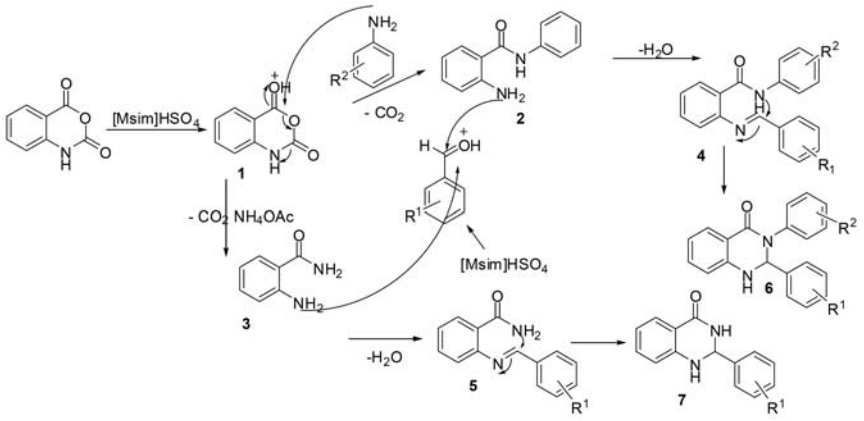
The reusability of the $[Msim]HSO_4$ was tested in the reaction of *p*-chlorobenzaldehyde and ammonium acetate with isatoic anhydride in $EtOH/H_2O$ mixed solvents. After each run, the reaction mixture was cooled to room temperature and filtered. The medium along with catalyst was evaporated under reduced pressure and the remaining catalyst was washed with $EtOAc\,(2\times 5ml)$ to exclude the impurities. The recovered catalyst was reused again and found to give comparable yield of the product (Table 1). The catalyst was employed four runs, although the activity of the catalyst gradually decreased. Subsequently, the decrease of the reaction yield in longer reaction time in the third and forth cycles, is due to the loss of some catalyst (0.002gr) after second cycle. These result clearly indicates that the Brønsted acidic ionic liquid $[Msim]HSO_4$ as a catalyst for the preparation of 2,3-dihydroquinazolin-4(1H)-one derivatives is recyclable.

Table 1. The reusability of the catalyst

Run No.	Time (min)	Yield (%)[a]
Fresh	30	95
1	30	90
2	30	90
3	45	85
4	45	80

[a] Yields refer to isolated product.

A plausible mechanism for the formation of the corresponding products is shown in Scheme 3. According to other reported mechanisms catalyzed by Brønsted and Lewis acids [7, 11, 14]. It is assumed that $[\text{Msim}]\text{HSO}_4$ as a Brønsted acid can protonated isatoic anhydride to give intermediate 1 to facilitate nucleophilic attack of aniline or ammonium acetate on the carbonyl unit, followed by decarboxylation to give intermediates 2 or 3. Condensation of intermediates 2 or 3 with protonated benzaldehyde and subsequent dehydration generate imines 4 or 5, which underwent intramolecular cyclization to afford final products 6 or 7.



Scheme 3. A plausible mechanism for the formation for products of 6 and 7.

Table 2. Condensation of isatoic anhydride, benzaldehyde, and ammonium acetate under different reaction conditions

Entry	Solvent	Catalyst (mol %)	Conditions	Yield (%)[a]
1	Dichloromethane	10	Reflux, 4h	Trace
2	Hexane	10	Reflux, 4h	20
3	Ethyl acetate	10	Reflux, 3h	20
4	Toluene	10	Reflux, 4h	Trace
5	Methanol	10	Reflux, 2.5h	90
6	Ethanol	10	Reflux, 1h	90
7	Water	10	Reflux, 2h	85
8	Water/Ethanol (1/1, v/v)	10	Reflux, 1.5h	80
9	Water/Ethanol (2/1, v/v)	10	Reflux, 2h	85
10	Water/Ethanol (1/2, v/v)	10	Reflux, 40mim	90
11	Water/Ethanol (1/2, v/v)	5	Reflux, 1h	90
12	Water/Ethanol (1/2, v/v)	7	Reflux, 30min	95
13	Water/Ethanol (1/2, v/v)	–	Reflux, 4h	No reaction
14[b]	Water/Ethanol (1/2, v/v)	10	Reflux,1h	80
15	Solvent free	10	90°C, 2h	60
16	Solvent free	10	120°C, 1.5h	70

[a]Isolated yield.

[b]The reaction occurred using 3-methyl-1-sulphonic acid imidazolium chloride {[Msim]Cl}.

[c]Catalyst was recycled for four runs.

Table 3. Synthesis of 2-substituted 2,3-dihydroquinazolin-4(1H)-ones catalyzed by $[Msim]HSO_4$[a]

Entry	R^1	Time (min)	Yield[b]	Mp (°C)[Ref]
1	C_6H_5-	30	95	220-222[43]
2	$2-ClC_6H_4-$	25	90	200-202[44]
3	$3-ClC_6H_4-$	30	90	185-187[45]
4	$4-ClC_6H_4-$	30	95	204-205[22]
5	$4-BrC_6H_4-$	25	90	196-197[45]
6	$2-H_3COC_6H_4-$	30	85	165-167[46]
7	$4-H_3COC_6H_4-$	35	80	190-192[13]
8	$4-H_3CC_6H_4-$	40	90	228-229[43]
9	$2-O_2NC_6H_4-$	40	90	186-188[44]
10	$4-O_2NC_6H_4-$	30	95	195-197[44]
11	$2-HOC_6H_4-$	25	85	208-209[45]
12	$4-HOC_6H_4-$	35	80	275-277[45]
13	$4-NCC_6H_4-$	30	90	252-253[47]
14	$4-(Me)_2NC_6H_4-$	45	85	206-208[48]

[a]Reaction conditions: isatoic anhydride: aromatic aldehyde: ammonium acetate (1:1:1) and $[Msim]HSO_4$ (7mol%) in 3mL of $EtOH/H_2O$ (2/1(v/v)).

[b]Yields to the isolated pure products.

Table 4. Synthesis of 2,3-disubstituted 2,3-dihydroquinazolin-4(1H)-ones catalyzed by $[Msim]HSO_4$[a]

Entry	R^1	R^2	Time (h:min)	Yield[b]	Mp (°C)[Ref]
1	C_6H_5-	C_6H_5-	1:30	95	203-204[49]
2	$4-FC_6H_4-$	C_6H_5-	1:15	90	224-226[50]
3	$4-ClC_6H_4-$	C_6H_5-	1:10	95	217-219[49]
4	$4-BrC_6H_4-$	C_6H_5-	1:20	95	222-225[49]
5	$4-O_2NC_6H_4-$	C_6H_5-	1:40	87	194-196[49]
6	$4-H_3CC_6H_4$	C_6H_5-	1:40	90	191-193[49]
7	$4-(Me)_2NC_6H_4-$	C_6H_5-	2:00	88	184-185[49]
8	$4-H_3COC_6H_4$	C_6H_5-	1:30	90	204-205[49]
9	C_6H_5-	$4-ClC_6H_4$	2:00	88	210-212[49]
10	C_6H_5-	$4-H_3CC_6H_4$	1:30	95	196-18[49]
11	$4-ClC_6H_4-$	$4-H_3CC_6H_4$	1:2	95	251-252[51]
12	$4-FC_6H_4-$	$4-H_3CC_6H_4$	1:20	83	241-242[52]
13	$4-NO_2C_6H_4-$	$4-H_3CC_6H_4$	1:30	85	212-214[50]
14	C_6H_5-	$4-H_3COC_6H_4$	1:50	95	209-211[49]
15	$4-ClC_6H_4-$	$4-H_3COC_6H_4$	1:40	97	238-240[50]
16	$4-H_3COC_6H_4$	$4-H_3COC_6H_4$	2:00	93	227-228[53]

[a]Reaction conditions: isatoic anhydride: aromatic aldehyde: primary amine (1:1:1) and $[Msim]HSO_4$ (7mol %) in 3mL of $EtOH/H_2O$ (2 / 1(v/v)).

[b]Yields to the isolated pure products.

4. Conclusion

In summary, we have developed an eco-friendly method to synthesize 2,3-dihydroquinazolin-4(1H)-ones in excellent yields using $[Msim][HSO_4]$ as an ionic liquid under reflux in $EtOH/H_2O$ in one-pot three-component procedure. High yields, recyclability of catalyst, cheap and commercially available starting materials, ease of work up procedure are some of the salient features of the developed protocol.

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