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Cellulose-SO_3H: an efficient and biodegradable solid acid for the synthesis of quinazolin-4(1*H*)-ones

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ABSTRACT

The condensation of 2-aminobenzamide with aldehydes or ketones has been achieved using cellulose-sulfonic acid under mild reaction conditions to furnish 2,3-dihydroquinazolin-4(1*H*)-ones in good yields with a high selectivity. The use of biodegradable solid acid catalyst, cellulosesulfonic acid makes this method quite simple, more convenient, and practical. This catalyst was also found to be very active for the synthesis of hydroxyalkylquinazolin-4-ones from cyclic enol ethers.

2,3-Dihydroquinazolinones exhibit a wide range of biological activities, such as antitumor, antibiotic, antidefibrillatory, antipyretic, analgesic, antihypertonic, diuretic, antihistamine, antidepressant, and vasodilating behavior.[1] They play a key role in various cellular processes. Furthermore, quinazolinone skeleton is frequently found in various natural products (Fig. 1).[2,3]

Therefore, several efforts have been made to develop elegant approaches for the synthesis of quinazolinone alkaloids.[3] In addition, a variety of methods have been developed for the synthesis of quinazolinone scaffolds.[4,5] Of these, the condensation of 2-aminobenzamide with aldehydes or ketones is one of the simplest and direct methods for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones. Various acid catalysts, such as *p*-TSA/$NaHSO_3$,[6a] $TiCl_4$/Zn,[6b] $CuCl_2$,[6c] ionic liquid–water,[6d] and TFA,[6e] ammonium chloride,[6f] and chiral phosphoric acids[7] have been utilized to accomplish this transformation. However, many of these methods involve the use of expensive reagents, extended reaction times, high temperatures, and also require tedious work-up procedures. Therefore, the development of novel methods for the synthesis of quinazolin-4(3*H*)-ones is of great importance because of their potential biological and pharmaceutical activities.

Recently, the use of heterogeneous catalysts has received a special attraction as user-friendly catalysts because of recyclability, operational simplicity, and minimal waste disposal.[8] Very recently, cellulose-SO_3H has been introduced as a biodegradable solid acid heterogeneous catalyst for the synthesis of quinolines, α-amino nitriles, aryl-*14H*-dibenzo[*a.j*]xanthenes, tetrahydroquinolines, and functionalized pyrrolidines.[9] However, there are no reports on the use of cellulose-SO_3H for the preparation of quinazolin-4(1*H*)-ones under mild reaction conditions.

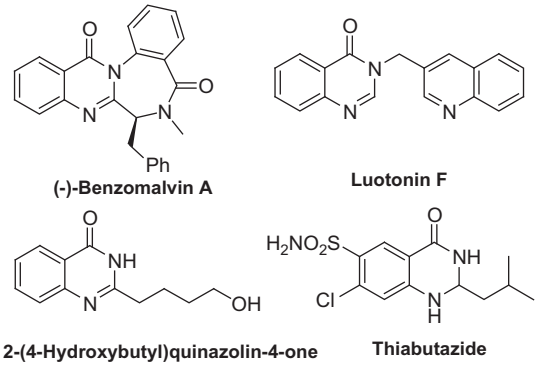

Figure 1. Examples of some natural products bearing quinazolin-4-one skeleton.

Following our interest on the catalytic applications of solid acid catalysts,[10] we herein report, for the first time, a novel method for the synthesis of hydroxyalkylquinazolin-4-ones using a biodegradable and recyclable solid acid catalyst, that is, cellulose-SO_3H. Initially, we have attempted the reaction of anthranilamide with 3,4-dihydro-2*H*-pyran in the presence of cellulose-SO_3H.

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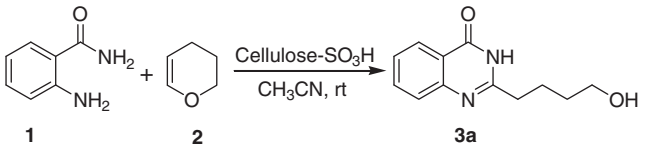

Scheme 1. Reaction of anthranilamide with 3,4-dihydro-2*H*-pyran.

Interestingly, the reaction went to completion at room temperature and the desired 2-(4-hydroxybutyl)quinazolin-4-one **3a** was obtained in 65% yield (Scheme 1, Table 1 entry a).

This result provided the incentive to extend this process for other enol ethers. Similarly, 2,3-dihydrofuran also reacted well with anthranilamide to give the corresponding 2-(3-hydroxypropyl)quinazolin-4-one (Table 1, entry b). Notably, 2-aminobenzenesulfonamide also participated well in this reaction (Table 1, entries c and d). Next, we have attempted the cyclization of anthranilamide with 3,4,6-tri-O-acetyl-D-glucal under similar conditions. Interestingly, the desired sugar substituted quinazolinone **3e** was obtained in 60% yield (Table 1, entry e).

The excellent catalytic activity of cellulose-SO_3H in the synthesis of hydroxyalkylquinazolin-4(1*H*)-ones prompted us to investigate its use in the condensation of anthranilamide with various aldehydes and ketones. Interestingly, several aromatic aldehydes, such as *p*-anisaldehyde, *p*-chlorobenzaldehyde, *m*-phenoxybenzaldehyde, 2-naphthaldehyde, 2-furfural, 3,4,5-trimethoxybenzaldehyde, 2-nitrobenzaldehyde, and 4-nitrobenzaldehyde participated well in this reaction (Table 2, entries a–h). Notably, acid sensitive cinnamaldehyde also gave the desired 2,3-dihydroquinazolin-4-one in 82% yield (Table 2, entry j). The reaction with aliphatic aldehydes, such as 3-phenylpropanaldehyde, cyclohexanecarboxaldehyde, butyraldehyde worked well to furnish the desired products in good yields (Table 2, entries i, k and l). In all cases, 2,3-dihydroquinazolin-4-ones were obtained exclusively in high yields (Scheme 2, Table 2).

This method works well with both electron-rich as well as electron deficient aryl aldehydes (Table 2). Next, we have attempted the coupling of anthranilamide with ketones, such as isatin, cyclohexanone, and cyclopentanone. Interestingly, these cyclic ketones underwent smooth coupling with anthranilamide under similar conditions to furnish the corresponding 2-spiro-2,3-dihydroquinazolin-4-ones in good yields (Table 2, entries m–o). The results are summarized in Table 2 after optimizing the reaction conditions with different aryl aldehydes and ketones.[11] It was found that all the reactions proceeded well at room temperature except with D-glucal affording the corresponding products in good yields (Tables 1 and 2). All the products were fully characterized and confirmed by ^{1}H NMR, ^{13}C NMR, IR, and mass spectroscopy. No significant change in yields was observed when either ketones or substituted aryl aldehydes were used. In the absence of cellulose-SO_3H, no cyclized product was observed even in refluxing acetonitrile. As solvent, acetonitrile gave the best results. It is noteworthy to highlight that the catalyst could be recycled three times without significant loss of activity. For example, the treatment of anthranilamide (1 mmol) with *p*-chlorobenzaldehyde (1 mmol) in the presence of cellulose-SO_3H (50 mg) gave the 2-phenyl-2,3-dihydroquinazolin-4-one in 90%, 87%, and 82% yields over three cycles. This clearly shows the heterogeneous nature of the catalyst with excellent recycling capability, which is the advantage of our method over existing ones.

In summary, we have demonstrated a novel protocol for the preparation of 2,3-dihydroquinazolin-4-ones using cellulose-SO_3H as a recyclable solid acid catalyst.[12] This method also describes for the first time the synthesis of hydroxyalkylquinazolin-4-ones. The use of cellulose-SO_3H makes this method simple, convenient, and economically viable for a large scale synthesis of 2,3-dihydroquinazolin-4-ones. The major advantages of the present method are short reaction times, high yields, simplicity in operation, cost-effective and mild reaction conditions.

Acknowledgments

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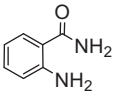
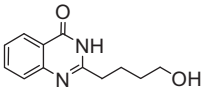
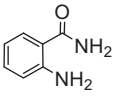
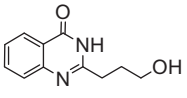
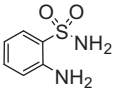
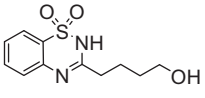
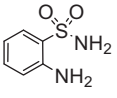
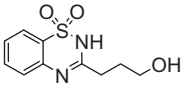
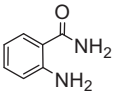
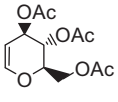
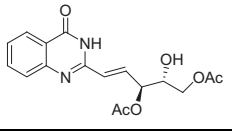
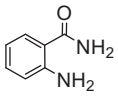
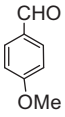
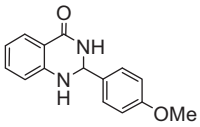
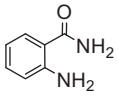
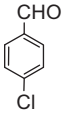
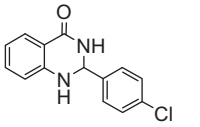
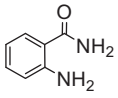
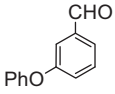
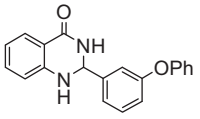
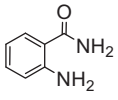
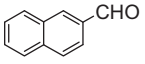
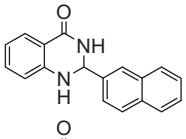
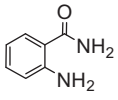
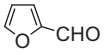
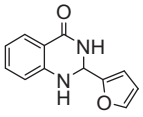
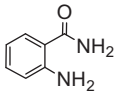
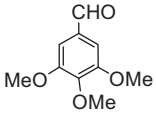
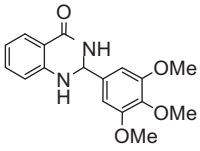
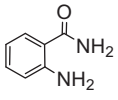
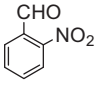
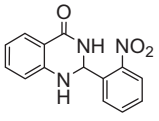
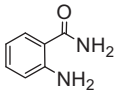
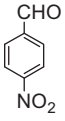
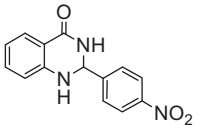
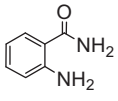
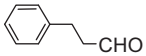
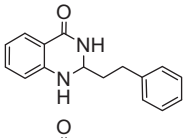
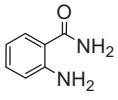
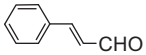
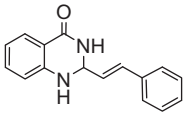
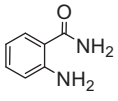
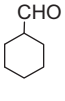
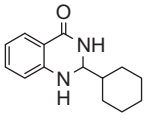
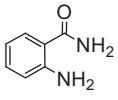
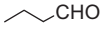
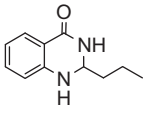
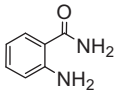
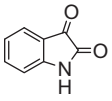
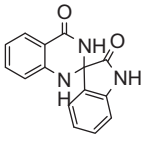
Table 1
Cellulose-SO_3H-catalyzed one-pot synthesis of hydroxyalkylquinazolinones

Entry	Anthranilamide	Enol eher	Product (**3**)[a]	Time (h)	Yield[b] (%)
a				1.0	65
b				1.5	60
c				1.0	62
d				1.5	57
e				12	60[c]

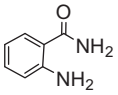
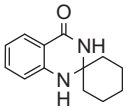
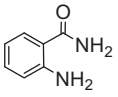
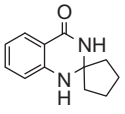
[a] All products were characterized by ^{1}H NMR, IR, and mass spectroscopy.
[b] Yield refers to pure products after chromatography.

Table 2
Cellulose-SO_3H-catalyzed synthesis of dihydroquinazolinones

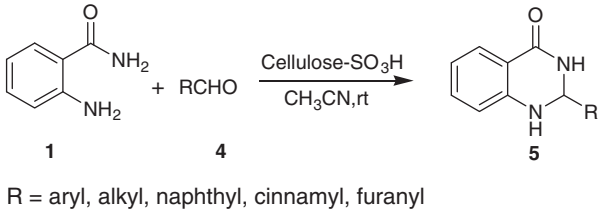
Entry	Anthranilamide (1)	Carbonyls (4)	Product (5)[a]	Time (min)	Yield[b] (%)
a				60	88
b				55	90
c				50	84
d				55	80
e				45	80
f				50	92
g				60	77
h				65	80
i				50	85
j				45	82
k				55	88
l				48	85
m				50	87

(*continued on next page*)

Table 2 (*continued*)

Entry	Anthranilamide (1)	Carbonyls (4)	Product (5)[a]	Time (min)	Yield[b] (%)
n				45	82
o				40	78

[a] All products were characterized by ^{1}H NMR, IR and mass spectroscopy.
[b] Yield refers to pure products after chromatography.



1 4 5

Scheme 2. Reaction of anthranilamide with aldehydes.

References and notes

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11. *General procedure*: A mixture of carbonyl compound (1 mmol), anthranilamide (1 mmol), and cellulose-SO_3H (50 mg) in acetonitrile (5 mL) was stirred at room temperature for a specified time (Table 1). After completion of the reaction, as indicated by TLC, the catalyst was filtered and washed with ethyl acetate (2×10 mL) and the resulting filtrate was dried over Na_2SO_4 and concentrated under *vacuum*. The product was purified by silica gel column chromatography (50:50, hexane/ethyl acetate) to afford the pure 2,3-dihydroquinazolin-4(1*H*)-one. The products thus obtained were characterized by IR, NMR and mass spectroscopy. The spectral data were found to be consistent with authentic samples.
3c (Table 1): Colorless liquid, ^{1}H NMR (300 MHz, DMSO-D_6): δ 7.44 (d, J = 6.6 Hz, 1H), 7.19 (td, J = 8.6, 11.8 Hz, 1H), 6.78 (d, J = 8.1 Hz, 1H), 6.66 (t, J = 7.7 Hz, 1H), 4.69 (t, J = 6.2 Hz, 2H), 1.38–0.82 (m, 6H); ^{13}C NMR (75 MHz, DMSO-D_6): δ 143.7, 132.5, 128.2, 123.6, 121.2, 116.1, 115.8, 65.7, 33.3, 32.1, 20.9.; IR (KBr): ν_{max} 3404, 2941, 2874, 1647, 1488, 1221, 1030, 769, 639; MS (ESI): 277 [M+Na]; HRMS Calcd for $C_{11}H_{14}N_2O_3Na$: 277.1253. Found: 277.1248.
3e (Table 1): (2S,3R,E)-2-hydroxy-5-(4-oxo-3,4-dihydroquinazolin-2-yl)pent-4-ene-1,3-diyl diacetate: Semi-solid, ^{1}H NMR (300 MHz, DMSO-D_6): δ 7.72 (brs, 1H), 7.63 (d, J = 7.9 Hz, 1H), 7.18 (t, J = 6.7 Hz, 1H), 6.68 (t, J = 6.7 Hz, 2H), 5.89 (dd, J = 16.8, 6.2 Hz, 1H), 5.19 (d, J = 5.6 Hz, 1H), 4.16–3.76 (m, 4H) 2.30 (s, 6H); ^{13}C NMR (75 MHz, DMSO-D_6): δ 170.5, 169.5, 163.6, 147.9, 133.4, 131.7, 128.7, 127.3, 114.4, 73.7, 69.3, 67.5, 20.9, 20.7.; IR(KBr): ν_{max} 3432, 3084, 2919, 1650, 1490, 1254, 1034, 760; MS (ESI): 347 [M+H]; HRMS Calcd for $C_{17}H_{19}N_2O_6$: 347.1243. Found: 347.1248.
5a (Table 2): 2-(4-Methoxyphenyl)-2,3-dihydroquinazolin-4(1H)-one: White solid, m.p. 189–191 °C, ^{1}H NMR (300 MHz, $CDCl_3$+DMSO, 3:1): δ 7.78 (brs, 1H), 7.64 (dd, J = 7.7 Hz, 1H), 7.43 (d, J = 8.6 Hz, 2H) 7.18 (t, J = 8.1 Hz, 1H), 6.88 (d, J = 8.4 Hz, 2H), 6.63–6.73 (m, 2H), 5.71 (s, 1H), 3.78 (s, 3H) ^{13}C NMR (75 MHz, $CDCl_3$+DMSO): δ 163.9, 159.9, 147.3, 132.7, 127.7, 127.0, 117.2, 113.8, 113.0, 67.0, 54.4; IR (KBr): ν_{max} 3443, 2924, 2853, 1656, 1612, 1511, 1385, 1247, 757; MS (ESI): 255 [M+H]; HRMS Calcd for $C_{15}H_{15}N_2O_2$: 255.1133. Found: 255.1134.
5c (Table 2): 2-(3-Phenoxyphenyl)-2,3-dihydroquinazolin-4(1H)-*one*: Semi-solid, ^{1}H NMR (300 MHz, $CDCl_3$+DMSO, 3:1): δ 7.87 (brs, 1H), 7.66 (dd, J = 7.7 Hz, 1H), 7.16–7.35 (m, 6H), 7.08 (t, J = 7.6 Hz, 1H), 6.96 (d, J = 8.5 Hz, 2H), 6.91 (brs, 1H), 6.63–6.73 (m, 3H), 5.74 (s, 1H); ^{13}C NMR (75 MHz, $CDCl_3$+DMSO): δ 161.9, 154.8, 154.5, 145.8, 141.9, 131.4, 128.0, 125.5, 121.5, 119.9, 116.7, 115.4, 115.2, 112.6, 64.4; IR (KBr): ν_{max} 3423, 3084, 2929, 2857, 1662, 1485, 1338, 1023, 764; MS (ESI): 317 [M+H]; HRMS Calcd for $C_{20}H_{17}N_2O_2$: 317.1290. Found: 317.1297.
5e (Table 2): 2-(Furan-2-yl)-2,3-dihydroquinazolin-4(1H)-one: Light yellow solid, m.p. 120–121 °C: ^{1}H NMR (300 MHz, $CDCl_3$+DMSO, 3:1): δ 8.06 (d, J = 2.5 Hz, 1H), 7.71 (brs, 1H), 7.67 (dd, J = 7.7 Hz, 1H), 7.19 (t, J = 8.3 Hz, 1H), 6.70 (t, J = 8.1 Hz, 2H), 6.65 (brs, 1H), 6.29 (s, 2H), 5.75 (d, J = 3.2 Hz, 1H); ^{13}C NMR (75 MHz, $CDCl_3$+DMSO): δ 163.9, 152.8, 146.3, 142.0, 141.9, 133.0, 127.3, 117.8, 114.3, 109.7, 107.1, 60.8; IR (KBr): ν_{max} 3416, 3259, 3034, 2921, 1650, 1611, 1467, 1386, 732; MS (ESI): 215 [M+H]; HRMS Calcd for $C_{12}H_{11}N_2O_2$: 215.0820. Found: 215.0830.
12. The catalyst, cellulose-sulfonic acid, was prepared according to the literature procedure: Shaabani, A.; Maleki, A. *Applied Catalysis A: General* **2007**, *331*, 149.