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Ion exchange resins as recyclable and heterogeneous solid acid catalysts for the Biginelli condensation: An improved protocol for the synthesis of 3,4-dihydropyrimidin-2-ones

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Abstract

A simple and efficient protocol has been developed for the synthesis of 3,4-dihydropyrimidin-2-ones using ion exchange resins as recyclable, heterogeneous, environmentally benign catalysts under mild reaction conditions. Among the various solid acid catalysts Nafion NR-50 was found to be most efficient catalyst.

Keywords: Biginelli condensation; 3,4-Dihydropyrimidin-2-ones; Ion-exchange resin; Heterogeneous catalysis

1. Introduction

Replacement of conventional, toxic and polluting Bronsted and Lewis acid catalysts with eco-friendly reusable solid acid heterogeneous catalysts like acidic zeolites, clays, sulfated zirconia and ion exchange resins is an area of current interest [1–5]. The use of solid acid catalyst instead of liquids includes many advantages such as reduced equipment corrosion, ease of product separation, recycling of the catalyst and environmental acceptability. In the recent past ion exchange resins in general and perflorinated resinsulfonic acid (Nafion-H^R) [6,7] in particular, which is strongly acidic and chemically as well as thermally stable has been found to be excellent catalysts for a variety of major organic reactions like alkylation, olefin isomerization, olefin oligomerization, acylation, estrification, etherification, hydration, dehydration and nitration [8–10].

3,4-Dihydropyrimidinones are well known heterocyclic units in the realm of natural and synthetic organic chemistry due to their therapeutic and pharmacological properties including antiviral, antitumor, antibacterial and anti-inflammatory activities [11–13]. Biginelli in 1893 reported for the first time the synthesis of these compounds by the one-pot cyclocondensation of ethyl acetoacetate, benzaldehyde and urea in the presence of strong acid [14] however this method suffers from the drawbacks such as the lower yields of the desired products (20–40%) particularly in case of substituted aldehydes and loss of sensitive functional groups during the reaction. Therefore, in the recent years several improved methodologies mainly using lewis acids [15–20], triflates [21–23], silica-sulfuric acid [24] and silver salts of heteropoly acids [25], silica supported sodium hydrogen sulfate [26], iodine-alumina system [27], poly(4-vinylpyridine-*co*-divinylbenzene)–Cu(II) complex[28], L-proline [29] microwave assisted methodologies [30–32], ultrasounic mediated methods [33] have been reported in the literature. However, inspite of their potential utility many of the existing methods suffers from the drawbacks such as the use of strong acidic conditions, longer reaction times, tedious work-up, environmental disposal problems and lower yields of the products, leaving scope for further development of an efficient and versatile method for Biginelli reaction. In continuation to our studies [34–37] herein, we wish to report for the first time a simple, facile, highly efficient and eco-friendly methodology for the synthesis of 3,4-dihydropyrimidin-2-ones in excellent yields by the reaction of aldehydes, urea and β-dicarbonyl compounds using ion exchange resins as solid heterogeneous catalysts (Scheme 1).

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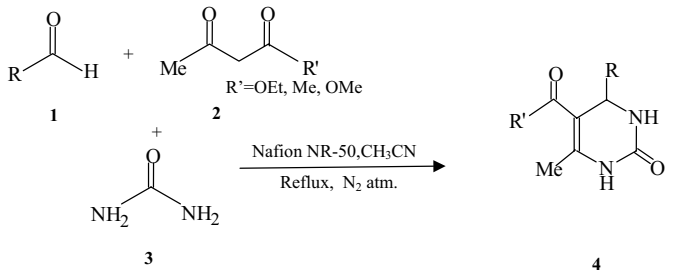

Scheme 1.

2. Results and discussion

To evaluate the catalytic effect of various ion exchange resins we started with the reaction of benzaldehyde, cinnamaldehyde, furfuraldehyde with urea and ethyl acetoacetate in refluxing acetonitrile without and with use of various acidic ion exchange resins as catalysts. These results are presented in Table 1 and clearly indicate that among the various acidic ion exchange resins studied, the Nafion NR-50 was found to be most efficient probably due its strong acidity. To generalize the reaction we have carried out the reactions of various aromatic, aliphatic and heterocyclic aldehydes with urea and β-dicarbonyl compound in presence of catalytic amount of Nafion NR-50 in refluxing acetonitrile under nitrogen atmosphere and these results are presented in Table 2. All the substrates were smoothly converted to their corresponding 3,4-dihydropyrimidinones in excellent yields, with aromatic aldehydes either containing electron donating or electron-withdrawing groups afforded high yields of the products. Similarly aliphatic, heterocyclic and α,β-unsaturated group containing aldehydes afforded good yields of the 3,4-dihydropyridinones without formation of any decomposition or polymerization products under these reaction conditions. The resin catalyst was separated from the reaction mixture by filtration and can be reused several times without any loss of activity, indicate the recylability and reusability of the catalyst. The effect of various solvents was also studied using the benzaldehyde, urea and ethyl acetoacetate as substrates in presence of catalytic amount of Nafion NR-50 and using various solvents such as acetonitrile, ethanol, THF, acetonitrile-water mixture and benzene. Among the various solvents studied, acetonitrile was found to be the most efficient for this transformation.

Table 1
Biginelli condensation using different acidic ion exchange resins as catalysts[a]

Entry	Substrate	Ion exchange resin	Reaction times (h)	Yields[b] (%)
1	Benzaldehyde	–	10	Trace
2	Benzaldehyde	Nafion NR-50	3.0	96
3	Benzaldehyde	Amberlyst-15	5.5	85
4	Benzaldehyde	Amberlyte^R IRP-64	6.0	70
5	Cinnamaldehyde	Nafion NR-50	6.0	74
6	Cinnamaldehyde	Amberlyst-15	7.0	60
7	Cinnamaldehyde	Amberlyte^R IRP-64	7.5	45
8	Furfuraldehyde	Nafion NR-50	5.0	82
9	Furfuraldehyde	Amberlyst-15	6.0	50
10	Furfuraldehyde	Amberlyte^R IRP-64	8.5	40

[a] Reaction conditions: aldehyde (5 mmol), urea (5 mmol), ethyl acetoacetate (5 mmol) in dry acetonitrile (5 ml), ion exchange resin (0.25 g) at refluxing temperature under nitrogen atmosphere.
[b] Isolated yields.

Table 2
Nafion NR-50 catalyzed synthesis of dihydropyrimidin-2 (1H)-ones

Entry	Product	R	R′	Reaction time (h)	Yield[a] (%)
1	4a	C_6H_5	OEt	3.0	96
2	4b	4-$CH_3C_6H_4$	OEt	4.5	87
3	4c	4-$CH_3OC_6H_4$	OEt	5.0	85
4	4d	4-$NO_2C_6H_4$	OEt	2.5	92
5	4e	4-ClC_6H_4	OEt	2.5	92
6	4f	2-ClC_6H_4	OEt	3.0	90
7	4g	4-$(CH_3)_2NC_6H_4$	OEt	6.0	80
8	4h	2,6-$Cl_2C_6H_3$	OEt	2.75	86
9	4i	2-pyridyl	OEt	4.5	80
10	4j	2-Furyl	OEt	5.0	82
11	4k	*n*-$CH_3CH_2CH_2$	OEt	5.0	84
12	4l	$(CH_3)_2CH$	OEt	5.0	85
13	4m	*n*-$CH_3(CH_2)_2CH_2$	OEt	5.5	82
14	4n	$C_6H_5CH=CH$	OEt	6.0	74
15	4o	C_6H_5	CH_3	3.0	94
16	4p	4-$NO_2C_6H_4$	CH_3	3.5	90
17	4q	4-$CH_3OC_6H_4$	CH_3	5.5	80
18	4r	4-ClC_6H_4	CH_3	4.0	87
19	4s	C_6H_5	OMe	3.5	95
20	4t	4-$NO_2C_6H_4$	OMe	4.0	92

[a] Isolated yields.

Although mechanism of this reaction is not clear at this stage, the reaction probably involves acid catalyzed in situ formation of acylimine intermediate by the reaction of urea and aldehyde, which undergoes the subsequent addition to β-dicarbonyl compound followed by cyclization and dehydration to yield dihydropyrimidinone.

3. Experimental

All the solvents and aldehydes were commercially available and purified before use. Nafion NR-50, Amberlyst-15 and Amberlyte^R IRP-64 were purchased from Aldrich.

The melting points were determined in open-capillaries on a Buchi apparatus and are uncorrected. The ^{1}H NMR spectra were recorded on Bruker 300 MHz spectrometer and the chemical shifts are expressed in δ parts per million relative to tetramethylsilane (TMS) as the internal standard. The IR spectra were recorded on a Perkin-Elmer FT-IR X 1760 instrument

3.1. Typical experimental procedure

A solution of benzaldehyde (1 mmol, 107 mg), urea (1 mmol, 60 mg) and ethylacetoacetate (1 mmol, 130 mg), Nafion NR-50 (250 mg) in dry acetonitrile (5 ml) was stirred and refluxed for 3 h under nitrogen atmosphere. The progress of the reaction was monitored by TLC using ethyl acetate/hexane (4:6) as eluent. After completion the reaction, the mixture was cooled to room temperature, the solid that separated was collected by filtration, washed with water and recrystallized from ethanol to give 5-ethoxycarbony-6-methyl-4-phenyl-3,4-

dihydropyrimidin-2-one in 96% yield, mp 201 °C (202 °C) [38]. Similarly other aldehydes were reacted with urea and β-dicarbonyl compound and their reaction times and yields are presented in Table 2. The physical and spectral data of all the products are given in the following section.

5-(Ethoxycarbony)-6-methyl-4-phenyl-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4a): mp 201 °C (202 °C)[38]; IR (KBr): 3240, 1725, 1635 cm^{-1}; ^1H NMR: $\delta = 9.20$ (s, 1H, N**H**), 7.75 (s, 1H, N**H**), 7.10–7.28 (m, 5H, arom C**H**), 5.14 (s, 1H, C**H**), 3.97 (q, 2H, OC**H**$_2$), 2.25 (s, 3H, C**H**$_3$), 1.09 (t, 3H, $OCH_2C\mathbf{H}_3$).

5-(Ethoxycarbonyl)-6-methyl-4-(4-methylphenyl)-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4b): mp 214–15 °C (215 °C) [38]; IR (KBr): 3242, 1715, 1633 cm^{-1}; ^1H NMR: $\delta = 9.16$ (s, 1H, N**H**), 7.80 (s, 1H, N**H**), 7.16–7.12 (m, 4H, arom C**H**), 5.09 (s, 1H, C**H**), 3.96 (q, 2H, OC**H**$_2$CH$_3$), 2.27 (s, 3H, C_6H_4-C**H**$_3$), 2.21 (s, 3H, C**H**$_3$), 1.08 (t, 3H, $OCH_2C\mathbf{H}_3$).

5-(Ethoxycarbonyl)-4-(4-methoxyphenyl)-6-methyl-3,4-dihydropyrimidine-2(1H)-one (Table 2, entry 4c): mp 199 °C (201 °C) [38]; IR (KBr): 3241, 1718, 1636 cm^{-1}; ^1H NMR: $\delta = 9.15$ (s, 1H, N**H**), 7.76 (s, 1H, N**H**), 7.15 (d, 2H, arom C**H**), 6.92 (d, 2H, arom C**H**), 5.12 (s, 1H, C**H**), 3.96 (q, 2H, OC**H**$_2$), 3.76 (s, 3H, C_6H_4-OC**H**$_3$), 2.24 (s, 3H, C**H**$_3$), 1.10 (t, 3H, C**H**$_3$).

5-(Ethoxycarbonyl)-6-methyl-4-(4-nitrophenyl)-3,4-dihydropyrimidin-2(1H)-one (Table 2,entry 4d): mp 207–08 °C (210 °C) [38]; IR (KBr): 3239, 1724, 1645, cm^{-1}; ^1H NMR: $\delta = 9.27$ (s, 1H, N**H**), 8.20 (d, 2H, arom C**H**), 7.91 (s, 1H, N**H**), 7.50 (d, 2H, arom C**H**), 5.18 (d, 1H, C**H**), 3.79 (q, 2H, OC**H**$_2$), 2.25 (s, 3H, $OCH_2C\mathbf{H}_3$), 1.10 (t, 3H, C**H**$_3$).

4-(4-Chlorophenyl)-5-(ethoxycarbonyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one(Table 2, entry 4e): mp 210–12 °C (212–13 °C) [38]; IR (KBr): 3240, 1723, 1643 cm^{-1}; ^1H NMR: $\delta = 9.26$ (s, 1, N**H**), 7.79 (s, 1H, N**H**), 7.40–7.30 (m, 4H, arom C**H**), 5.14 (s, 1H, C**H**), 3.99 (q, 2H, OC**H**$_2$), 2.25 (s, 3H, C**H**$_3$), 1.09 (t, 3H, $OCH_2C\mathbf{H}_3$).

4-(2-Chlorophenyl)-5-ethoxycarbonyl-6-methyl-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4f): mp 222–23 °C (223–24 °C) [38]; IR (KBr): 3242, 1715, 1645 cm^{-1}; ^1H NMR: $\delta = 9.24$ (s, 1H, N**H**), 7.80 (s, 1H, N**H**), 7.42–7.39 (m, 4H, arom C**H**), 5.12 (s, 1H, C**H**), 3.87 (q, 2H, OC**H**$_2$CH$_3$), 2.26 (s, 3H, C**H**$_3$), 1.07 (t, 3H, $OCH_2C\mathbf{H}_3$).

4-(4-*N*,*N*-Dimethylaniline)-5-(ethoxycarbonyl)-6-methyl-3,4-dihydropyrimidine-2 (1H)-one (Table 2, entry 4 g): mp 250 °C; IR (KBr): 3240, 1715, 1650 cm^{-1}; ^1H NMR: $\delta = 9.20$ (s, 1H, N**H**), 7.75 (s, 1H,N**H**), 7.20–7.14 (m, 4H, arom C**H**), 5.16 (s, 1H, C**H**), 3.92 (q, 2H, OC**H**$_2$), 3.31 (s, 6H, N(C**H**$_3$)$_2$), 2.22 (s, 3H, C**H**$_3$), 1.10 (t, 3H, $OCH_2C\mathbf{H}_3$).

4-(2,6-Dichlorophenyl)-5-(ethoxycarbonyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4h): mp 226 °C; IR (KBr): 3235, 1710, 1635 cm^{-1}; ^1H NMR: $\delta = 9.18$ (s, 1H, N**H**), 7.72 (s, 1H, N**H**), 7.31–7.28 (m, 3H, arom C**H**), 5.15 (s, 1H,C**H**), 3.89 (q, 2H, OC**H**$_2$), 2.21 (s, 3H, C**H**$_3$), 1.08 (t, 3H, $OCH_2C\mathbf{H}_3$).

5-(Ethoxycarbonyl)-4-(2-pyridyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4i): mp 193–94 °C (194–95 °C) [38] IR (KBr): 3242, 1714, 1642 cm^{-1}; ^1H NMR: $\delta = 9.13$ (s, 1H, N**H**), 8.25 (s, 1H, N**H**), 7.71 (m, 1H, pyridyl C**H**), 7.51–7.25 (m, 3H, pyridyl C**H**), 5.31 (s, 1H, C**H**),4.02 (q, 2H, OC**H**$_2$), 2.21 (s, 3H, C**H**$_3$), 1.07 (s, 3H, $OCH_2C\mathbf{H}_3$).

5-(Ethoxycarbonyl)-4-(2-furyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4j): mp 202–04 °C (204.5–05 °C) [38]; IR (KBr): 3239, 1705, 1644 cm^{-1}; ^1H NMR: $\delta = 9.22$ (s, 1H, N**H**), 7.74 (s, 1H, N**H**), 7.53 (d, 1H, furyl C**H**), 6.30–6.08 (d, 2H, furyl-C**H**), 5.20 (s, 1H, C**H**), 4.02 (q, 2H, C**H**$_2$CH$_3$), 2.22 (s, 3H, C**H**$_3$), 1.12 (t, 3H, $OCH_2C\mathbf{H}_3$).

5-(Ethoxycarbonyl)-6-methyl-4-(*n*-propyl)-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4k): mp 152–54 °C (153–55 °C) [38]; IR (KBr): 3247, 1709, 1646 cm^{-1}; ^1H NMR: $\delta = 9.01$ (s, 1H, N**H**), 7.52 (s, 1H, N**H**), 4.85 (t, 1H, C**H**), 3.89 (q, 2H, OC**H**$_2$), 2.28 (s, 3H, C**H**$_3$), 1.52–1.42 (m, 4H, -C**H**$_2$C**H**$_2$), 1.28 (t, 3H, C**H**$_3$), 0.90 (t, 3H, -(CH2)$_2$C**H**$_3$).

5-(Ethoxycarbonyl)-6-methyl-4-(*iso*-propyl)-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4l): mp 194 °C (193–94 °C) [38]; IR (KBr): 3234, 2985, 1710, 1642 cm^{-1}; ^1H NMR: $\delta = 8.99$ (s, 1H, N**H**), 7.55 (s, 1H, N**H**), 4.04 (q, 2H, OC**H**$_2$), 4.82 (s, 1H, C**H**), 2.18 (s, 3H, C**H**$_3$), 1.68 (m, 1H, C**H**), 1.07 (t, 3H, $OCH_2C\mathbf{H}_3$), 0.80–0.75 (m, 6H, (C**H**$_2$)$_3$).

4-*n*-Butyl-5-(ethoxycarbonyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4m): mp 157 °C (157–58 °C) [38]; IR (KBr): 3242, 1715, 1647 cm^{-1}; ^1H NMR: $\delta = 9.02$ (s, 1H, N**H**), 7.59 (s, 1H, N**H**), 4.65 (t, 1H, CH), 4.05 (m, 2H, OC**H**$_2$), 2.16 (s, 3H, C**H**$_3$), 1.40–1.15 (m, 6H, -(C**H**$_2$)$_{3-}$), 1.05 (t, 3H, C**H**$_3$), 0.85 (t, 3H, C**H**$_3$).

5-(Ethoxycarbonyl)-6-methyl-4-styryl-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4n): mp 233–34 °C (232–35 °C) [38]; IR (KBr): 3246, 1704, 1650 cm^{-1}; ^1H NMR: $\delta = 9.12$ (s, 1H, N**H**), 7.79 (s, 1H, N**H**), 7.42–7.25 (m, 5H, arom C**H**), 6.33 (d, 1H, HC = C**H**), 6.20 (dd, 1H, C**H** = CH), 4.74 (d, 1H, C**H**), 4.09 (q, 2H, OC**H**$_2$), 2.20 (s, 3H, C**H**$_3$), 1.10 (t, 3H, OCH_2CH_3).

5-Aceto-6-methyl-4-phenyl-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4o): mp 233–34 °C (236 °) [38]; IR (KBr): 3241, 1715, 1643 cm^{-1}; ^1H NMR: $\delta = 9.20$ (s, 1H, N**H**), 7.76 (s, 1H, N**H**), 7.35–7.25 (m, 5H, arom C**H**), 5.25 (s, 1H, C**H**), 2.24 (s, 3H, C**H**$_3$CO), 2.07 (s, 3H, C**H**$_3$).

5-Acetyl-6-methyl-4(4-nitrophenyl)-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4p): mp 234 °C (235–37 °C) [38]; IR (KBr): 3241, 1710, 1640 cm^{-1}; ^1H NMR: $\delta = 9.20$ (s, 1H, N**H**), 8.21 (d, 2H, arom C**H**), 7.93 (s, 1H, N**H**), 7.51 (d, 2H, arom C**H**), 5.32 (s, 1H, C**H**), 2.32 (s, 3H, C**H**$_3$CO), 2.10 (s, 3H, C**H**$_3$).

5-Aceto-6-methyl-4-(4-methoxyphenyl)-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4q): mp 189–90 °C (190–93 °C) [38]; IR (KBr): 3242, 1714, 1642 cm^{-1}; ^1H NMR: $\delta = 9.16$ (s, 1H, N**H**), 7.78 (s, 1H, N**H**), 7.16 (d, 2H, arom C**H**), 6.88 (d, 2H, arom C**H**), 5.30 (s, 1H, C**H**), 3.82 (s, 3H, OC**H**$_3$), 2.32 (s, 3H, C**H**$_3$CO), 2.10 (s, 3H, C**H**$_3$).

5-Aceto-4-(4-chlorophenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4r): mp 232 °C; IR (KBr): 3232, 1708, 1640 cm^{-1}; ^1H NMR: $\delta = 9.22$ (s, 1H, N**H**), 7.78 (s, 1H, N**H**), 7.38 (d, 2H, arom C**H**), 7.25 (d, 2H, arom C**H**), 5.16 (s, 1H, C**H**), 2.30 (s, 3H, C**H**$_3$CO), 2.18 (s, 3H, C**H**$_3$).

5-(Methoxycarbonyl)-6-methyl-4-phenyl-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4s): mp 207–08 °C; IR (KBr): 3231, 1700, 1641 cm^{-1}; ^1H NMR: $\delta = 9.23$ (s, 1H, N**H**), 7.77 (s, 1H, N**H**), 7.35–7.25 (m, 5H, arom C**H**), 5.15 (d, 1H, C**H**), 3.53 (s, 3H, OC$\mathbf{H}_3$), 2.20 (s, 3H, C$\mathbf{H}_3$).

5-(Methoxycarbonyl)-4-(4-nitrophenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (Table 2, entry 4t): mp 235–36 °C (235–37 °C) [38]; IR (KBr): 3232, 1711, 1692 cm^{-1}; ^1H NMR: $\delta = 9.30$ (s, 1H, N**H**), 8.21 (d, 2H, arom C**H**), 7.88 (s, 1H, N**H**), 7.51 (d, 2H, arom C**H**), 5.29 (s, 1H, C**H**), 3.54 (s, 3H, $\mathbf{CH}_3$OCO), 2.27 (s, 3H, C$\mathbf{H}_3$).

4. Conclusion

In conclusion we have developed a simple, efficient, environmentally benign and improved protocol for the synthesis of 3,4-dihydropyrimidin-2-ones in excellent yields. The simplicity of the system, ease of separation/reuse of the catalyst due to its heterogeneous nature, excellent yields of the products and ease of work-up make this method an attractive, environmentally acceptable synthetic tool for Biginelli condensation.

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