Supporting Information

AN IONIC LIQUID-BASED GREEN SYNTHESIS STRATEGY: SYNTHESIS OF DIHYDROPYRIMIDINONES BY THREE-COMPONENT BIGINELLI-TYPE REACTION OF ALIPHATIC ALDEHYDES, AROMATIC ALDEHYDES AND UREA

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1. The recycle of IL ........................................................................................................................................1
2. Characterization of compounds..................................................................................................................2
   2.1 1H-NMR and 13C-NMR data of target products......................................................................................2
   2.2 1H-NMR and 13C-NMR spectra for target products..................................................................................7
1. The recycle of IL

In order to implement the concept of green chemistry and reduce energy consumption, the recycling of ionic liquids was also examined. The results are shown in Table S1. As the number of cycles increased from 1 to 4, the yield decreased only slightly, which shows that the ionic liquid still has a catalytic effect on the reaction after the cycle.

Table S1. The recycle of IL

<table>
<thead>
<tr>
<th>Entry</th>
<th>Cycle numbers</th>
<th>Yield(%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1</td>
<td>85</td>
</tr>
<tr>
<td>2</td>
<td>2</td>
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<td>3</td>
<td>3</td>
<td>83</td>
</tr>
<tr>
<td>4</td>
<td>4</td>
<td>79</td>
</tr>
</tbody>
</table>

*Reaction conditions: 4-nitrobenzaldehyde (1 mmol), n-hexanal (1.5 mmol), methylurea (1.5 mmol), 90°C, 6 h, 1 mL 30% IL aqueous solution. *Yields of pure products isolated by chromatography.
2. Characterization of compounds

2.1 $^1$H-NMR, $^{13}$C-NMR data of DHPMs products

1-methyl-4-(4-nitrophenyl)-5-propyl-3,4-dihydropyrimidin-2(1H)-one

Yellow solid, $^1$H NMR (500 MHz, CDCl$_3$) $\delta$ 8.20 (d, $J = 8.5$ Hz, 2H), 7.46 (d, $J = 8.5$ Hz, 2H), 5.82 (s, 1H), 5.46 (s, 1H), 5.03 (s, 1H), 3.08 (s, 3H), 1.86 – 1.54 (m, 2H), 1.34 – 1.11 (m, 2H), 0.85 (t, $J = 7.3$ Hz, 3H). $^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$ 153.48, 149.99, 147.72, 129.37, 127.99, 125.21, 124.23, 59.06, 39.21, 31.63, 22.63, 14.15.

1-methyl-4-(3-nitrophenyl)-5-propyl-3,4-dihydropyrimidin-2(1H)-one

Yellow solid, $^1$H NMR (500 MHz, CDCl$_3$) $\delta$ 8.09 (dt, $J = 3.5$, 1.5 Hz, 2H), 7.59 (d, $J = 7.8$ Hz, 1H), 7.48 (t, $J = 7.9$ Hz, 1H), 5.79 (s, 1H), 5.42 (s, 1H), 4.99 (s, 1H), 3.03 (s, 3H), 1.65 (t, $J = 7.8$ Hz, 2H), 1.41 – 1.27 (m, 2H), 0.79 (t, $J = 3.5$ Hz, 3H). $^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$ 153.50, 150.01, 147.74, 129.39, 128.01, 124.25, 123.70, 113.02, 59.08, 34.48, 31.65, 22.65, 14.17.

1-methyl-4-(2-nitrophenyl)-5-propyl-3,4-dihydropyrimidin-2(1H)-one

Yellow solid, $^1$H NMR (500 MHz, CDCl$_3$) $\delta$ 7.86 (d, $J = 7.1$ Hz, 1H), 7.59 (t, $J = 7.6$ Hz, 1H), 7.52 (d, $J = 7.8$ Hz, 1H), 7.40 (t, $J = 7.7$ Hz, 1H), 6.01 (s, 1H), 5.57 (s, 1H), 5.27 (s, 1H), 3.03 (s, 3H), 1.71 – 1.65 (m, 2H), 1.26 – 1.16 (m, 2H), 0.79 (t, $J = 7.3$ Hz, 3H). $^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$ 153.48, 149.99, 147.72, 129.37, 127.99, 125.21, 124.23, 113.00, 59.06, 34.46, 31.63, 22.63, 14.15.
4-(1-methyl-2-oxo-5-propyl-1, 2, 3, 4-tetrahydropyrimidin-4-yl)benzonitrile

Yellow solid. $^1$H NMR (500 MHz, CDCl$_3$) δ 7.53 (d, $J$ = 7.6 Hz, 2H), 7.30 (t, $J$ = 17.5 Hz, 2H), 6.37 (s, 1H), 5.72 (s, 1H), 4.86 (s, 1H), 2.90 (s, 3H), 1.60 (t, $J$ = 14.3 Hz, 2H), 1.25 (ddd, $J$ = 20.6, 13.2, 6.7 Hz, 2H), 0.76 (t, $J$ = 6.9 Hz, 3H). $^{13}$C NMR (126 MHz, CDCl$_3$) δ 153.51, 147.75, 129.40, 128.02, 125.24, 124.26, 116.45, 113.03, 59.09, 34.49, 31.66, 22.66, 14.18.

5-isopropyl-1-methyl-4-(4-nitrophenyl)-3,4-dihydropyrimidin-2(1H)-one

Yellow solid. $^1$H NMR (500 MHz, CDCl$_3$) δ 8.11 (d, $J$ = 8.7 Hz, 2H), 7.40 (d, $J$ = 8.7 Hz, 2H), 6.40 (s, 1H), 5.81 (s, 1H), 5.00 (s, 1H), 3.01 (s, 3H), 1.89 (dt, $J$ = 13.5, 6.7 Hz, 1H), 0.93 (d, $J$ = 6.8 Hz, 3H), 0.89 (d, $J$ = 6.8 Hz, 3H). $^{13}$C NMR (126 MHz, CDCl$_3$) 153.49, 150.00, 147.73, 129.38, 128.00, 125.22, 124.24, 59.07, 34.47, 31.64, 22.64.

5-butyl-1-methyl-4-(4-nitrophenyl)-3,4-dihydropyrimidin-2(1H)-one

Yellow solid. $^1$H NMR (500 MHz, CDCl$_3$) δ 8.13 (d, $J$ = 8.7 Hz, 2H), 7.40 (d, $J$ = 8.7 Hz, 2H), 5.79 (s, 1H), 5.75 (s, 1H), 4.97 (s, 1H), 2.99 (s, 3H), 1.67 (t, $J$ = 11.9 Hz, 2H), 1.23 – 1.11 (m, 4H), 0.78 (t, $J$ = 6.2 Hz, 3H). $^{13}$C NMR (126 MHz, CDCl$_3$) δ 153.48, 149.99, 147.72, 127.99, 125.21, 124.23, 123.68, 59.06, 34.46, 31.63, 30.34, 22.63, 14.15.

1-methyl-4-(4-nitrophenyl)-5-pentyl-3,4-dihydropyrimidin-2(1H)-one
Yellow solid, $^1$H NMR (500 MHz, CDCl$_3$) $\delta$ 8.09 (d, $J$ = 8.6 Hz, 2H), 7.39 (d, $J$ = 8.5 Hz, 2H), 6.01 (s, 1H), 5.75 (s, 1H), 4.97 (s, 1H), 2.98 (s, 3H), 1.64 (t, $J$ = 7.5 Hz, 2H), 1.09 (d, $J$ = 10.0 Hz, 6H), 0.76 (t, $J$ = 6.9 Hz, 3H). $^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$ 153.48, 149.99, 147.72, 127.99, 125.21, 124.23, 113.00, 59.06, 34.46, 31.63, 30.34, 26.99, 22.63, 14.15.

5-hexyl-1-methyl-4-(4-nitrophenyl)-3,4-dihydropyrimidin-2(1H)-one

Yellow solid, $^1$H NMR (500 MHz, CDCl$_3$) $\delta$ 8.18 (d, $J$ = 8.4 Hz, 2H), 7.44 (d, $J$ = 8.4 Hz, 2H), 5.88 (s, 1H), 5.80 (s, 1H), 5.02 (s, 1H), 3.05 (s, 3H), 1.70 (t, $J$ = 7.2 Hz, 2H), 1.18 (d, $J$ = 7.3 Hz, 8H), 0.83 (t, $J$ = 6.9 Hz, 3H). $^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$ 153.40, 149.91, 147.64, 127.91, 125.13, 124.15, 112.92, 58.98, 34.38, 31.56, 30.27, 28.78, 26.91, 22.55, 14.07.

4-(4-methoxyphenyl)-1-methyl-5-propyl-3,4-dihydropyrimidin-2(1H)-one

Yellow oily liquid, $^1$H NMR (500 MHz, CDCl$_3$) $\delta$ 7.12 (d, $J$ = 8.6 Hz, 2H), 6.78 (d, $J$ = 8.6 Hz, 2H), 5.68 (s, 1H), 5.11 (s, 1H), 4.80 (s, 1H), 3.72 (s, 3H), 2.99 (s, 3H), 1.69 – 1.56 (m, 2H), 1.31 (dd, $J$ = 14.6, 7.2 Hz, 2H), 1.14 (t, $J$ = 7.0 Hz, 3H). $^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$ 153.46, 149.97, 129.35, 127.97, 125.19, 124.21, 112.98, 59.04, 54.17, 34.44, 31.61, 22.61, 14.13.

4-(4-(dimethylamino)phenyl)-1-methyl-5-propyl-3,4-dihydropyrimidin-2(1H)-one

Yellow solid, $^1$H NMR (500 MHz, CDCl$_3$) $\delta$ 7.05 (d, $J$ = 8.7 Hz, 2H), 6.60 (d, $J$ = 8.7 Hz, 2H), 5.66 (s, 1H), 5.09 (s, 1H), 4.75 (s, 1H), 2.99 (s, 3H), 2.86 (s, 6H), 1.70 – 1.54 (m, 2H), 1.26 – 1.12 (m, 2H), 0.77 (t, $J$ = 7.3 Hz, 3H). $^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$ 153.48, 152.38,
129.37, 127.99, 125.21, 124.23, 113.00, 59.06, 39.02, 34.46, 31.63, 22.63, 14.15.

4-(4-bromophenyl)-1-methyl-5-propyl-3,4-dihydropyrimidin-2(1H)-one

White solid, $^1$H NMR (500 MHz, CDCl$_3$) $\delta$ 7.44 (d, $J$ = 8.3 Hz, 2H), 7.14 (d, $J$ = 8.3 Hz, 2H), 5.75 (s, 1H), 5.34 (s, 1H), 4.87 (s, 1H), 3.04 (s, 3H), 1.78 – 1.59 (m, 2H), 1.45 – 1.25 (m, 2H), 0.83 (t, $J$ = 7.3 Hz, 3H). $^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$ 153.50, 147.74, 129.39, 128.01, 125.23, 124.25, 123.70, 59.08, 34.48, 31.65, 22.65, 14.17.

4-(4-chlorophenyl)-1-methyl-5-propyl-3,4-dihydropyrimidin-2(1H)-one

White solid, $^1$H NMR (500 MHz, CDCl$_3$) $\delta$ 7.30 (d, $J$ = 8.4 Hz, 2H), 7.21 (d, $J$ = 8.4 Hz, 2H), 5.76 (s, 1H), 5.23 (s, 1H), 4.89 (s, 1H), 3.06 (s, 3H), 1.69 (dd, $J$ = 16.9, 8.7 Hz, 2H), 1.38 (dt, $J$ = 14.8, 7.4 Hz, 2H), 0.83 (t, $J$ = 7.3 Hz, 3H). $^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$ 153.48, 147.72, 129.37, 127.99, 125.21, 124.23, 123.68, 59.06, 34.46, 31.63, 22.65, 14.15.

4-(4-bromophenyl)-1-ethyl-5-propyl-3,4-dihydropyrimidin-2(1H)-one

White solid, $^1$H NMR (500 MHz, CDCl$_3$) $\delta$ 7.47 (d, $J$ = 8.3 Hz, 2H), 7.16 (d, $J$ = 8.5 Hz, 2H), 5.80 (s, 1H), 5.29 (s, 1H), 4.87 (s, 1H), 3.61 – 3.30 (m, 2H), 1.79 – 1.61 (m, 2H), 1.50 – 1.27 (m, 2H), 1.19 (t, $J$ = 7.1 Hz, 3H), 0.86 (t, $J$ = 7.4 Hz, 3H). $^{13}$C NMR (126 MHz, CDCl$_3$) $\delta$ 152.74, 142.10, 131.90, 128.70, 123.16, 121.95, 113.61, 59.01, 41.62, 32.48, 20.23, 14.24, 13.67.

1-ethyl-5-propyl-4-(p-tolyl)-3,4-dihydropyrimidin-2(1H)-one

5
Yellow solid, $^1$H NMR (500 MHz, CDCl$_3$) δ 7.18 (d, $J = 13.8$ Hz, 4H), 5.79 (s, 1H), 4.87 (s, 1H), 3.94 (s, 1H), 3.62 – 3.40 (m, 2H), 2.35 (s, 3H), 1.85 – 1.63 (m, 2H), 1.48 – 1.30 (m, 2H), 1.22 (t, $J = 7.1$ Hz, 3H), 0.86 (t, $J = 6.2$ Hz, 3H). $^{13}$C NMR (126 MHz, CDCl$_3$) δ 152.82, 140.13, 137.83, 129.32, 126.86, 122.59, 114.38, 59.35, 41.57, 32.55, 21.06, 20.23, 14.26, 13.69.

1-ethyl-4-(4-nitrophenyl)-5-propyl-3,4-dihydropyrimidin-2(1$H$)-one

Yellow solid, $^1$H NMR (500 MHz, CDCl$_3$) δ 8.13 (d, $J = 8.6$ Hz, 2H), 7.38 (d, $J = 8.6$ Hz, 2H), 5.78 (s, 1H), 5.59 (s, 1H), 4.94 (s, 1H), 3.47 – 3.37 (m, 2H), 1.72 – 1.57 (m, 2H), 1.38 – 1.31 (m, 2H), 1.12 (t, $J = 7.1$ Hz, 3H), 0.79 (t, $J = 7.3$ Hz, 2H). $^{13}$C NMR (126 MHz, CDCl$_3$) δ 152.71, 150.04, 147.64, 127.86, 124.16, 123.80, 112.89, 58.87, 41.73, 32.45, 20.25, 14.23, 13.63.
$^1$H-NMR, $^{13}$C-NMR spectra for DHPMs products

$^1$H NMR spectra of compound 4a

$^{13}$C NMR spectra of compound 4a
$^1$H NMR spectra of compound 4b

$^{13}$C NMR spectra of compound 4b
$^1$H NMR spectra of compound 4c

$^{13}$C NMR spectra of compound 4c
$^1$H NMR spectra of compound 4d

$^{13}$C NMR spectra of compound 4d
$^1$H NMR spectra of compound 4e

$^{13}$C NMR spectra of compound 4e
$^1$H NMR spectra of compound 4f

$^{13}$C NMR spectra of compound 4f
$^1$H NMR spectra of compound 4g

$^{13}$C NMR spectra of compound 4g
$^1$H NMR spectra of compound $4h$

$^{13}$C NMR spectra of compound $4h$
$^1$H NMR spectra of compound 4i

$^{13}$C NMR spectra of compound 4i
$^1$H NMR spectra of compound 4j

$^{13}$C NMR spectra of compound 4j
$^1$H NMR spectra of compound 4k

$^{13}$C NMR spectra of compound 4k
$^1$H NMR spectra of compound 4l

$^{13}$C NMR spectra of compound 4l
$^1$H NMR spectra of compound 4m

$^{13}$C NMR spectra of compound 4m
$^1$H NMR spectra of compound 4n

$^{13}$C NMR spectra of compound 4n
$^1$H NMR spectra of compound 4o

$^{13}$C NMR spectra of compound 4o