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A fast, simple and convenient procedure for the synthesis of fused pyrimidinone derivatives by using [Hmim][HSO_4] as a green, efficient and reusable catalyst under solvent-free conditions

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ABSTRACT

In the present of methylimidazolium hydrogen sulfate, the synthesis of arylidene heterobicyclic pyrimidinones is studied by condensation of aromatic aldehyde, cyclopentanone, and urea or thiourea. Using solvent-free conditions, non-toxic and inexpensive materials, simple and clean work-up, short reaction times and good yields of the products are the advantages of this method.

***Keywords**: Pyrimidinone, Methylimidazolium hydrogen sulfate, Bronsted acidic ionic liquid, Biginelli-type reaction.*

1. Introduction

In recent decades, multicomponent reactions (MCRs), have played an increasingly important role in organic and medicinal chemistry [1-4]. Moreover, this approach is known as an important, economical and environmentally benign process in synthetic chemistry because it decreases the number of reaction steps, energy consumption and waste [5,6]. It is known that pyrimidinone and its derivatives are used in various pharmaceutical and biochemical fields [7,8]. Therefore, an interest for the synthesis of 3,4-dihydropyrimidin-2(1*H*)-ones (DHPMs) and their derivations is tremendously increasing [9]. One of the most important functionalized pyrimidinones are fused derivatives with an arylidene group. These heterobicyclic compounds are significant intermediates for the preparation of many biologically active products. For example, some of them show a broad-spectrum antitumor activity [10a]. Previously, they were synthesized by the reaction of α, α'-bis (arylidene) cycloalkanones with urea or thiourea (Scheme 1). In most cases, using strong Brönsted acid such as HCl, or base such as sodium alkoxide or potassium hydroxide was necessary for the progression of the reaction [10-12].

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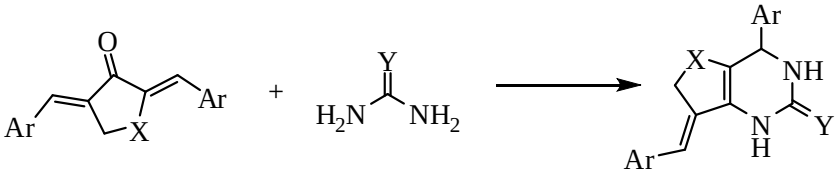
Recently, Pan and coworkers have reported an efficient one-pot method for the synthesis of these fused pyrimidinones by condensation of aromatic aldehyde, cyclopentanone, and urea or thiourea as the starting materials named as Biginelli-type reaction [13]. Using stoichiometric amounts of TMSCl as a condensation reagent, and mixed DMF/CH_3CN as the reaction solvent is necessary to progress the reaction. More recently, lanthanide Lewis acids have been studied as the catalysts for the one-pot synthesis of these pyrimidinone derivatives under solvent-free conditions [14].

In recent times, using ionic liquids (ILs) is increasing with a very fast rate because of their beneficial properties such as undetectable vapor pressure, non-inflammability, wide liquid range, reusability and high thermal stability [15-17]. A subdivision of ILs is protic ionic liquids (PILs), which are produced through the combination of a Brönsted acid and Brönsted base [18]. These acidic ionic liquids have widely been applied in electrochemistry, synthesis of nanostructure materials, reaction media and catalyst. Furthermore, Brønsted acidic ionic liquids can be designed to replace traditional mineral liquid acids such as sulfuric acid and hydrochloric acid in organic synthesis. Although, there are various Brønsted acidic ionic liquids applied in organic synthesis [18], the majority of them are synthesized with a time-consuming and expensive procedure. Methyl imidazolium hydrogen sulfate ([Hmim][HSO_4]) has easily



X = -CH_2-, -CH_2CH_2-, -N(CH_3)-CH_2-
Y = O, S

Scheme 1

been synthesized and used as an efficient, inexpensive and reusable catalyst in organic synthesis [19-22]. Moreover, the present ionic liquid is halogen free and because of less carbon numbers, this ionic liquid has less toxicity [23]. Therefore, [Hmim][HSO_4] can be introduced as a green ionic liquid.

Herein we report, an efficient and convenient procedure for the synthesis of these arylidene heterobicyclic pyrimidinones by the one-pot three-component condensation of aromatic aldehyde, cyclopentanone, and urea or thiourea in the presence of [Hmim]HSO_4 as a green and economical catalyst under solvent-free conditions (Scheme 2).

2. Experimental

All reagents were purchased from Merck and Aldrich and used without further purification. All yields refer to the isolated products after purification. The products were characterized by comparison with authentic samples and by spectroscopic data (IR, ^{1}H NMR, ^{13}C NMR spectra and melting point). All melting points were taken on a Gallenkamp melting apparatus and were uncorrected. IR spectra were recorded on a JASCO FT/IR-680 PLUS spectrometer. ^{1}H NMR spectra were recorded on a Bruker 400 MHz.

2.1. General Procedure for the Synthesis of arylidene heterobicyclic pyrimidinones

A mixture of the aldehyde (1 mmol), cyclopentanone (1.1 mmol), urea or thiourea (1.2 mmol) and methylimidazolium hydrogen sulfate (0.25 mmol) [19] was heated in an oil bath at 110 °C for the specified time. The reaction was followed by TLC (EtOAc/cyclohexane, 50:50). After completion of reaction, the mixture was cooled to room temperature and then water was added to the reaction mixture and stirred for 5–10 min. The product was collected by filtration, washed with water and then washed with ethanol to afford the pure product.

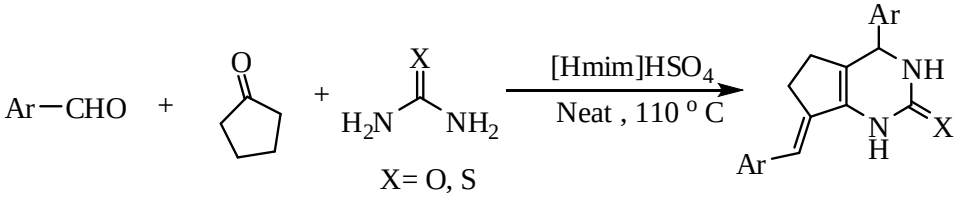
The spectral data of some representative products of arylidene heterobicyclic pyrimidinones

7-(benzylidene)-3,4,6,7-tetrahydro-4-phenyl-1*H*-cyclopenta[*d*]pyrimidin-2(5*H*)-one (Table 2, entry 1): ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ: 8.76 (1 H, s), 7.14-7.40 (11 H, m), 6.62 (1 H, s), 5.15 (1 H, s), 2.78-2.90 (2 H, m), 2.31-2.41 (1 H, m), 1.94-2.05 (1 H, m). IR (KBr, cm^{-1}): 3410, 3215, 3118, 2922, 2848, 1672, 1467, 1444, 1353, 1071, 754.

7-(2-chlorobenzylidene)-4-(2-chlorophenyl)-3,4,6,7-tetrahydro-1*H*-cyclopenta[*d*]pyrimidin-2(5*H*)-one (Table 2, entry 2): ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ: 9.08 (1 H, s), 7.17-7.54 (9 H, m), 6.76 (1 H, s), 5.62 (1 H, s), 2.65-2.80 (2 H, m), 2.37-2.45 (1 H, m), 1.92-2.01 (1 H, m). IR (KBr, cm^{-1}): 3421, 3229, 3115, 2925, 2851, 1677, 1616, 1492, 1459, 1439, 1037, 867, 813, 754.

7-(4-(methoxycarbonyl)benzylidene)-3,4,6,7-tetrahydro-4-(4-methoxycarbonyl)phenyl)-1*H*-cyclopenta[*d*]pyrimidin-2(5*H*)-one (Table 2, entry 7): ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ: 8.91 (1 H, s), 7.98 (2 H, d, *J* = 7.20 Hz), 7.91 (2 H, d, *J* = 7.60 Hz), 7.43 (4 H, m), 7.33 (1 H, s), 6.72 (1 H, s), 5.29 (1 H, s), 3.83 (6 H, s), 2.76-2.94 (2 H, m), 2.38-2.45 (1 H, m), 1.95-2.05 (1 H, m). ^{13}C NMR (100 MHz, DMSO-d_6, ppm) δ: 166.45, 153.61, 148.72, 142.90, 142.52, 136.76, 130.09, 129.89, 129.31, 128.36, 127.29, 127.11, 120.15, 116.60, 57.66, 52.56, 52.43, 29.11, 28.76. IR (KBr, cm^{-1}): 3382, 3219, 3119, 2950, 2850, 1688, 1599, 1432, 1282, 1108, 898, 770. Anal. Calcd for $C_{24}H_{22}N_2O_5$: C, 68.89; H, 5.30; N, 6.69. Found: C, 68.80; H, 5.41; N, 6.70.

7-(4-fluorobenzylidene)-4-(4-fluorophenyl)-3,4,6,7-tetrahydro-1*H*-cyclopenta[d]pyrimidin-2(5H)-one (Table 2, entry 8): ^{1}H NMR (400 MHz, DMSO-*d6*) δ: 8.77 (1 H, s),



Scheme 2.

7.27-7.38 (4H, m), 7.13-7.22 (5 H, m), 6.62 (1 H, s), 5.17 (1 H, s), 2.71-2.85 (2 H, m), 2.33-2.40 (1 H, m), 1.95-2.02 (1 H, m). IR (KBr, cm^{-1}): 3415, 3220, 3123, 2987, 2918, 1681, 1603, 1507, 1455, 1352, 1229, 1157, 1076, 886, 825, 755.

3,4,6,7-tetrahydro-4-(naphthalene-2-yl)-7- ((naphtha lene - 2-yl)methylene)-1H-cyclopenta[d]pyrimidin-2(5H)-one (Table 2, entry 10): ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ: 8.89 (1 H, s), 7.77-7.97 (8 H, m), 7.41-7.55 (6 H, m), 7.33 (1 H, s), 6.83 (1 H, s), 5.36 (1 H, s), 2.85-3.05 (2 H, m), 2.28-2.35 (1 H, m), 1.98-2.08 (1 H, m). ^{13}C NMR (100 MHz, DMSO-d_6, ppm) δ: 154.10, 141.62, 140.72, 137.11, 136.25, 134.11, 133.73, 133.41, 132.33, 129.33, 128.71, 128.45, 128.26, 127.63, 127.23, 127.10, 126.87, 126.50, 125.82, 125.77, 119.64, 117.68, 58.60, 29.46, 29.25 IR (KBr, cm^{-1}): 3433, 3207, 3107, 2923, 2849, 1671, 1615, 1507, 1457, 1360, 1123, 901, 813, 745. Anal. Calcd for C28H22N2O: C, 83.56; H, 5.51; N, 6.96. Found: C, 83.42; H, 5.64; N, 6.88.

7-(4-methybenzylidene)-3,4,6,7-tetrahydro-4-p-tolyl-1H-cyclopenta[d]pyrimidin-2(5H)-one (Table 2, entry 11): ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ: 8.70 (1 H, s), 7.11-7.25 (9 H, m), 6.57 (1 H, s), 5.08 (1 H, s), 2.73-2.82 (2 H, m), 2.31-2.39 (1 H, m), 2.27 (6 H, s), 1.94-2.02 (1 H, m). IR (KBr, cm^{-1}): 3445, 3217, 3119, 2917, 2847, 1687, 1512, 1458, 1348, 1081, 889, 808, 750.

7-(4-methoxybenzylidene)-3,4,6,7-tetrahydro-4-(4-methoxyphenyl)-1H-cyclopenta[d]pyrimidin-2(5H)-one (Table 2, entry 14): ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ: 8.65 (1 H, s), 7.26 (2 H, d, J = 8.70 Hz), 7.17 (2 H, d, J = 8.50 Hz), 7.08 (1 H, s), 6.75-6.87 (4 H, m), 6.55 (1 H, s), 5.07 (1 H, s), 3.74 (3 H, s), 3.73 (3 H, s), 2.68-2.83 (2 H, m), 2.32-2.38 (1 H, m), 1.65-2.02 (1 H, m). IR (KBr, cm^{-1}): 3374, 3214, 3114, 2957, 2932, 2835, 1682, 1604, 1511,

7-(4-chlorobenzylidene)-4-(4-chlorophenyl)-3,4,6,7-tetrahydro-1H-cyclopenta[d]pyrimidin-2(5H)-thione (Table 2, entry 16): ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ: 8.80 (1 H, s), 7.45 (2 H, d, J = 8.36 Hz), 7.41 (2 H, d, J = 8.60 Hz), 7.27-7.34 (4 H, m), 7.23 (1 H, s), 6.62 (1 H, s), 5.18 (1 H, s), 2.71-2.87 (2 H, m), 2.34-2.42 (1 H, m), 1.95-2.02 (1 H, m). IR (KBr, cm^{-1}): 3409, 3223, 3122, 2919, 2850, 1673, 1489, 1453, 1406, 1352, 1272, 1090, 1013, 887, 827, 815, 755.

7-(4-nitrobenzylidene)-3,4,6,7-tetrahydro-4-(4-nitrophenyl)-1H-cyclopenta[d]pyrimidin-2(5H)-thione (Table 2, entry 17): ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ: 10.31 (1 H, s), 9.19 (1 H, s), 8.29 (2 H, d, J = 7.60 Hz), 8.19 (2 H, d, J = 8.40 Hz), 7.51-7.60 (4 H, m), 7.09 (1 H, s), 5.49 (1 H, s), 2.82-2.99 (2 H, m), 2.54-2.61 (1 H, m), 2.06-2.14 (1 H, m). IR (KBr, cm^{-1}): 3387, 3202, 2923, 2848, 1667, 1587, 1518, 1475, 1340, 1181, 1110, 858, 750.

7-(2-methoxybenzylidene)-3,4,6,7-tetrahydro-4-(2-methoxyphenyl)-1H-cyclopenta[d]pyrimidin-2(5H)-thione Table 2, entry 18: ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ: 10.18 (1 H, s), 8.67 (1 H, s), 7.25-7.33 (2 H, m), 7.12-7.21 (2 H, m), 6.87-7.04 (5 H, m), 5.48 (1 H, s), 3.79 (6 H, s), 2.62-2.73 (2 H, m), 2.33-2.42 (1 H, m), 2.01-2.11 (1 H, m). IR (KBr, cm^{-1}): 3423, 3163, 2962, 2900, 2834, 1666, 1594, 1552, 1488, 1461, 1243, 1194, 1178, 1028, 873, 760.

3. Results and discussion

To optimize the reaction conditions, the reaction of 3-nitrobenzaldehyde (1 mmol) with cyclopentanone (1.1 mmol) and urea (1.2 mmol) was studied as a simple model substrate under different conditions (Table 1). It was found that the best result was obtained when the reaction carried out at 110 °C by using 0.25 mmol of methylimidazolium hydrogen sulfate under solvent-free conditions (Table 1, entry 6). In the absence of [Hmim]HSO_4, using the same reaction conditions, the reaction was carried out in low yield even after 90 minutes (Table 1, entry 1). As shown in Table 1, [Hmim]HSO_4 is

Table 1. The reaction of 3-nitrobenzaldehyde, cyclopentanone and urea by using different amount of [Hmim]HSO4 under solvent-free conditions [a].

Entry	[Hmim]HSO_4 (mmol)	Temperature	Time (min)	Yield (%)
1	0	110 °C	90	10
2	0.05	110 °C	60	38
3	0.10	110 °C	60	52
4	0.15	110 °C	45	65
5	0.20	110 °C	20	77
6	0.25	110 °C	20	85
7	0.30	110 °C	20	82
8	0.25	25 °C	90	Trace
9	0.25	80 °C	60	63
10	0.25	100 °C	20	80

[a]The yields refer to the isolated pure products.

Table 2. Synthesis of arylidene heterobicyclic pyrimidinones by using catalytic amount of methylimidazolium hydrogen sulfate [a].

Entry	Ar	X	Time (min)	Yield (%)[a]	m.p (°C)		Ref.
					Found	Reported	
1	C_6H_5-	O	20	80	231-233	236-239	13
2	2-ClC_6H_4-	O	20	78	229-232	232-234	13
3	4-ClC_6H_5-	O	20	86	248-251	252-255	13
4	4-BrC_6H_4-	O	20	80	218-221	221-222	14
5	3-$NO_2C_6H_4$-	O	20	85	232-234	235-239	13
6	4-NCC_6H_4-	O	20	86	221-223	222-225	14
7	4-$CH_3OCOC_6H_4$-	O	20	78	219-222	-	-
8	4-FC_6H_4-	O	20	84	207-210	203-205	14
9	1-Naphtyl	O	45	72	235-237	238-240	14
10	2-Naphtyl	O	45	70	208-211	-	-
11	4-$CH_3C_6H_4$-	O	25	73	234-237	238-241	13
12	2-$CH_3OC_6H_4$-	O	50	62	248-250	250-251	14
13	3-$CH_3OC_6H_4$-	O	40	71	222-224	226-227	14
14	4-$CH_3OC_6H_4$-	O	40	70	245-248	250-252	13
15	C_6H_5-	S	45	77	215-218	219-223	13
16	4-ClC_6H_5-	S	45	81	221-224	226-228	13
17	4-$NO_2C_6H_4$-	S	45	74	202-205	203-207	13
18	2-$CH_3OC_6H_4$-	S	60	60	223-226	223-226	13

[a]The yields refer to isolated pure products which were characterized from their spectroscopic data and by comparison with authentic samples.

necessary for the progression of the reaction. An increase of the amount of the catalyst from 0.25 to 0.30 mmol did not improve the result to a greater extent.

In order to study the generality of the present procedure, different kinds of aromatic aldehydes were treated with cyclopentanone, and urea or thiourea in the presence of [Hmim]HSO_4 under solvent-free conditions (Table 2). Aromatic aldehydes, with electron-donating or electron-withdrawing groups, reacted very well to produce the corresponding arylidene heterobicyclic pyrimidinones in good yields. We observed electronic effect on these reactions so that electron-withdrawing groups on the aromatic aldehydes, in shorter reaction times, increased the yields of the products than those of electron-donating groups. The steric effects of *ortho*-substituents had influence on the reaction time and yield. It was found that *ortho*-substituted benzaldehydes afforded the corresponding fused pyrimidinones in relatively lower yields (Table 2, entries 2, 12 and 18). Moreover, thiourea was successfully used to provide the corresponding products in good yields (Table 2, entries 15-18).

To show the advantage of the present work in comparison with reported results in the literature, we compared results of methyl imidazolium hydrogen sulfate with some catalyst in the synthesis of arylidene heterobicyclic pyrimidinones from aldehyde, cyclopentanone and urea. As shown in Table 4 (entries 1-12), [Hmim][HSO_4] can act as an effective catalyst with respect to reaction times, yields and simplified conditions of the obtained products.

The thermal properties of Methylimidazolium hydrogen sulfate ([Hmim]HSO_4) was evaluated by means of TGA and DSC under argon atmosphere. This ionic liquid exhibited good resistance to thermal decomposition. 10% weight loss of them occurred at around 311 °C. 10 % weight of ionic liquid left after TGA analysis at maximum temperature 600°C (Fig. 1). To study the reusability of the present catalyst, after each run, water was added to the reaction mixture and the product was filtered. To recycle the catalyst, all the water added for filtering and washing the product, was collected and washed with CH_2Cl_2 (3 × 10 ml) to remove organic impurities. Then water was evaporated and the catalyst was dried at 65 °C under reduced pressure for 2 h. This catalyst reused for the reaction of benzaldehyde with cyclopentanone and urea under solvent-free conditions at 110 ºC. As shown in Table 3, the catalyst could be

Table 3. Reusability of the catalyst.

Run	Time (min)	Yield
1	20	80
2	20	76
3	20	70

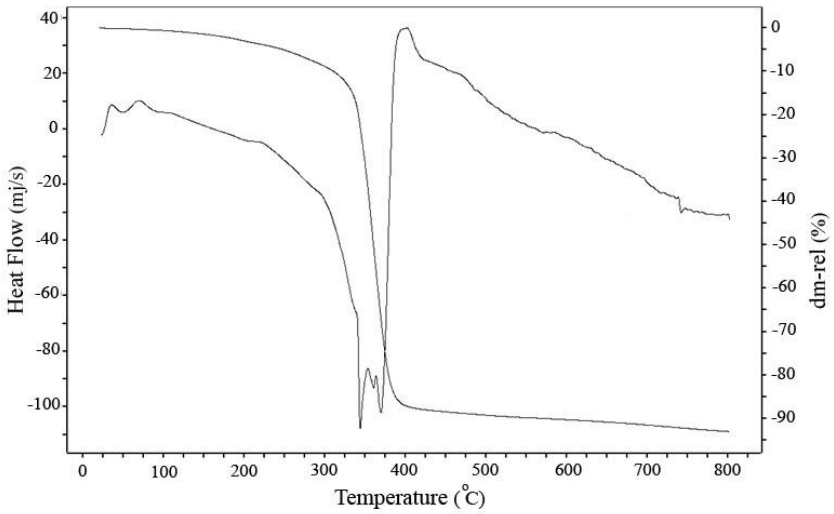


Fig. 1. TGA/ DSC thermogram of methylimidazolium hydrogen sulfate ([Hmim]HSO_4) under argon atmosphere.

employed three times, although its activity gradually decreased.

4. Conclusion

In summary, we introduced methyl imidazolium hydrogen sulfate as an inexpensive, easily available, non-corrosive and environmentally benign catalyst for the synthesis of fused pyrimidinone derivatives by one-pot three component condensation reactions. Using solvent-free conditions, non-toxic and inexpensive materials, simple and clean work-up, short reaction times and high yields of the products are the advantages of this method.

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Table 4. Synthesis of arylidene heterobicyclic pyrimidinones by using different acidic catalysts.

Entry	Catalyst (mol%)	Ar	X	Conditions	Time	Yield (%)	Ref.
1	$YbCl_3$(3)	Ph	O	90 °C, neat	3 h	79	14
2	$YbCl_3$(3)	3-NO_2Ph-	O	90 °C, neat	5h	50	14
3	$YbCl_3$(3)	4-CH_3Ph-	O	90 °C, neat	5h	70	14
4	TMSCl (1)	Ph	O	DMF-CH_3CN, rt	3 h	93	13
5	TMSCl (1)	3-NO_2Ph-	O	DMF-CH_3CN, rt	3 h	95	13
6	TMSCl (1)	4-CH_3Ph-	O	DMF-CH_3CN, rt	3 h	78	13
7	IL (15) [a]	Ph	O	100 °C, neat	5 min	86	9e
8	IL (15)	3-NO_2Ph-	O	100 °C, neat	5 min	88	9e
9	IL (15)	4-CH_3Ph-	O	100 °C, neat	10 min	75	9e
10	[Hmim][HSO_4] (10)	Ph	O	110 °C, neat	20 min	80	-
11	[Hmim][HSO_4] (10)	3-NO_2Ph-	O	110 °C, neat	20 min	85	-
12	[Hmim][HSO_4] (10)	4-CH_3Ph-	O	110 °C, neat	25 min	73	-

[a] IL= $(CH_3CH_2)_3N^+CH_2$ $(CH_2)_2CH_2SO_3H$ HSO_4^-

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