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PIPERAZINE CATALYZED ONE-POT, THREE-COMPONENT SYNTHESIS OF 4*H*-CHROMENE AND 3,4-DIHYDROPYRANO[*c*]CHROMENE DERIVATIVES UNDER SOLVENT-FREE CONDITIONS

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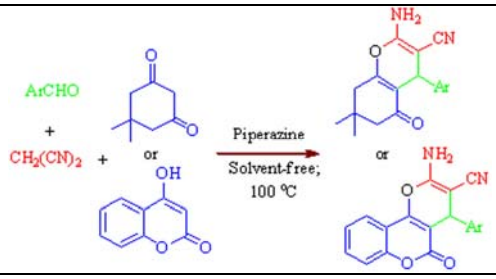
A convenient and efficient method for one-pot synthesis of 4*H*-chromene and 3,4-dihydropyrano[c]chromene derivatives by a multi-component condensation reaction of aromatic aldehydes, malononitrile and dimedone or 4-hydroxycoumarin in the presence of piperazine as a catalyst under solvent-free conditions has been developed. The features of this procedure were characterized by the following: good to high yields, one-pot procedure, short reaction time, and operational simplicity.



INTRODUCTION

Multicomponent reactions (MCRs) is a valuable synthetic tool for the synthesis of highly functionalized organic molecules and various drug like compounds.[1-3] The MCRs protocol has significant advantages upon the conventional two-component reactions in several aspects including lower costs, shorter reaction times, high degree of atom economy, and environmental friendliness. These reactions play a fundamental role in combinatorial chemistry and drug discovery research as they can provide an easy access to large information of organic compounds with diverse substitution patterns.[4-6] Consequently, the introduction of new MCRs and improving the already known MCRs are of special interest.

In recent years, the syntheses of 4H-chromene and 3,4-dihydropyrano[c]chromene derivatives have attracted great interest due to their biological and pharmacological activities[7,8] such as anti-coagulant, anticancer, spasmolytic, anti-anaphylactic, etc.[9] Furthermore, these compounds can be used as pigments,[10] photoactive materials[11] and utilized as potential biodegradable agrochemicals.[12] This moiety also occurs in different natural products.[13] A literature search revealed that different reagents have been used for preparation of 4H-chromene or 3,4-dihydropyrano[c]chromene derivatives involving piperidine,[14] Rare earth perfluorooctanoate ($Re(PFO)_3$),[15] $KF–Al_2O_3$,[16] Tetrabutylammonium bromide (TBAB),[17] (S)-proline,[18] $(NH_4)_2HPO_4$,[19] tetrabutylammonium fluoride (TBAF),[20] MgO,[21] 1,1,3,3-*N,N,N',N'*-Tetramethylguanidinium trifluoroacetate (TMGT)[22] and 1,8-Diazabicyclo[5.4.0]undec-7-ene (DBU).[23] Although several methods have been reported for the synthesis of 4*H*-chromene and 3,4-dihydropyrano[c] chromene derivatives, there is still a demand for preparation of these valuable compounds using a cheap and readily available catalyst.

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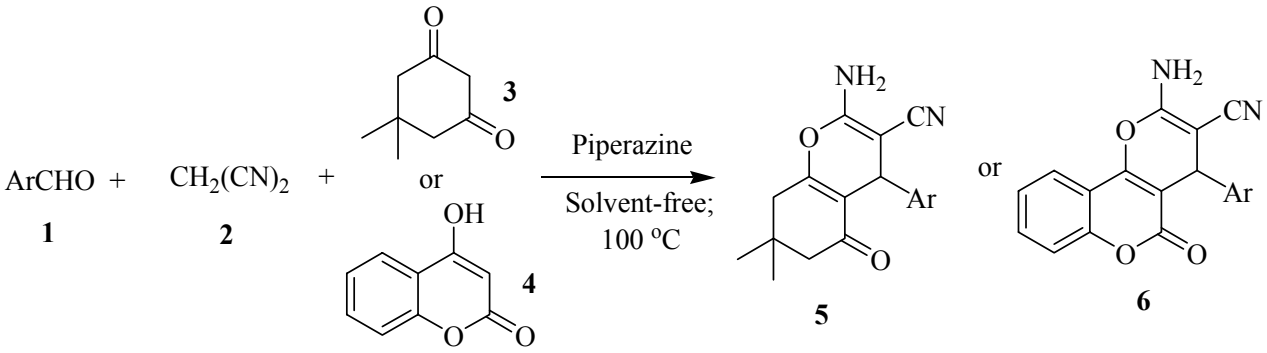
RESULTS AND DISCUSSION

As a part of our continuous interest directed towards the development of practical safe and environmentally friendly procedures for some important transformations,[24-29] we wish to report a new and efficient method for the synthesis of 4H-chromene and 3,4-dihydropyrano[c]chromene derivatives in the presence of piperazine as an odourless and easy to work catalyst under solvent-free conditions (Scheme 1).

Preliminary study, the condensation reaction of benzaldehyde with malononitrile and dimedone in the presence of piperazine was explored in order to search for the optimal conditions, such as the amount of catalysts and reaction conditions (Table 1). As shown in Table 1, the best conditions for this reaction were solvent-free, 10 mol% of piperazine and solvent-free at 100 ºC, since the reaction could be carried out in high yield (Table 1, entry 7).



Scheme 1 – The synthetic pathway for preparation of 4*H*-chromene and 3,4-dihydropyrano[c]chromenes.

Table 1

Optimization of reaction conditions

Entry	Conditions	Piperazine (mol%)	Time (min)	Yield/ %[a]
1	C_2H_5OH/R.T.	10	60	<20
2	C_2H_5OH/Reflux	10	60	83
3	CH_3CN/ Reflux	10	60	70
4	Toluene/ Reflux	10	60	55
5	THF/ Reflux	10	60	68
6	Solvent-free/70 ºC	10	10	67
7	Solvent-free/100 ºC	10	6	94
8	Solvent-free/100 ºC	20	6	94

[a] Isolated yields.

To explore the generality of the reaction, we extended our study using piperazine as catalyst under solvent-free conditions with different aromatic aldehydes containing both electron withdrawing and donating substitutes to prepare a series of 2-amino-7,7-dimethyl-5-oxo-4-(aryl)-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile derivatives (Table 2). The results clearly indicate that reactions can tolerate a wide range of differently substituted aldehydes. Reaction of aromatic aldehydes and malononitrile, with 4-hydroxycoumarin instead of dimedone also underwent successful condensation under similar conditions, to afford a series of 2-amino-4-(aryl)-5-oxo-4,5-dihydropyrano[3,2-c]chromene-3-carbonitrile derivatives in good yields. To demonstrate the scope and limitation of the procedure, the reactions of *ortho*-substituted aromatic aldehydes including 2-methoxybenzaldehyde, 2-chlorobenzaldehyde and 2-chloro-6-fluorobenzaldehyde and heteroaromatic aldehydes such as thiophene-2-carbaldehyde were studied, and corresponding products were obtained in good to high yields. On the other hand, only traces of corresponding product were produced in the reaction with *α, β*-unsaturated aldehydes such as cinnamaldehyde and aliphatic aldehydes.

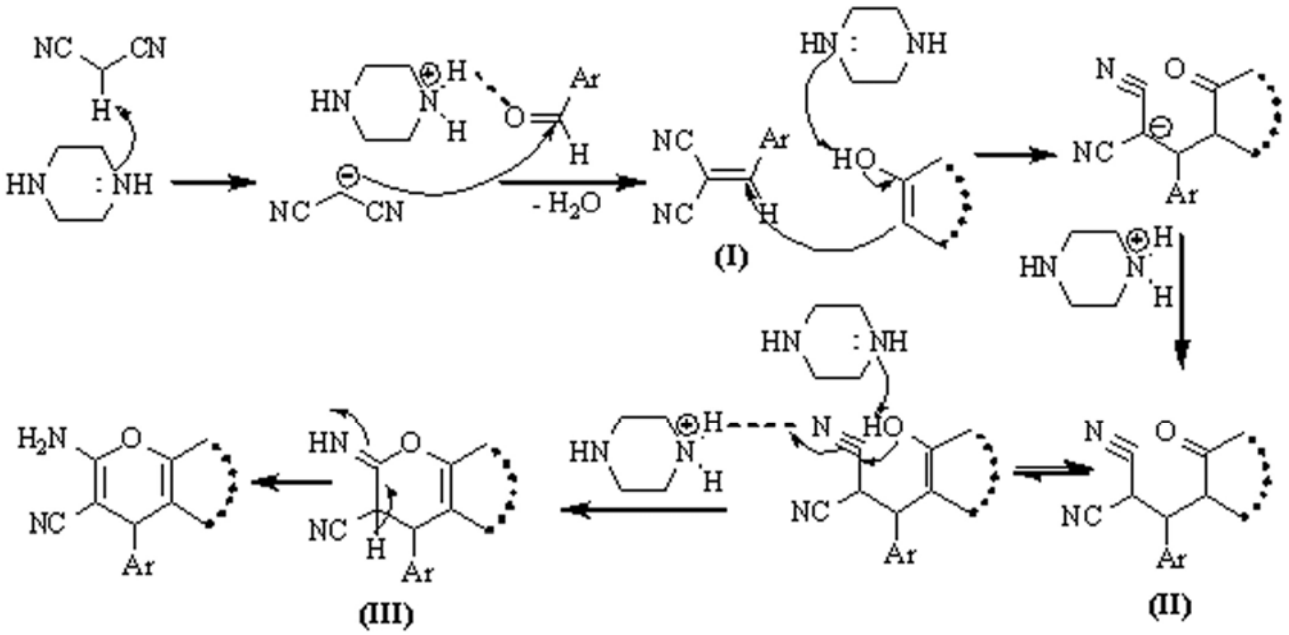
A plausible mechanism for the formation of chromene derivatives catalyzed by piperazine is shown in Scheme 2. The Knoevenagel reaction in the presence of piperazine occurs via an initial formation of α-cyanocinnamonitrile derivatives (I), after removal of one molecule of water from the condensation of aromatic aldehydes and malononitrile. Dimedone or 4-hydroxycoumarin is converted to the enol form after tautomerisation and attacks the α-cyanocinnamonitrile derivatives (I), as Michael acceptor, to give intermediate (II). The latter is then cyclized by nucleophilic attack of the OH group on the cyano (CN) moiety, giving intermediate (III), and is subsequently tautomerized to yield the chromene derivatives.

Table 2

Synthesis of 4*H*-chromene and 3,4-dihydropyrano[c]chromene derivatives

Entry	Ar (1)	Time (min)	Yield (%) [a]	TOF (min^{-1})[b]	Product	M.P (° C)	
						Found	Reported [lit.]
1	C_6H_5	7	92	1.31	**5a**	232-233	234-235 [20]
2	4-MeO C_6H_4	10	84	0.84	**5b**	203-204	201-202 [20]
3	2-Cl C_6H_4	12	81	0.67	**5c**	189-191	191-192 [20]
4	4-Cl C_6H_4	8	88	1.10	**5d**	217-218	215-216 [20]
5	4-Br C_6H_4	10	79	0.79	**5e**	202-204	201-203 [15]
6	3,4-$(MeO)_2$ C_6H_3	15	77	0.51	**5f**	190-192	-
7	2-Cl-6-F C_6H_3	10	75	0.75	**5g**	205-206	-
8	2-thiophene	5	85	1.70	**5h**	230-232	-
9	2-Me C_6H_4	10	86	0.86	**6a**	250-251	251-253 [23]
10	4-MeO C_6H_4	12	81	0.67	**6b**	238-240	240-242 [19]
11	4-Cl C_6H_4	8	82	1.02	**6c**	260-262	263-265 [19]
12	3,4-$(MeO)_2$ C_6H_3	15	76	0.51	**6d**	230-233	-
13	4-OH-3-MeO-5-NO_2 C_6H_2	10	68	0.68	**6e**	236-238	-
14	2-thiophene	7	79	1.13	**6f**	228-230	-

[a] Isolated yields.
[b]TOF = (mmol of product/mmol of catalyst)/time (min).



Scheme 2 – The possible mechanism for one-pot synthesis of of chromenes using piperazine as a catalyst.

EXPERIMENTAL

Aldehydes, malononitrile, dimedone, 4-hydroxycoumarin and other chemicals were purchased from Fluka and Merck companies. Products were characterized by IR, ^{1}H NMR and by comparison of their physical properties with those reported in the literature. IR spectra were recorded as KBr disc on a galaxy series FT-IR 5000 spectrometer. NMR spectra were recorded on a brucker spectrometer in DMSO-d_6 with TMS as an internal standard.

General procedure for preparation of 4*H*-chromene and 3,4-dihydropyrano[c] chromene derivatives

To a mixture of aldehyde (1 mmol), malononitrile (1.2 mmol) and dimedone/4-hydroxycoumarin (1 mmol), Piperazine (0.1 mmol) was added. The reaction mixture stirred magnetically at 100 ºC for appropriate time as shown in Table 2. The progress of the reaction was monitored by thin layer chromatography. After completion of the reaction, the reaction mixture was cooled to room temperature, washed with water, and the residue was recrystallized from ethanol.

Compound 5a: ^{1}H NMR (300 MHz, DMSO-d_6): δ= 7.28 (t, *J*=7.3 Hz, 2H), 7.21-7.12 (m, 3H), 6.98 (s, 2H), 4.17 (s, 1H), 2.51 (s, 2H), 2.25 (d, *J*=16.1 Hz, 1H), 2.10 (d, *J*=16.1 Hz, 1H), 1.04 (s, 3H), 0.96 (s, 3H).

Compound 5b: ^{1}H NMR (300 MHz, DMSO-d_6): δ= 7.05 (d, *J*=8.1 Hz, 2H), 6.94 (s, 2H), 6.84 (d, *J*=8.1 Hz, 2H), 4.12 (s, 1H), 3.71 (s, 1H), 2.50 (s, 2H), 2.24 (d, *J*=16.1 Hz, 1H), 2.09 (d, *J*=16.1 Hz, 1H), 1.03 (s, 3H), 0.94 (s, 3H).

Compound 5c: ^{1}H NMR (300 MHz, DMSO-d_6): δ= 7.22-7.14 (m, 2H), 7.10 (t, *J*=8.8 Hz, 2H), 7.02 (s, 1H), 4.21 (s, 1H), 2.50 (s, 2H), 2.24 (d, *J*=16.1 Hz, 1H), 2.10 (d, *J*=16.1 Hz, 1H), 1.02 (s, 3H), 0.94 (s, 3H).

Compound 5d: ^{1}H NMR (300 MHz, DMSO-d_6): δ= 7.34 (d, *J*=8.4 Hz, 2H), 7.17 (d, *J*=8.4 Hz, 2H), 7.04 (s, 2H), 4.19 (s, 1H), 2.50 (s, 2H), 2.24 (d, *J*=16.1 Hz, 1H), 2.10 (d, *J*=16.1 Hz, 1H), 1.03 (s, 3H), 0.94 (s, 3H).

Compound 5e: 1H NMR (300 Hz, DMSO-d_6): δ= 1.03 (s, 3H), 1.12 (s, 3H), 2.15 (d, *J* = 16.4 Hz, 2H), 2.29 (d, *J* = 16.4 Hz, 2H), 2.45 (s, 2H), 4.35 (s, 1H), 4.64 (s, 2H), 7.13 (d, *J* = 8.4Hz, 2H), 7.44 (d, J = 8.4 Hz, 2H).

Compound 5f: IR (KBr): ν_{max} = 3393, 3327, 3217, 2959, 2197, 1680, 1660, 1605, 1512, 1417, 1368, 1250, 1140, 1026, 858, 752 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6): δ= 6.93 (s, 2H), 6.86 (d, *J* = 8.1 Hz,1), 6.69-6.63 (m, 2H), 4.1 (s, 1H), 3.70 (s, 6H), 2.49 (s, 2H), 2.25 and 2.10 (AB system, *J* = 17.2 Hz, 2H), 1.04 (s, 3H), 0.97 (s, 3H); ^{13}C NMR(75 MHz, DMSO-d_6): δ= 196.1, 162.8, 158.9, 149.0, 148.0, 137.8, 120.3, 119.6, 113.3, 112.3, 111.5, 59.1, 56.0, 55.9, 50.5, 35.5, 32.2, 29.0, 27.1. Anal Calcd for $C_{20}H_{22}N_2O_4$: C, 67.78; H, 6.26; N, 7.90%. Found: C, 67.59; H, 6.21; N, 7.80%.

Compound 5g: IR (KBr): ν_{max} = 3412, 3333, 3213, 2964, 2199, 1698, 1662, 1601, 1452, 1370, 1215, 1144, 1040, 899, 781 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6): δ= 7.27 (d, *J* = 7.1 Hz, 2H), 7.17-7.09 (m, 3H), 4.89 (s, 1H), 2.55 and 2.35 (AB system, *J* = 17.8 Hz, 2H), 2.27 and 2.05 (AB system, *J* = 16.1 Hz, 2H), 1.06 (s, 3H), 0.99 (s, 3H); ^{13}C NMR(75 MHz, DMSO-d_6): δ= 196.1, 164.0, 159.8, 134.1, 129.6, 129.5, 126.3, 126.1, 119.7, 115.4, 110.7, 56.5, 50.3, 32.1, 29.5, 29.1, 26.8. Anal Calcd for $C_{18}H_{16}ClFN_2O_2$: C, 62.34; H, 4.65; N, 8.08%. Found: C, 62.25; H, 4.71; N, 8.02%.

Compound 5h: IR (KBr): ν_{max} = 3383, 3325, 3207, 2964, 2199, 1701, 1678, 1602, 1375, 1215, 1035, 700 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6): δ= 7.31 (d, *J* = 4.9 Hz, 1H), 7.10 (s, 2H), 6.92-6.86 (m, 2H), 4.53 (s, 1H), 2.54 and 2.43 (AB system, *J* = 17.6 Hz, 2H), 2.30 and 2.15 (AB system, *J* = 16.1 Hz, 2H), 1.04 (s, 3H), 0.98 (s, 3H); ^{13}C NMR(75 MHz, DMSO-d_6): δ= 196.0, 163.0, 159.4, 149.7, 127.3, 124.8, 124.5, 120, 113.4, 58.6, 50.5, 40.1, 32.2, 30.9, 29.1, 27.0. Anal Calcd for $C_{16}H_{16}N_2O_2S$: C, 63.98; H, 5.37; N, 9.33; S, 10.67%. Found: C, 63.88; H, 5.40; N, 10.70; S, 10.65%.

Compound 6a: ^{1}H NMR (300 MHz, DMSO-d_6): δ= 7.89 (d, *J*=7.8 Hz, 1H), 7.66–7.73 (m, 1H, Ar), 7.46 (t, *J*=7.6 Hz, 1H), 7.41 (d, *J*=8.3 Hz, 1H), 7.34 (s, 2H), 6.90–7.20 (m, 4H), 4.73 (s, 1H), 2.48 (s, 3H).

Compound 6b: ^{1}H NMR (300 MHz, DMSO-d_6): δ= 7.89 (d, *J* = 7.7 Hz, 1H), 7.70 (t, *J* = 7.7 Hz, 1H), 7.49 (t, *J* = 7.8 Hz, 1H), 7.45 (d, *J* = 8.3 Hz, 1H), 7.37 (br s, 2H), 7.18 (d, *J* = 8.1 Hz, 2H), 6.87 (d, *J* = 8.1 Hz, 2H), 4.40 (s, 1H), 3.72 (s, 3H),

Compound 6c: ^{1}H NMR (300 MHz, DMSO-d_6): δ= 7.92 (d, *J* = 7.8 Hz, 1H), 7.71 (t, J = 7.8 Hz, 1H), 7.49 (t, *J* = 7.6 Hz, 1H), 7.44 (d, *J* = 8.2 Hz, 1H), 7.38 (m, 2H), 7.36 (br s, 2H), 7.31 (d, *J* = 8.2 Hz, 2H), 4.50 (s, 1H),

Compound 6d: IR (KBr): ν_{max} = 3406, 3325, 3080, 2937, 2197, 1711, 1672, 1608, 1516, 1419, 1379, 1255, 1141, 1049, 954, 760 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6): δ= 7.89 (d, *J* = 7.7 Hz, 1H), 7.70 (t, *J* = 7.7 Hz, 1H), 7.50-7.42 (m, 2H), 7.34 (s, 2H), 6.70-6.85 (m, 2H), 6.75 (d, *J* = 7.8 Hz, 1H), 4.41 (s, 1H), 3.72 (s, 3H), 3.68 (s, 3H); ^{13}C NMR(75 MHz, DMSO-d_6): δ= 160.0, 158.4, 153.6, 152.6, 149.0, 148.4, 136.3, 133.3, 125.0, 122.9, 120.1, 119.8, 117.0, 113.5, 112.4, 112.1, 104.6, 58.7, 56.0, 55.9, 37.0 . Anal Calcd for $C_{21}H_{16}N_2O_5$: C, 67.02; H, 4.28; N, 7.46%. Found: C, 66.91; H, 4.22; N, 7.51%.

Compound 6e: IR (KBr): ν_{max} = 3400, 3292, 2982, 2218, 1716, 1674, 1607, 1516, 1408, 1373, 1315, 1265, 1159, 1060, 943, 761 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6): δ= 10.50 (br, 1H), 7.87 (d, *J* = 7.9 Hz, 1H), 7.66 (t, *J* = 7.9 Hz, 1H), 7.59 (s, 2H), 7.49-7.39 (m, 2H), 7.33 (s, 1H), 7.23 (s, 1H), 4.53 (s, 1H), 3.86 (s, 3H); ^{13}C NMR(75 MHz, DMSO-d_6): δ= 160.1, 158.5, 154.1, 152.6, 149.8, 142.2, 137.3, 134.5, 133.3, 125.0, 123.0, 119.6, 116.8, 116.4, 115.1, 113.4, 103.4, 57.7, 56.6, 36.9. Anal Calcd for $C_{20}H_{13}N_3O_7$: C, 58.97; H, 3.22; N, 10.32%. Found: C, 58.87; H, 3.27; N, 10.24%.

Compound 6f: IR (KBr): ν_{max} = 3368, 3281, 3173, 2201, 1711, 1670, 1607, 1383, 1172, 1055, 955, 760, 706 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6): δ= 7.86 (d, *J* = 7.5 Hz, 1H), 7.67 (t, *J* = 7.3 Hz, 1H), 7.51 (s, 2H), 7.46-7.37 (m, 4H), 7.02-6.95 (m, 2H), 4.81 (s, 1H); ^{13}C NMR(75 MHz, DMSO-d_6): δ= 159.9, 158.9, 153.4, 152.5, 147.9, 133.6, 127.6, 125.7, 125.6, 125.2, 122.9, 119.6, 117.0, 113.2, 104.5, 58.2, 32.3. Anal Calcd for $C_{17}H_{10}N_2O_3S$: C, 63.34; H, 3.13; N, 8.69; S, 9.95%. Found: C, 63.25; H, 3.17; N, 8.61; S, 10.03%.

CONCLUSIONS

In conclusion we have developed a facile protocol for the synthesis of 4*H*-chromene and 3,4-dihydropyrano[c] chromene derivatives from condensation reaction of aromatic aldehydes, malononitrile and dimedone or 4-hydroxycoumarin in the presence of piperazine as a catalyst under solvent-free conditions. Some advantages of this solvent-free protocol include a simple reaction set-up, good to high products yields, short reaction times, and elimination of toxic solvents.

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