ONE POT BF_3.MeCN CATALYZED SOLVENT FREE SYNTHESIS OF 3,4-DIHYDROPYRIMIDINE-2-ONE ANALOGUES

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Keywords: BF_3.ACN, solvent-free, 3,4-dihydropyrimidine-2-one.

One-pot solvent free three components coupling of aryl aldehydes, β-dicarbonyl compounds, urea or thiourea was performed to afford corresponding 3,4-dihydropyrimidine-2-ones and their sulfur analogs 3,4-dihydro-pyrimidine-2-thiones. It is the first report of BF_3.ACN catalyzed the solvent-free synthesis of pyrimidone analogs.

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INTRODUCTION

The multicomponent reactions (MCRs) are established as a simple, convenient method in synthetic chemistry.[1-3] Furthermore, MCRs are extremely economical, high yielding, less time consuming and with less side reactions.[4-5] Therefore, the design of new MCRs with the green procedure has engaged huge attention, especially in the areas of drug discovery, organic synthesis and material science.

Pyrimidines have extremely biological importance,[6-11] they and their analogs are considered as important bioactive heterocycles -+exhibiting interesting biological activities like antiviral,[12] antiprotozoan,[13] anti-proliferative,[14] cytotoxic activity[15] and anti-inflammatory .[16]

As a part of our ongoing efforts to develop new routes for the synthesis of heterocyclic compounds,[17] herein, we like to report a solvent-free single step multicomponent synthesis of 3,4-dihydropyrimidine-2-one and 3,4-dihydropyrimidine-2-thione derivatives.

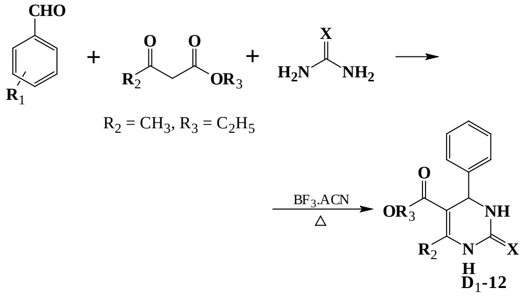

Figure 1. BF_3.ACN catalyzed solvent-free synthesis 3,4-dihydropyrimidine-2-one and 3,4-dihydropyrimidine-2-thione derivatives.

It is the first report of solvent-free condensation of β-keto esters, aryl aldehydes and urea or thiourea in the presence of BF_3.MeCN (BF3*ACN)) as an effective catalyst (**Figure 1**).

RESULTS AND DISCUSSION

Initially, a mixture of benzaldehyde, ethyl acetoacetate and urea was refluxed in ethanol in the presence of BF_3.ACN (Table 1) to obtain the corresponding 3,4-dihydropyrimidine-2-one derivative. The product was obtained in good yield (90 %). Solvent optimization studies of the above reaction were carried out and are summarized in Table 1. The reaction proceeded very well in solvent-free condition (Table 1, 97%).

Table 1. Solvent optimization for one-pot synthesis 3,4-dihydropyrimidine-2-one in the presence of 10 mol % BF3.MeCN catalyst[a]

Solvent	**Condition**	**Time, min**	**Yield, %[b]**
Ethanol	Reflux	60	90
Water	Reflux	130	85
Water : Ethanol (1:1)	Reflux	120	88
Methanol	Reflux	90	88
Acetonitrile	Reflux	35	92
Solvent Free	90 °C	20	97

a) Experimental conditions: benzaldehyde (2 mmol), urea (3 mmol), ethyl acetoacetate (2 mmol); b) Isolated yield.

Similarly, catalyst optimization studies of the above reaction were also carried out in solvent-free conditions and are summarized in Table 2. When catalyst was used from 5 mol%, 10 mol%, 15 mol% both yield and rate of the reaction was increased. However, the further increment of catalyst amount did not appreciably affect the yield and rate of the reaction. Finally, among all the experimental variations, the 10 mol% BF_3.ACN solvent-free condition at 90 ° C temperature gave the best results with 97% yield (Table 2).

To check the generality and scope of the optimized reaction, different aromatic aldehydes, β-ketoesters, urea and thiourea were used. The resultant 3,4-dihydropyrimidine-2-one (**D_{1-9}**) and 3,4-dihydropyrimidine-2-thione derivatives (**D_{10-12}**) were obtained in good to excellent yields as mentioned in Table 3.

Table 2. Catalyst optimization for one-pot synthesis 3,4-dihydropyrimidine-2-one [a]

Sr. No.	Catalyst, mol %	Time, min	Yield, %[b]
1	5%	35	85
2	10%	20	97
3	15%	15	95
4	20%	15	95
5	25%	15	95

a)Experimental conditions: benzaldehyde (2 mmol), urea (3 mmol), ethyl acetoacetate (2 mmol) at 90 °C; b)Isolated yield.

Table 3. Synthesis of 3,4-dihydropyrimidine-2-ones and 3,4-dihydropyrimidine-2-thiones from aryl aldehydes, β-ketoesters and urea/thiourea[a]

Aldehyde	X	β–keto-ester[c]	Yield[b]	Melting point, °C	
				Measured	Reported
C_6H_5	O	EAA	97	203-204	206[18]
m-$NO_2C_6H_4$	O	EAA	94	226-227	227-228[20]
p-HOC_6H_4	O	EAA	99	223-226	227-228[20]
p-ClC_6H_4	O	EAA	95	208-210	209-212[18]
m-ClC_6H_4	O	EAA	98	194-196	193-194[20]
m-HOC_6H_4	O	EAA	97	166-169	167-170[18]
C_6H_5	O	MAA	92	211-213	212-213[18]
p-$MeOC_6H_4$	O	EAA	91	198-199	199-201[19]
p-FC_6H_4	O	EAA	94	175-176	176-178[21]
C_6H_5	S	EAA	99	206-208	207-208[19]
m-$NO_2C_6H_4$	S	MAA	98	273-274	273-275[18]
p-HOC_6H_4	S	EAA	97	201-203	202-203[21]

a) Reaction conditions: Aromatic aldehyde (2 mmol), Urea/Thiourea (3 mmol), MAA or EAA (2 mmol) and catalyst (10 mol%) solvent free at 90^{0}C; b)Isolated yield, c) MAA-methyl acetoacetate, EAA-ethyl acetoacetate

EXPERIMENTS

All the chemicals were purchased from Sigma Aldrich and used as received without further purification. All compounds were matched with and confirmed by literature data for Melting point, IR, ^{1}H NMR, ^{13}C NMR and mass spectrometry. The melting points were determined on Labstar melting point apparatus and are uncorrected. The IR spectra were taken on a Perkin-Elmer FTIR-1600 spectrophotometer and the data expressed in cm (KBr). ^{1}H and ^{13}C NMR spectra were recorded on Bruker Avance (300 MHz) spectrometer in $CDCl_3$ using TMS as the internal standard. Mass spectra were recorded on an Agilent spectrometer.

General procedure for the preparation of 3,4-dihydropyrimidine-2-one and 3,4-dihydropyrimidine-2-thione derivatives (D_{1-12})

A mixture of β-ketoester (2 mmol), urea/thiourea (3 mmol), aryl aldehyde (2 mmol) and BF_3.ACN (10 mol%) was heated at 90°C till the completion of the reaction, monitored by TLC in Dichloromethane : Methanol (9:1) as a mobile phase. The reaction mixture was cooled and poured in 10 mL ice-water and precipitated solid was filtered out to give the desired crude product. The crude product was recrystallized with ethanol to get pure 3,4-dihydropyrimidine-2-one and 3,4-dihydropyrimidine-2-thione product as shown in (Table 3). The products were analyzed by IR, ^{1}H and ^{13}C NMR.

Ethyl 1,2,3,4-tetrahydro-6-methyl-2-oxo-4-phenylpyrimidine-5-carboxylate

White solid, mp. 203–204 °C; IR (KBr) ν: 3228, 3106, 2936, 1721, 1695, 1604, 1221 cm^{-1}; ^{1}H NMR (300 MHz, $CDCl_3$) δ: 1.23 (t, 3H), 2.35 (s, 3H), 4.10 (m, 2H), 5.25 (s, 1H), 5.98 (s, 1H), 7.88–7.13 (m, 5H), 8.25 (s, 1H); ^{13}C NMR (75 MHz, $CDCl_3$) δ: 14.1, 18.3, 54.4, 61.4, 102.3, 126.2, 127.2, 128.7, 143.5, 146.1, 163.6 ppm.

Ethyl 1,2,3,4-tetrahydro-6-methyl-4-(3-nitrophenyl)-2-oxopyrimidine-5-carboxylate (D_2)

Off-white solid, mp. 226-227°C; IR (KBr) ν: 3408, 3106, 2954, 1670, 1605, 1590, 1524, 1348, 1215 cm^{-1}; ^{1}H NMR (300 MHz, $CDCl_3$) δ: 1.21 (t, 3H), 2.54 (s, 3H), 4.37 (q, 2H), 5.21 (s, 1H), 7.18-7.25 (m, 2H), 7.88 (d, 2H, *3J* = 8.7 Hz), 8.17 (s, 1H), 8.81 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 165.7, 158.5, 148.7, 148.4, 131.8, 130.6, 129.5, 125.7, 121.8, 118.8, 61.2, 53.4, 25.4, 17.3 ppm.

Ethyl 1,2,3,4-tetrahydro-4-(4-hydroxyphenyl)-6-methyl-2-oxopyrimidine-5-carboxylate (D_3)

White solid, mp. 223-226°C; IR (KBr) ν: 3510, 3285, 3115, 2968, 1658, 1523, 1466, 1218 cm^{-1}; ^{1}H NMR (300 MHz, $CDCl_3$) δ: 1.14(t, 3H), 2.24 (s, 3H), 3.96 (m, 2H), 5.06 (s, 1H), 6.75 (d, 2H), 7.05 (d, 2H), 9.15 (s, 1H), 9.36 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 166.5, 159.1, 152.8, 147.9, 136.8, 126.3, 124.8, 115.8, 62.8, 49.3, 24.4, 19.4 ppm.

Ethyl 4-(4-chlorophenyl)-1,2,3,4-tetrahydro-6-methyl-2-oxopyrimidine-5-carboxylate (D4)

White solid, mp 208-210°C; IR (KBr) ν: 3239, 3117, 2969, 1715, 1646, 1458, 1225, 1093 cm^{-1}; ^{1}H NMR (300 MHz, $CDCl_3$) δ: 1.21 (t, 3H), 2.38 (s, 3H), 4.11 (m, 2H), 5.85 (s, 1H), 7.31 (d, 2H), 7.30 (d, 2H), 8.06 (s, 1H); ^{13}C NMR (100MHz, $CDCl_3$) δ: 168.2, 158.6, 146.8, 143.3, 145.5, 132.1, 129.2, 117.1, 61.4, 51.2, 22.4, 18.3 ppm.

Ethyl 4-(3-chlorophenyl)-1,2,3,4-tetrahydro-6-methyl-2-oxopyrimidine-5-carboxylate (D_5)

White solid, mp. 194-196°C; IR (KBr) ν:3245, 3110, 2975, 1705, 1655 cm^{-1}; ^{1}H NMR ($CDCl_3$ 300 MHz) δ: 1.21 (t, 3H), 2.43 (s,3H), 4.21 (m, 2H), 5.42 (s, 1H), 7.22 (d, 2H), 7.33 (d, 2H), 7.61 (brs, 1H), 8.12 (brs, 1H). ^{13}C NMR ($CDCl_3$ 100 MHz) δ: 168.2, 158.4, 146.5, 143.2, 145.3, 131.6, 129.2, 117.1, 61.4, 51.4, 22.3, 18.5 ppm.

Ethyl 1,2,3,4-tetrahydro-4-(3-hydroxyphenyl)-6-methyl-2-oxopyrimidine-5-carboxylate (D_6)

White solid, mp. 166-169°C; IR (KBr) ν: 3515, 3310, 3106, 2958, 1724, 1645, 1612, 1466, 1223 cm^{-1}; ^{1}H NMR (300 MHz, $CDCl_3$) δ: 1.14 (t, 3H), 2.25 (s, 3H), 4.06 (m, 2H), 5.06 (s, 1H), 6.62 (d, 1H), 6.68 (d, 2H), 7.10 (t, 2H), 9.11 (s, 1H), 9.31 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 163.7, 157.8, 150.2, 146.4, 133.9, 131.7, 130.2, 124.7, 121.3, 115.8, 60.9, 54.7, 26.2, 18.1 ppm.

Methyl 1,2,3,4-tetrahydro-6-methyl-2-oxo-4-phenylpyrimidine-5-carboxylate (D_7)

White solid, mp. 211-213°C, IR (KBr) ν: 3415, 3320, 3106, 2950, 1728, 1660, 1632, 1475, 1234 cm^{-1}; ^{1}H NMR ($CDCl_3$ 300 MHz) δ: 9.23 (s, 1H), 7.74 (s, 1H), 7.45-7.35 (m, 2H), 7.28-7.26 (m, 3H), 5.18 (d, 1H), 3.55 (s, 3H), 2.26 (s, 3H); ^{13}C NMR ($CDCl_3$ 100 MHz) δ: 166.3, 152.8, 150.1, 145.3, 129.4, 128.4, 127.5, 99.8, 54.6, 51.8, 18.8 ppm.

Ethyl 1,2,3,4-tetrahydro-4-(4-methoxyphenyl)-6-methyl-2-oxopyrimidine-5-carboxylate (D_8)

White solid, mp. 198-199°C; IR (KBr) ν: 3254, 3105, 2955, 1710, 1645, 1515, 1464, 1225 m^{-1}; ^{1}H NMR (300 MHz, $CDCl_3$) δ: 1.13 (t, 3H), 2.24 (s, 3H), 3.38 (s, 3H), 4.1 (m, 2H), 5.11 (s, 1H), 6.90 (d, 2H), 7.16 (d, 2H), 7.71 (s, 1H), 9.14 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 168.0, 158.2, 152.4, 149.5, 136.7, 130.2, 123.3, 118.8, 62.4, 61.8, 49.7, 25.7, 19.7 ppm.

Ethyl 4-(4-fluorophenyl)-1,2,3,4-tetrahydro-6-methyl-2-oxo-pyrimidine-5-carboxylate (D_9)

White solid, mp. 175-176°C; IR (KBr) ν: 3243, 1698, 1638 cm^{-1}; ^{1}H NMR (300 MHz, $CDCl_3$) δ: 9.24 (s, 1H), 7.81 (s, 1H), 7.23 (m, 4H), 5.14 (s, 1H), 4.12 (m, 2H), 2.23 (s, 3H), 1.11 (t, 3H); ^{13}C NMR (100 MHz $CDCl_3$) δ: 165.8, 160.1, 152.2, 148.5, 141.4, 128.3, 115.3, 99.4, 59.4, 53.8, 17.6, 14.8 ppm.

Ethyl 1,2,3,4-tetrahydro-6-methyl-4-phenyl-2-thioxopyrimidine-5-carboxylate (D_{10})

White solid, mp. 206-208°C, IR (KBr) ν: 3236, 3126, 2946, 1728, 1698, 1226 cm^{-1}; ^{1}H NMR (300 MHz, $CDCl_3$) δ: 9.63 (1H, s), 8.94 (1H, s), 6.67-6.54 (m, 5H), 4.53 (d, 1H), 3.37 (m, 2H), 1.61 (3H, s), 0.44 (t, 3H); ^{13}C NMR ($CDCl_3$ 100 MHz) δ: 175.3, 166.3, 145.9, 144.4, 129.3, 128.4, 127.1, 101.7, 60.5, 54.8, 18.1,14.8 ppm.

Ethyl 1,2,3,4-tetrahydro-6-methyl-4-(3-nitrophenyl)-2-thioxopyrimidine-5-carboxylate(D_{11})

White solid, mp. 273-274°C, IR (KBr) ν: 3325, 3215, 3105, 2963, 1715, 1634, 1520 cm^{-1}; ^{1}H NMR (300 MHz,$CDCl_3$) δ: 9.38 (s, 1H), 8.17- 7.69 (m, 4H), 3.88 (m, 2H), 2.24 (s, 3H), 1.13 (t, 3H); ^{13}C NMR ($CDCl_3$ 100 MHz) δ: 165.3, 151.9, 149.6, 147.9, 147.2, 133.2, 130.2, 122.3, 121.4, 98.4, 59.3, 53.7, 17.8, 14.2 ppm.

Ethyl 1,2,3,4-tetrahydro-4-(4-hydroxyphenyl)-6-methyl-2-thioxopyrimidine-5-carboxylate (D_{12})

White solid, mp 201-203 °C; IR (KBr) ν: 3223, 3098, 2980, 1742, 1655, 1459, 1251 cm^{-1}; ^{1}H NMR (300 MHz, $CDCl_3$) δ: 1.24 (t, 3H), 1.89 (s, 3H), 4.16 (m, 2H), 5.87 (s, 1H), 7.32(d, 2H), 7.33 (d, 2H), 8.07 (s, 1H); ^{13}C NMR (100MHz, $CDCl_3$) δ: 168.2, 158.6, 146.8, 143.3, 145.5, 132.1, 129.2, 117.1, 61.4, 51.2, 22.4, 18.3 ppm.

CONCLUSION

In summary, it is the first report of cost-effective, solvent-free mild protocol for the synthesis of 3,4-dihydropyrimidine-2-one and 3,4-dihydropyrimidine-2-thione derivatives using BF_3.ACN as a catalyst. This MCRs protocol offers several significant advantages like operational simplicity, superior atom-economy, shorter reaction time with good to excellent yields.

ACKNOWLEDGMENT

The authors are thankful to the Head, Department of Chemistry, Maulana Azad College, Dr. Babasaheb Ambedkar Marathwada University, Aurangabad-431 004, India, for allowing the work.

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This paper was presented at the "*International Symposium on Exploring New Horizons in Chemical Sciences*," January 10–12, **2019**, Aurangabad, India (ENHCS–2019).

Received: 11.03.2019
Accepted: 22.06.2019