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Microwave assisted synthesis of 6-methyl-1,2,3,4-tetrahydro-*N*-aryl-2-oxo-4-arylpyrimidine-5-carboxamide and 3, 4-dihydropyrimidin-2(*1H*)-ones under solvent free conditions

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ABSTRACT

6-Methyl-1,2,3,4-tetrahydro-N-aryl-2-oxo-4-arylpyrimidine-5-carboxamide and 5-unsubstituted 3, 4-dihydropyrimidin-2(1H)-ones have been synthesised from three component cyclocondensation of N-aryl-3-oxobutanamide/acetophenone, aldehyde and urea /thiourea in presence of PTSA under solvent free microwave irradiation.

Key words: Pyrimidine-5-carboxamide, dihydropyrimidin-2(*1H*)-ones, *N*-phenyl-3-oxobutanamide, solvent free , PTSA.

INTRODUCTION

The exploitation of simple molecules with different functionalities is a worthwhile contribution in the chemistry of heterocycles. Dihydropyrimidin-2(*1H*)-ones (DHPMs) are well known to possess varied pharmacological and biological activities [1] and hence their synthesis has always been of attraction to organic chemists. Of the different pyrimidine derivatives, dihydropyrimidinones are the most important in the synthesis of different drugs such as anticancer [2], antihypertensive [3] and antiviral [4]. As a consequence, many approaches have been developed to synthesize dihydropyrimidinones. Several methods using promoters such as

AcOH [5a], $InCl_3$ [5b], lanthanide triflate [5c], $BF_3.OEt_2$ [5d], PPE [5e], KSF clay [5f], $LaCl_3$ [5g], H_2SO_4 [5h], Ceric ammonium nitrate (CAN) [5i], $Mn(OAc)_3$ [5j], ion-exchange resin [5k], $InBr_3$[5l], $FeCl_3$[5m], $CdCl_2$[5n], 1-n-butyl-3-methyl imidazolium tetrafluoroborate[5o], ytterbium triflates [5p], SiO2/NaHSO4[5q], BiCl3[5r], LiClO4[5s], $ZrCl_4$[5t], $Cu(OTf)_2$[5u], $Bi(OTf)_3$[5v], $AlCl_3.H_2O$[5w], etc. have been found to be effective. However, some of these methods require the use of toxic reagents [in combination with bronsted acids, such as hydrochloric acid[6] and acetic acid [7] as additives, expensive reagents [8], stoichiometric amounts of catalysts, strongly acidic conditions [5h], long reaction times [9], and unsatisfactory yields [10]. Therefore, the development of a mild and efficient alternative procedure would extend the scope of pyrimidinone chemistry.

EXPERIMENTAL SECTION

N-aryl-3-oxobutanamide was prepared according to the literature procedure [16]. Microwave reactions were carried out in a CEM Discover Benchmate microwave digester. Melting points are uncorrected. Infrared spectra were recorded on a BOMEM DA-8 FTIR instrument and the frequencies are expressed in cm^{-1}. ^1H and ^{13}C NMR spectra were recorded on a Bruker Avance II-400 spectrometer using $CDCl_3$ as the solvent. Chemical shifts are reported in ppm downfield from internal tetramethylsilane and are given on the δ scale. Mass spectral data were obtained with a JEOL D-300 (EI) mass spectrometer. Elemental analyses were carried out on a Heraeus CHN-O-Rapid analyzer. All compounds give satisfactory elemental analyses within ± 0.4% of the theoretical values. All reactions were monitored by TLC using precoated aluminum sheets (silica gel 60 F_{254} 0.2 mm thickness) and developed in an iodine chamber or under UVGL-15 mineral light 254 lamp. Column chromatographic separations were carried out using ACME silica gel (60–120 mesh).

General Procedure for the Synthesis of Title compound

To a mixture of *N*-aryl-3-oxobutanamide/acetophenone(1 mmol), aldehyde(1 mmol) and urea /thiourea(1.5 mmol), a catalytic amount of p-toluenesulfonic acid was added and grounded thoroughly. The mixture was then subjected to Microwave Irradiation in Microwave digester at 100^{o}C and 200 watts for the specified time (**Table 1).** After completion of the reaction, (monitored by TLC), the reaction mixture was treated with water to remove the catalyst and the unreacted urea/thiourea and was extracted with ethylacetate (3 x 5 mL). The combined extract was dried (anhydrous Na_2SO_4) and the solvent was removed on rotavapor to give the crude product which was purified by recrystallisation from dicloromethane : hexane or by column chromatography using dichloromethane: methanol as an eluent.

Spectroscopic and Analytical Data:

1,2,3,4-Tetrahydro-6-methyl-*N*,4-diphenyl-2-thioxopyrimidine-5-carboxamide 4a.

mp 213-215°C (lit.[13] mp 216-217°C); IR (KBr): 3205, 1670, 1605,1519 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.07 (s, 3H, CH_3), 5.15 (d, 1H), 7.29-8.24 (m, 10H, Ar-H), 9.19 (s, 1H, NH), 9.63 (s, 1H), 9.77 (s, 1H); ^{13}C NMR ($CDCl_3$): δ 14.81, 53.73, 106.51, 120.96, 125.41, 126.61,127.73, 128.55, 129.35, 136.31, 144.22, 158.71, 162.90, 179.33; Mass: m/z 323 [M^+]; Anal. Calcd for $C_{18}H_{17}N_3OS$: C, 66.85; H, 5.30; N, 12.99; Found: C 67.09; H, 5.25; N, 12.87%.

1,2,3,4-Tetrahydro-6-methyl-2-oxo-*N*,4-diphenylpyrimidine-5-carboxamide 4b
mp 229-231°C; IR (KBr): 3200, 1675, 1610 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.31 (s, 3H, CH_3), 5.65 (d, 1H), 7.08-8.95 (m, 10H, Ar-H), 9.02 (s, 1H, NH), 9.43 (s, 1H), 9.83 (s, 1H,); ^{13}C NMR ($CDCl_3$): δ 13.61, 48.00, 105.41, 120.46, 123.81, 125.00, 126.66, 127.13, 128.15, 133.41, 142.61, 145.86, 148.38, 161.25; Mass: m/z 307 [M^+]; Anal. Calcd for $C_{18}H_{17}N_3O_2$: C, 70.34; H, 5.58; N, 13.67; Found: C, 69.93; H, 5.25; N, 13.33%.

4-(4-Chlorophenyl)-1,2,3,4-tetrahydro-6-methyl-*N*-phenyl-2-thioxopyrimidine-5-carboxamide 4c. mp 217–218°C (lit.[13] mp 221-223°C); IR (KBr): 3222 , 1695 , 1605 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.23(s, 3H, CH_3), 5.54 (d, 1H), 7.37-8.67 (m, 9H, ArH), 9.37 (s, 1H, NH),9.61 (s, 1H, NH), 9.70 (s, 1H,); ^{13}C NMR ($CDCl_3$): δ 13.66, 53.43, 104.68, 120.96, 123.82, 127.32, 127.94, 128.69, 131.42, 134.20, 140.83, 158.00, 162.06, 178.44; Mass: m/z 357 [M^+]; Anal. Calcd for $C_{18}H_{16}ClN_3OS$: C, 60.41; H, 4.51; N, 11.74; Found: C, 60.12; H, 4.27; N, 12.07%.

1,2,3,4-Tetrahydro-*N*-(4-methoxyphenyl)-6-methyl-2-oxo-4-phenylpyrimidine-5-carboxamide 4d. mp 237–238°C (lit.[16] mp 240-242°C); IR (KBr): 3237, 3112, 2307, 1674, 1526 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.07 (s, 3H, CH_3), 3.52 (s, 3H, OCH_3), 5.56 (s, 1H, CH), 7.23–7.79 (m, 9H, ArH), 8.03 (s, 1H, NH), 8.87 (s,1H, NH), 9.41 (s, 1H, NH); ^{13}C NMR ($CDCl_3$): δ 13.91, 48.70, 53.27, 106.65, 113.28, 121.82, 125,12, 126.00, 127.12, 127.85, 143.27, 146.00, 157.41, 149.72, 161.61; Mass: m/z 337 [M^+]; Anal. Calcd for $C_{19}H_{19}N_3O_3$: C, 67.64; H, 5.68; N, 12.46; Found: C, 67.74; H, 5.67; N, 12.37%.

1,2,3,4-Tetrahydro-*N*-(4-methoxyphenyl)-6-methyl-4-phenyl-2-thioxopyrimidine-5-carboxamide 4e. mp 206-208°C (lit.[16] mp 209-212°C); IR (KBr): 3254, 3078, 1632 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.11 (s, 3H, CH_3), 3.89 (s, 3H, OCH_3), 5.66 (s, 1H, CH), 6.95-7.88 (m, 9H, ArH), 9.06 (s, 1H, NH), 9.59 (s, 1H, NH), 9.95 (s, 1H, NH); ^{13}C NMR ($CDCl_3$): δ 13.90, 54.82, 54.71, 105.22, 112.63, 120.82, 125.17, 126.00, 127.41, 127.86, 141.22, 152.13, 157.00, 161.27, 178.86; Mass: m/z 353[M^+]; Anal. Calcd for $C_{19}H_{19}N_3O_2S$: C, 64.57; H, 5.42; N, 11.89; Found: C, 64.78; H, 5.67; N, 11.67%.

1,2,3,4-Tetrahydro-*N*,4-bis(4-methoxyphenyl)-6-methyl-2-oxopyrimidine-5-carboxamide 4f. mp 237-239°C (lit.[16] mp 238-240°C); IR (KBr): 3311, 3032, 2987, 1667 , 1515 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.09 (s, 3H, CH_3), 3.87 (s, 3H, OCH_3), 5.21 (s, 1H, CH), 7.07-8.65 (m, 8H, ArH), 7.89 (s, 1H, NH), 8.78 (s, 1H, NH), 9.34 (s, 1H, NH); ^{13}C NMR ($CDCl_3$): δ 13.55, 53.01, 49.87, 107.63, 113.65, 115.01, 121.82, 127.59, 127.97, 134.80, 153.65, 156.41, 145.38, 148.05, 161.87, Mass: m/z 367[M^+]; Anal. Calcd for $C_{20}H_{21}N_3O_4$: C, 65.38; H, 5.76; N, 11.44: Found: C, 65.87; H, 5.23; N, 11.37%.

1,2,3,4-Tetrahydro-*N*,4-bis(4-methoxyphenyl)-6-methyl-2-thioxopyrimidine-5-carboxamide 4g. mp 216-218°C (lit.[16] mp 216-220°C)**;** IR (KBr): 3243, 3017, 2965, 1657 $cm^{-1;}$ ^1H NMR ($CDCl_3$): δ 2.12 (s,3H, CH_3), 3.73 (s, 3H, OCH_3), 5.40 (s, 1H, CH), 7.21-8.11 (m, 8H, ArH), 9.11 (s, 1H, NH), 9.54 (s, 1H, NH), 9.79 (s, 1H, NH);^{13}C NMR ($CDCl_3$): δ 13.95, 53.06, 54.41, 106.00, 113.65, 114.00, 121.36, 127.58, 127.99, 134.51, 155.00, 156.89, 158.16, 160.87, 178.47; Mass: m/z 383[M^+]; Anal. Calcd for $C_{20}H_{21}N_3O_3S$: C, 62.64; H, 5.52; N, 10.96; Found: C, 62.53; H, 5.67; N, 10.34%.

4-(3-Chlorophenyl)-1,2,3,4-tetrahydro-*N*-(4-methoxyphenyl)-6-methyl-2-oxopyrimidine-5-carboxamide 4h. mp 212-215°C(lit.[16] mp 212-214°C); IR (KBr): 3256, 3029, 1645 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.05 (s, 3H, CH_3), 3.74 (s, 3H, OCH_3), 5.45 (s, 1H, CH), 6.89- 7.56 (m, 8H, ArH), 8.74 (s, 1H, NH), 9.46 (s, 1H, NH), 9.69 (s, 1H, NH); ^{13}C NMR ($CDCl_3$): δ 14.13, 53.18, 56.82, 104.82, 112.51, 121.28, 125.26, 126.81, 128.31, 128.76, 129.83, 130.03, 131.47. 132.12, 146.81, 163.11, 178.12; Mass: m/z 371[M^+]; Anal. Calcd for $C_{19}H_{18}ClN_3O_2S$: C, 58.83; H, 4.60; N, 10.83; Found: C, 58.72; H, 4.64; N, 10.33%.

1,2,3,4-Tetrahydro-6-methyl-4-phenyl-2-thioxo-*N*-o-tolylpyrimidine-5-carboxamide 4i. mp 225-226°C(lit.[16] mp 229-231°C); IR (KBr): 3207, 1680, 1608 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.01 (s, 3H, p-CH_3), 2.33 (s, 3H,CH_3), 5.34 (d, 1H), 7.08-8.43 (m, 9H, ArH), 9.07 (s, 1H, NH), 9.35 (s, 1H, 3-H), 9.76 (br, 1H, NH); ^{13}C NMR ($CDCl_3$): δ 13.68, 54.72, 105.82, 120.61, 122.11, 123.26, 125.21, 126.86, 126.91, 127.60, 128.00, 133.52, 141.60, 158.00, 161.58, 178.32; Mass: m/z 337 [M^+]; Anal. Calculated for $C_{19}H_{19}N_3OS$: C, 67.63; H 5.68; N 12.45; Found: 67.54; H, 5.32; N, 12.34%.

3,4-Dihydro-4,6-diphenylpyrimidin-2(*1H*)-one 6a.
mp 239-241°C (lit.[14b] mp 245-246°C); IR (KBr): 3225, 2922, 1687, 1602, 1451 cm^{-1}; ^1H NMR ($CDCl_3$): δ 5.07 (d, 1H, CH), 5.45 (d, 1H, CH), 7.12–7.95 (m, 10H, ArH), 8.57 (s, 1H, NH), 9.04 (s, 1H, NH); ^{13}C NMR ($CDCl_3$): δ 52.91, 98.55, 125.68, 126.01, 126.98, 127.00, 127.71, 127.99, 132.31, 141.27, 135.61, 168.97; Mass: m/z 250 [M^+]; Anal. Calcd for $C_{16}H_{14}N_2O$: C, 76.78; H, 5.64; N, 11.19; Found: C, 76.42; H, 5.33; N, 11.27%.

3,4-Dihydro-4,6-diphenylpyrimidine-2(*1H*)-thione 6b.
mp 251-253°C (lit.[14a] mp 257-258°C); IR (KBr): 3307, 3075, 1673, 1602, 1554 cm^{-1}; ^1H NMR ($CDCl_3$): δ 5.48 (s, 1H), 5.89 (s, 1H), 7.00-7.87 (m, 10H ArH), 8.03 (s, 1H), 8.72 (s, 1H); ^{13}C NMR ($CDCl_3$): δ 54.71, 95.37, 125.61, 126.52, 127.30, 128.50, 129.31, 130.50, 135.31, 137.42, 150.23, 163.33; Mass: m/z 266 [M^+]; Anal. Calcd. For $C_{17}H_{16}N_2O$; C, 72.18; H, 5.26; N, 10.53; Found: C, 72.20; H, 5.37; N, 10.47%.

3,4-Dihydro-4-(4-methoxyphenyl)-6-phenylpyrimidin-2(*1H*)-one 6c.
mp 247-249°C (lit.[14a] mp 259-261°C); IR (KBr): 3348, 2932, 1654, 1535, 1421 cm^{-1}; ^1H NMR ($CDCl_3$): δ 3.69 (s, 3H, OCH_3), 5.27 (d, 1H, CH), 5.89 (d, 1H, CH), 7.17-7.85 (m, 9H, ArH), 8.07 (s,1H, NH), 9.25 (s, 1H, NH); ^{13}C NMR ($CDCl_3$): δ 52.47, 57.83, 115.23, 118.32, 126.43, 127.81, 129.38, 130.81, 131.21, 134.81, 145.92, 163.18; Mass: m/z 280 [M^+]; Anal. Calcd. For $C_{17}H_{16}N_2O$; C, 72.84; H, 5.75; N, 9.99; Found: C, 72.82; H, 5.78; N, 10.02%.

3,4-Dihydro-4-(4-methoxyphenyl)-6-phenylpyrimidine-2(*1H*)-thione 6d.
mp 227-229°C (lit.[15b] mp 227-231°C); IR (KBr): 3332, 2971, 1644, 1581 cm^{-1}; ^1H NMR ($CDCl_3$): δ 3.45(s, 3H, OCH_3), 5.17 (d, 1H, CH), 5.74 (d, 1H, CH), 7.21-7.96 (m, 9H, ArH), 8.21 (s, 1H, NH), 9.15 (s, 1H, NH); ^{13}C NMR ($CDCl_3$): δ 54.46, 60.82, 117.13, 120.62, 125.76, 129.74, 130.68, 131.41, 132.22, 134.31, 145.42, 165.71. Mass: m/z 296 [M^+]; Anal. Calcd. For $C_{17}H_{16}N_2OS$: C, 68.89; H, 5.44; N, 9.45; Found:C, 68.65; H, 5.54; N, 9.37%.

3,4-Dihydro-6-(4-methoxyphenyl)-4-phenylpyrimidin-2(*1H*)-one 6e.
mp 209-211°C (lit.[15b] mp 210-213°C); IR (KBr): 3353, 2953, 1637, 1581 cm^{-1}; ^1H NMR ($CDCl_3$): δ 3.53 (s, 3H, OCH_3), 5.51 (d, 1H, CH), 5.83 (d, 1H, CH), 7.10-8.12 (m, 9H, ArH), 8.32 (s, 1H, NH), 9.21 (s, 1H, NH); Mass: m/z: 280 [M^+]; ^{13}C NMR ($CDCl_3$): δ 50.87, 56.73, 98.12, 115.83, 124.82, 126.52, 127.41, 128.00, 128.61, 130.89, 138.90, 145.80, 157.92; Anal. Calcd. For $C_{17}H_{16}N_2O$; C, 72.84; H, 5.75; N, 9.99; Found: C, 72.68; H, 5.68; N, 10.04%.

3,4-Dihydro-6-(4-methoxyphenyl)-4-phenylpyrimidin-2(*1H*)-one 6f.
mp 203-204°C (lit.[15b] mp 203-204°C); IR (KBr): 3307, 3007, 1626, 1574 cm^{-1}; ^1H NMR ($CDCl_3$): δ 3.45 (s, 3H, OCH_3), 5.23 (d, 1H, CH), 5.78 (d, 1H, CH), 7.34-7.98 (m, 9H, ArH), 8.16 (s, 1H, NH), 9.22 (s, 1H, NH); ^{13}C NMR ($CDCl_3$): δ 54.87, 57.81, 96.31, 117.21, 123.82, 126.84, 127.18, 128.63, 129.66, 130.67, 137.82, 150.58, 177.38; Mass: m/z 296 [M^+]; Anal. Calcd. For $C_{17}H_{16}N_2OS$: C, 68.89; H, 5.44; N, 9.45; Found: C, 68.86; H, 5.39; N, 9.31%.

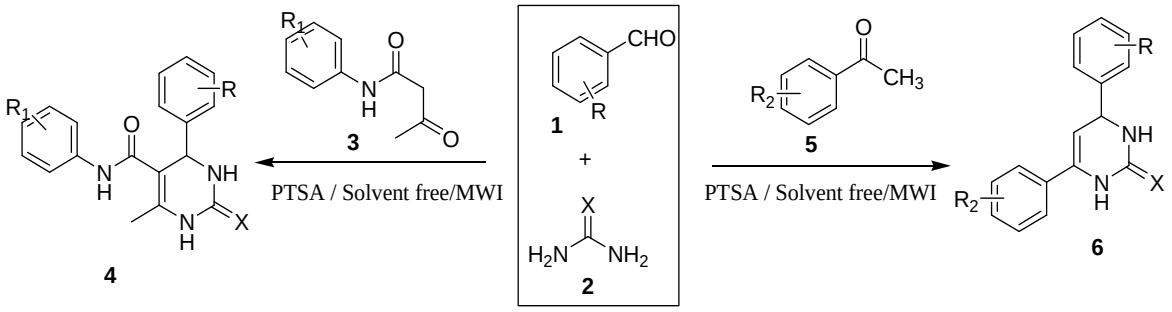
3,4-Dihydro-6-phenyl-4-p-tolylpyrimidin-2(*1H*)-one 6g.
mp 247-249°C (lit.[14a] mp 248-250°C); IR (KBr): 3232, 2915, 1637, 1589, 1416 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.54 (s, 3H, CH_3), 5.45 (d, 1H, CH), 5.63 (d, 1H, CH), 7.08–7.88 (m, 9H, ArH), 8.56 (s, 1H, NH), 9.21 (s, 1H, NH); ^{13}C NMR ($CDCl_3$): δ 23.87, 58.11, 98.23, 120.81, 124.91, 126.81, 127.00, 127.86, 128.23, 129.17, 133.20, 139.80, 159.76; Mass: m/z 264 [M^+]; Anal. Calcd for $C_{17}H_{16}N_2O$: C, 77.25; H, 6.10; N, 10.60; Found: C, 77.33; H, 5.89; N, 10.58%.

4-(4-Chlorophenyl)-3,4-dihydro-6-phenylpyrimidin-2(*1H*)-one 6h.
mp 259-261°C(lit.[14a] mp 267-269°C); IR (KBr): 3221, 2930, 1673, 1565, 1465 cm^{-1}; ^1H NMR ($CDCl_3$): δ 5.12 (d, 1H, CH); 5.34 (d, 1H, CH), 7.09–7.78 (m, 9H, Ar–H), 8.23 (s, 1H, NH), 8.96 (s, 1H, NH); ^{13}C NMR ($CDCl_3$): δ 51.87, 96.82, 124.82, 127.33, 128.00, 128.77, 129.37, 129.89, 130.72, 123.88, 144.86, 155.58; Mass: m/z [M^+]; Anal. Calcd for $C_{16}H_{13}ClN_2O$: C, 67.49; H, 4.60; N, 9.84; Found: C, 66.99; H, 4.62; N, 10.02%.

RESULTS AND DISSCUSSION

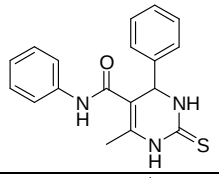
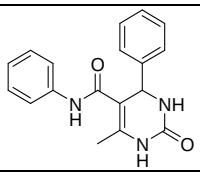
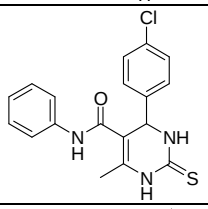
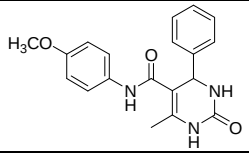
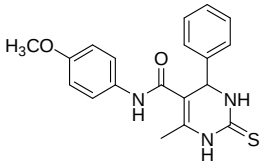
Solvent free reactions [11] in fine chemical synthesis are of increasing interest to synthetic organic chemists and industrialists. The development of solvent-free synthesis using microwave irradiation (MWI), technique has contributed significantly to the eco-friendly synthesis due to the increasing environmental consciousness worldwide. Our literature survey revealed that in almost all the reported methods, DHPMs have been synthesized starting from diketones or ketoesters [12], while the syntheses from *N*-aryl-3-oxobutanamide [13] or acetophenone [14] are not well documented. Therefore, in continuation with our investigation on the methodology of green synthesis [15], we report herein a three-component cyclocondensation of *N*-aryl-3-oxobutanamide/acetophenone, aldehyde and urea /thiourea to synthesise pyrimidinone derivatives using inexpensive p-toluenesulfonic acid as catalyst under solvent-free condition and microwave irradiation, which is an efficient and environmentally friendly method (**Scheme 1**). p-toluenesulfonic acid (*p*-TsOH) is water soluble which can be easily removed from the crude product by washing with water.

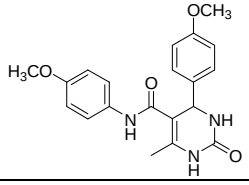
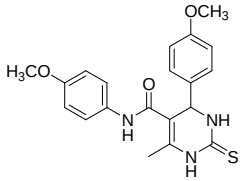
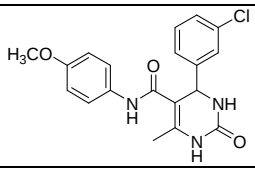
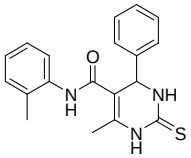
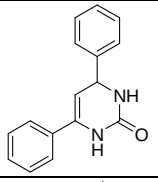
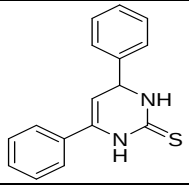
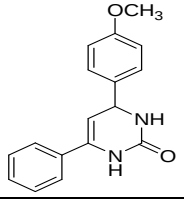
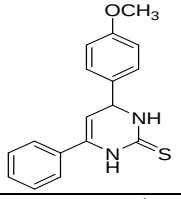
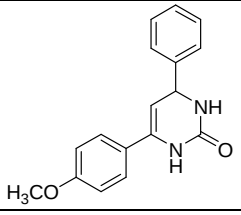
In summary, this paper discloses a rapid and simple protocol of the pyrimidinone synthesis through the use of readily available p-TsOH as reaction mediator. The excellent yields with short reaction time, makes this method better than many existing ones.

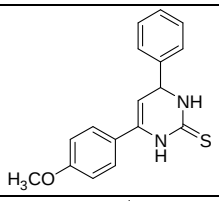
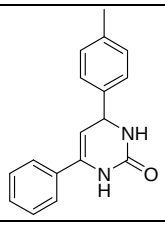
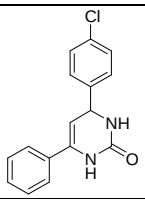


Scheme 1. General synthetic route of the title compounds

Table I. List of the title compound with reaction time and yields

Entry	R	R_1/R_2	X	Product	Reaction Time.(Min)	Yield(%)[a]
4a	H	H	S		5	75
4b	H	H	O		5	73
4c	4-Cl	H	S		5	60
4d	H	4-OCH_3	O		6	66
4e	H	4-OCH_3	S		4.5	58

4f	4-OCH_3	4-OCH_3	O		4.5	70
4g	4-OCH_3	4-OCH_3	S		4	62
4h	3-Cl	4-OCH_3	O		5	74
4i	H	2-CH_3	S		5	65
6a	H	H	O		7	76
6b	H	H	S		7	69
6c	4-OCH_3	H	O		5	67
6d	4-OCH_3	H	S		5	65
6e	H	4-OCH_3	O		5	72

6f	H	4-OCH_3	S		5	65
6g	4-CH_3	H	O		6.5	67
6h	4-Cl	H	O		6	63

[a] *refer to isolated yields*

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