



Three Component Condensation: Synthesis and Characterization of Bioactive 4-Substituted Aryl Pyrimidinones and Pyrimidinethiones

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(Received: May 01, 2011; Accepted: June 10, 2011)

ABSTRACT

A series of some novel 4-aryl substituted pyrimidine(1H)-2-one and its thio analogue pyrimidine(1H)-2-thione have been synthesized in appreciable yield by three component condensation of urea/thiourea, ethylacetoacetate and appropriate aromatic aldehydes in alcoholic medium in presence of catalytic amount of acid. The structure of newly synthesized compounds have been confirmed by physicochemical as well as spectral i.e. IR and NMR (¹H & ¹³C) studies.

Key words: Bioactive 4-substituted Aryl pyrimidinones, Pyrimidinethiones.

INTRODUCTION

Cyclic compounds bearing hetero atoms are well known compounds which exhibit a wide range of bio activities and have great application in the field of medicine as well in synthetic organic chemistry^{1,2}. Pyrimidines, their derivatives and their thio analogues have received significant attention owing to their diverse range of therapeutic and pharmacological properties such as analgesic, antitumor, anti-inflammatory, antihypertensive, antiviral, antibacterial, fungicidal, as calcium channel blockers and as potent inhibitor of cancer cell proliferation³⁻¹².

The biological activity of some recently isolated alkaloids has also been attributed due to

the presence of dihydropyrimidinone moiety in the molecule^{13,14}. These compounds were found to be potent human immunodeficiency virus inhibitors and considered as a source for the development of new anticancer drugs^{11,15-17}.

Therefore, the synthesis of dihydropyrimidinones has gained much acceptance and popularity among organic chemists and numerous synthetic methods for preparing these compounds have been reported by using Lewis as well as protonic acid promoters and anionic liquids as catalyst¹⁸⁻³².

Keeping in view of the importance of these, it was planned to synthesize a series of pyrimidin(1H)-2-one and (their thioanalogues) pyrimidine(1H)-2-

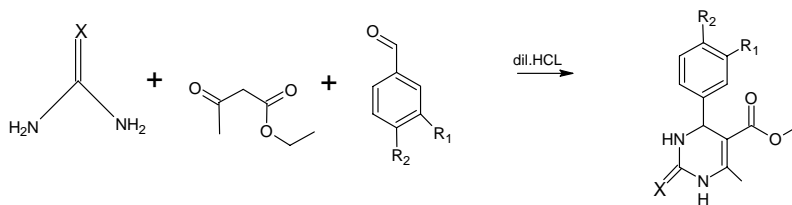
thiones by three component condensation of urea/thiourea, substituted aldehydes and α - ketoester.

EXPERIMENTAL

The chemicals used for the synthesis were of analytical grade (BDH, E.Merck and Spectrochem.) and were used as received. The solvents were used after double distillation. All the analyses were carried out at RSIC, CDRI, Lucknow.

These compounds were prepared by

refluxing a mixture of urea/thiourea and substituted aldehyde in equimolar ratio with slightly higher ratio of acetoacetic ester and catalytic amount of acid was refluxed for required period of time. After the completion of reaction the reaction mixture was kept in refrigerator overnight. The solid separated out was filtered, washed with suitable solvent and crystallized by appropriate solvent or solvent system. The filtrate was again refluxed for few hours to check further precipitation. These compounds were characterized by MPs, elemental analysis and spectral studies (IR, ^1H & ^{13}C NMR).



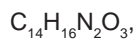
Comp. No.	X	R1	R2
1, 8	O, S	H	H
2, 9	O, S	H	Me
3, 10	O, S	H	OMe
4, 11	O, S	NO ₂	H
5, 12	O, S	H	Cl
6, 13	O, S	H	N (Me) ₂
7, 14	O, S	OMe	OMe

5-carboethoxy-6-methyl-4-phenyl (1H) pyrimidin-2-one

Reactants

Urea (0.1 mole, 6.0 gm), Ethyl acetoacetate (0.15 mole, 19 ml), Benzaldehyde (0.1 mole, 10.1ml), Yield: - 55.6 gm; M.P. - 201-209

Molecular formula



Elemental analysis

Calcd. (Found) C, 64.61(64.49), H, 6.15(6.04), N, 10.76 (10.56)

IR (v)

3300, 3030, 2960, 2870, 1740, 1690, 1580, 1460

$^1\text{HNMR}$

δ 7.09-7.18 (5H, m, Ar-H), 5.46 (1H, s, CH), 6.19 (1H, br, NH), 8.18 (1H, s, NH), 1.71 (3H, s, CH₃), 4.19 (2H, m, CH₂CH₃), 1.18 (3H, t, CH₂CH₃)

$^{13}\text{CNMR}$

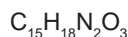
143.2, 128.6, 127.1, 126.6 (Benzene ring resonances) 167.2, 150.3, 137.3, 106.4, 61.7, 49.5, 17.2, 14.2 (Pyrimidine ring resonances)

Mass

m/z 260 [M]⁺, 232 [M-CO]⁺, 218 [M-NCO]⁺, 187 [M-COOC₂H₅]⁺, 162 [M-C₃H₄NO]⁺, 159[187-CO]⁺, 145 [187-NCO]⁺, 117 [187-C₃H₄NO]⁺

5-carboethoxy-6-methyl-4-(4-methyl phenyl) (1H) pyrimidin-2-one**Reactants**

Urea (0.1 mole, 6.0 gm), Ethylacetoacetate (0.15 mole, 19 ml), 4-methyl benzaldehyde (0.1mole, 11.8ml), Yield: - 60.1gm; M.P. - 170-172

Molecular formula**Elemental analysis**

Calcd. (Found), C, 65.69(65.76), H, 6.56(6.72), N, 10.21(10.35)

IR (ν)

3050, 3010, 2980, 2870, 1745, 1690, 1580, 1460

¹HNMR

δ 6.84-7.02 (4H,m,Ar-H), , 5.39 (1H,s,Py ring) 7.99(1H,br,NH) 8.45(1H,s,NH), 2.29(3H,s,CH₃), 1.88 (3H,s,CH₃), 4.11 (2H,m,CH₂CH₃), 1.08(3H,t,CH₂CH₃)

¹³CNMR

135.7, 130.4, 129, 127 (Benzene ring resonances) 157, 139.3, 106.7, 59.8, 48.8, 20.9, 17.2, 13.7 (Pyrimidine ring resonances)

5-carboethoxy-4-(4-methoxy phenyl)- 6-methyl (1H) pyrimidin -2-one**Reactants**

Urea (0.1 mole, 6.0 gm), Ethylacetoacetate (0.15 mole, 19 ml), 4-methoxy benzaldehyde (0.1mole, 12.1ml), Yield: - 64.7gm; M.P. - 202-205

Molecular formula**Elemental analysis**

Calcd. (Found), C, 62.06 (61.05), H, 6.20 (6.09), N, 9.65 (9.49)

IR (ν)

3040, 3015, 2990, 2880, 1740, 1685, 1570, 1460

¹HNMR

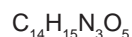
δ 6.90-6.65 (4H,m,Ar-H), 3.72 (3H,S,OCH3), 5.41 (1H,s,Py ring), 6.21 (1H,br,NH), 8.25 (1H,s,NH), 1.71 (3H,s,CH₃), 4.10 (2H,m,CH₂CH₃), 1.20(3H,t,CH₂CH₃)

¹³CNMR

165.2, 135.5, 128, 114.1 (Benzene ring resonances) 168.1, 150.3, 145.2, 108.2, 60, 48.8, 15, 14.7 (Pyrimidine ring resonances)

5-carboethoxy-6-methyl-4-(3-nitro phenyl) (1H) pyrimidin -2-one**Reactants**

Urea (0.1 mole, 6.0 gm), Ethylacetoacetate (0.15 mole, 19 ml), 3-nitro benzaldehyde (0.1mole, 15.1 gm), Yield: - 57.8 gm; M.P.-226-229

Molecular formula**Elemental analysis**

Calcd. (Found), C, 55.08 (54.09), H, 4.91 (4.62), N, 13.77 (13.47)

IR (ν)

3310, 3246, 3108, 2950, 2870, 1725, 1700, 1480

¹HNMR

δ 7.45-8.10 (4H,m,Ar-H), 5.40 (1H,s,CH Py ring), 8.45-6.20(1H,s,NH), 8.24 (1H, br,NH), 1.83 (3H,S,CH3), 4.16 (2H,m,CH₂CH₃), 1.24 (3H,t,CH₂CH₃)

¹³CNMR

148.8, 141.4, 133.1, 129.5, 122.2 (Benzene ring resonances) 166.6, 151.2, 146.3, 108.1, 60.5, 47.8, 14.5, 14.0 (Pyrimidine ring resonances)

5-carboethoxy-4-(4-chloro phenyl)-6-methyl (1H) pyrimidin-2-one**Reactants**

Urea (0.1 mole, 6.0 gm), Ethylacetoacetate (0.15 mole, 19 ml), 4-chlorobenzaldehyde (0.1mole, 14 gm), Yield: - 54.2 gm; M.P. - 208-214

Molecular formula**Elemental analysis**

Calcd. (Found), C, 57.04 (56.92), H, 5.09 (5.05), N, 9.50 (9.41), Cl, 12.05 (12.01)

IR (ν)

3410, 3110, 2980, 2860, 1710, 1690, 1510

¹HNMR

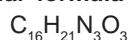
δ 7.01-7.29, (4H,m,Ar-H), 5.16(1H,s,CH Py ring),,7.05(1H, br,NH),8.78, (1H,s,NH), 1.91(3H,S,CH3),4.15 (2H,m,CH₂CH₃),1.19 (3H,t,CH₂CH₃)

¹³CNMR

141.3, 133.2, 129.1, 128.4 (Benzene ring resonances) 166.6, 149.8, 146.6, 105.8, 60.2, 47.8, 14.5, 14.1 (Pyrimidine ring resonances)

5- Carboethoxy-4-(4-N,N dimethyl phenyl)-6-methyl (1H) pyrimidin-2-one**Reactants**

Urea (0.1 mole, 6.0 gm), Ethyl acetoacetate (0.15 mole, 19 ml), 4-Dimethylaminobenzaldehyde (0.1mole, 14.9gm) Yield: - 62.7gm; M.P. 257-258

Molecular formula**Elemental analysis**

Calcd. (Found), C, 63.36 (63.23), H, 6.93 (6.88), N, 13.86 (13.79)

IR (ν)

3350, 3240, 2990, 1730, 1690, 1570, 1460

¹HNMR

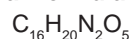
δ 6.88-6.47, 2.64 (6H, s, N(CH₃)₂), 5.12 (1H,s,Py ring), 7.60 (1H,br,NH), 8.82(1H,s,NH), 2.22 (3H,s,CH₃), 4.05 (2H,m,CH₂CH₃), 1.20 (3H,t,CH₂CH₃)

¹³CNMR

142.5, 132.7, 127.9, 112.4 (Benzene ring resonances) 167.2, 157.3, 139.3, 106.4, 61.7, 59.5, 43.6, 17.2, 14.7 (Pyrimidine ring resonances)

5-Carboethoxy-4-(3,4-dimethoxy phenyl)-6-methyl (1H) pyrimidin-2-one**Reactants**

Urea (0.1 mole, 6.0 gm), Ethyl acetoacetate (0.15 mole, 19 ml), And 3, 4-Dimethoxybenzaldehyde (0.1mole, 16.6gm) Yield: 58.7gm; M.P.; 176-177

Molecular formula**Elemental analysis**

Calcd. (Found), C, 60.0 (59.97), H, 6.25 (6.20), N, 8.75 (8.67)

IR (ν)

3040, 3015, 2990, 2880, 1740, 1685, 1590, 1460

¹HNMR

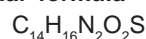
δ 6.41-6.65 (3H, m, Ar-H), 3.73 (6H, 2xOCH₃), 5.56(1H, s, Py ring), 6.39 (1H,br,NH), 8.33 (1H,s,NH), 1.79(3H,s,CH₃), 4.12 (2H,m,CH₂CH₃), 1.23(3H,t,CH₂CH₃)

¹³CNMR

116.1-148.2, 56.8(2xOCH₃), 149.8(CO), 144.1(C-CH₃), 14.2(C-CH₃), 107.9(C-CO), 167.8(CO), 59.9(CO-CH₂), 14.7(CO-CH₂CH₃)

5-carboethoxy-6-methyl-4-phenyl (1H) pyrimidine -2-thione**Reactants**

Thiourea (0.1mole, 7.6gm), Ethylacetoacetate (0.15 mole, 19 ml), Benzaldehyde (0.1 mole, 10.1ml) Yield: - 55.7 gm; M.P. – 202-209

Molecular formula**Elemental analysis**

Calcd. (Found), C, 60.86 (60.80), H, 5.79 (5.74), N, 10.14(10.09), S, 11.59 (11.51)

IR (ν)

3280, 3120, 2970, 2850, 1760, 1690, 1580

¹HNMR

δ 7.06-7.14 (5H, m, Ar-H), 4.16 (1H, s, CH

py ring), 9.66(1H, br, NH), 10.32(1H, s, NH), 1.81(3H, s, CH₃), 4.19(2H, m, CH₂CH₃), 1.11(3H, t, CH₂CH₃)

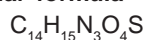
¹³CNMR

143.2, 128.6, 127, 126.6 (Benzene ring resonances) 54.6, 184.1, 152.3, 104.2, 165, 59.9, 17.9, 13.7 (Pyrimidine ring resonances)

5-carboethoxy-6-methyl-4-(4-methyl phenyl) (1H) pyrimidine-2-thione

Reactants

Thiourea (0.1mole, 7.6gm), Ethylacetoacetate (0.15 mole, 19 ml), 4-Methylbenzaldehyde (0.1mole, 11.8ml) Yield: - 66.4 gm; M.P. – 204-208

Molecular formula**Elemental analysis**

Calcd. (Found), C, 52.33(52.38), H, 4.67 (4.79), N, 13.08 (13.14), S, 9.96 (9.99)

IR (v)

3050, 3010, 2980, 2870, 1749, 1690, 1580, 1460

¹HNMR

δ 7.40-7.99 (4H, m, Ar-H), 4.65(1H, s, CH py ring), 9.71(1H, br, NH), 10.39(1H, s, NH), 1.81(3H, s, CH₃), 4.12(2H, m, CH₂CH₃), 1.16 (3H, t, CH₂CH₃)

¹³CNMR

135.7, 130.4, 129, 127 (Benzene ring resonances) 53.6, 183.9, 151.8, 103.7, 164.9, 60.1, 17.8, 13.8 (Pyrimidine ring resonances)

5-carboethoxy-4-(4-methoxy phenyl) 6-methyl (1H) pyrimidine -2-thione

Reactants

Thiourea (0.1mole, 7.6gm), Ethylacetoacetate (0.15 mole, 19 ml), 4-Methoxybenzaldehyde (0.1mole, 12.1ml), Yield: - 59.5gm; M.P. – 150-152

Molecular formula**Elemental analysis**

Calcd. (Found), C, 58.82(58.76), H, 5.88 (5.64), N, 9.15(9.06), S, 10.45 (10.22)

IR (v)

3040, 3015, 2990, 2880, 1750, 1685, 1570, 1460

¹HNMR

6.65-6.95(4H, m, Ar-H), 3.73(3H, s, OCH₃), 5.45(1H, s, CH pyring), 6.29(1H, br, NH), 8.30(1H, s, NH), 1.72(3H, s, CH₃), 4.12(2H, m, CH₂CH₃), 1.22(3H, t, CH₂CH₃)

¹³CNMR

165.2, 135.5, 128, 114.1 (Benzene ring resonances) 55.8 (OCH₃), 48.5, 149.3, 145.1, 14.4, 108.2, 168.0, 59.8, 15.0 (Pyrimidine ring resonances)

5-carboethoxy-6-methyl-4-(3-nitro phenyl) (1H) pyrimidine -2-thione

Reactants

Thiourea (0.1mole, 7.6gm), Ethylacetoacetate (0.15 mole, 19 ml), 3-Nitrobenzaldehyde (0.1mole, 15.1 gm), Yield: - 63.6gm; M.P. – 204-208

Molecular formula**Elemental analysis**

Calcd. (Found), C, 52.33 (52.45), H, 4.67(4.79), N, 13.08(13.28), S, 9.96(10.1)

IR (v)

3310, 3246, 3108, 2950, 2870, 1730, 1700, 1480

¹HNMR

δ 7.06-7.14(5H, m, Ar-H), 4.16(1H, s, CH py ring), 9.66(1H, br, NH), 10.32(1H, s, NH), 1.81(3H, s, CH₃), 4.19(2H, m, CH₂CH₃), 1.11(3H, t, CH₂CH₃)

¹³CNMR

148.8, 141.4, 133.1, 129.5, 122.2 (Benzene ring resonances) 54.6, 184.1, 152.3, 104.2, 165, 59.9, 17.9, 13.7(Pyrimidine ring resonances)

5-carboethoxy-4-(4-chloro phenyl)-6-methyl(1H) pyrimidin-2-thione**Reactants**

Thiourea (0.1mole, 7.6gm), Ethylacetoacetate (0.15 mole, 19 ml), and 4-Chlorobenzaldehyde (0.1mole, 14 gm) Yield: -68.1 gm; M.P. – 192-210

Molecular formula**Elemental analysis**

Calcd. (Found), C, 54.10 (55.08), H, 4.83 (4.92), N, 9.01 (9.19), Cl, 10.30 (10.41)

IR (v)

3410, 3110, 2980, 2860, 1710, 1690, 1510

¹HNMR

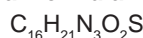
δ 7.0-7.18 (4H, m, Ar-H), 5.19(1H, s, CH py ring), 7.11(1H, br, NH), 8.84(1H, s, NH), 1.97(3H, s, CH₃), 4.13(2H, m, CH₂CH₃), 1.16(3H, t, CH₂CH₃)

¹³CNMR

141.3, 133.2, 129.1, 128.4 (Benzene ring resonances) 47.5, 149.7, 146.6, 14.2, 105.9, 166.7, 60.2, 14.1 (Pyrimidine ring resonances)

5- Carboethoxy-4-(4- N,N-dimethyl phenyl)-6-methyl (1H) pyrimidine-2-thione**Reactants**

Thiourea (0.1mole, 7.6gm), Ethylacetoacetate (0.15 mole, 19 ml), 4-Dimethylaminobenzaldehyde (0.1mole, 14.9gm) Yield: 68.9gm; M.P. – 210-225

Molecular formula**Elemental analysis**

Calcd (Found), C, 60.18(59.98), H, 6.58(6.57), N, 13.16 (13.06), S, 10.03 (9.97)

IR (v)

3350, 3240, 2990, 1730, 1690, 1570, 1460

¹HNMR

δ 6.42-6.91 (4H, m, Ar-H), 2.79 (6H, s, CH₃), 5.14(1H, s, CH py ring), 7.54(1H, br, NH), 9.14(1H, s, NH), 2.22(3H, s, CH₃), 4.04(2H, m,

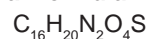
CH₂CH₃), 1.21(3H, t, CH₂CH₃)

¹³CNMR

142.5, 132.7, 127.9, 112.4 (Benzene ring resonances) 43.3, 150.3, 144.9, 14.6, 108.4, 167.9, 60.1, 14.9 (Pyrimidine ring resonances)

5- Carboethoxy-4-(3,4-dimethoxy phenyl)-6-methyl (1H) pyrimidin-2-thione**Reactants**

Thiourea (0.1mole, 7.6gm), Ethyl acetoacetate (0.15 mole, 19 ml), 3, 4-Dimethoxybenzaldehyde (0.1mole, 16.6gm) Yield: 60.6 gm; M.P.; 140-145

Molecular formula**Elemental analysis**

Calcd. (Found), C, 57.14 (57.22), H, 5.95 (6.05), N, 8.33 (8.67), S, 9.52 (9.66)

IR (v)

3040, 3015, 2990, 2880, 1740, 1685, 1590, 1460

¹HNMR

δ 6.41-6.65 (3H, m, Ar-H), 3.72 (6H, 2xOCH₃), 5.58 (1H, s, Py ring), 6.40 (1H,br,NH), 8.38 (1H,s,NH), 1.81 (3H,s,CH₃), 4.15 (2H,m,CH₂CH₃), 1.27 (3H,t,CH₂CH₃)

¹³CNMR

56.7(2xOCH₃), 149.6 (CO), 144.2 (C-CH₃), 14.3 (C-CH₃), 107.8 (C-CO), 167.9(CO), 60.0 (CO-CH₂), 14.9 (CO-CH₂CH₃)

RESULTS AND DISCUSSION

The 4-aryl substituted pyrimidine(1H)-2-one and pyrimidine(1H)-2-thiones were prepared by refluxing urea and/or thiourea, ethylacetoacetate and different aromatic aldehydes in alcoholic medium in presence of catalytic amount of acid for required period of time. The crude product so obtained was recrystallized from appropriate solvent/solvent system. These newly synthesized compounds were characterized by melting points and spectral studies like IR and NMR (¹H & ¹³C).

The synthesized compounds were obtained in appreciable yield and gave sharp melting points. The spectral studies showed well resolved signals for characteristic groups. The IR spectra of all the compounds exhibit the absorption bands for C=O (1690-1710 cm^{-1}) and ester groups (1720-1735 cm^{-1}). The absorption bands at 3335-3350 cm^{-1} were assigned for N-H frequency while those appearing at 1480-1540 cm^{-1} were due to C-N stretching vibrations. The sharp band appeared around 1650 cm^{-1} was clearly due to -CONH grouping. The ^1H NMR spectra of all the compounds showed sharp signals of aromatic protons in the range of δ 6.61-7.14. A singlet appeared at δ 5.40-5.50 was clearly assignable to methane proton of pyrimidine ring. Two downfield signals at δ 6.20 and δ 8.80 were due to NH protons. The methylene proton of ester

resonates at δ 4.16-4.20. The structure of the compounds was further supported by ^{13}C NMR spectra which in addition to signals of aromatic carbon atoms, also exhibit two downfield signal at δ 150 and 166 assignable to carbonyl and ester carbon atoms respectively. Another downfield signal appeared at 145 was due to pyrimidine ring carbon to which methyl group is attached and at 106 for pyrimidine carbon adjacent to ester carbon. The methylene carbon of ester group resonates around 60. The mass ion fraction of compound (1) with mass spectrum showed at m/z 260 corresponding to the molecular formula $\text{C}_{14}\text{H}_{16}\text{N}_2\text{O}_3$. Other prominent fraction appeared at m/z 187 reasonably due to the loss of ester group. The fractions found at 232, 218, 162, 159 and 145 were due to loss of CO and NCO groups.

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