Synthesis of Some Novel Fused Oxo- and Thiopyrimidines *via* an Intramolecular Friedel-Crafts Reaction

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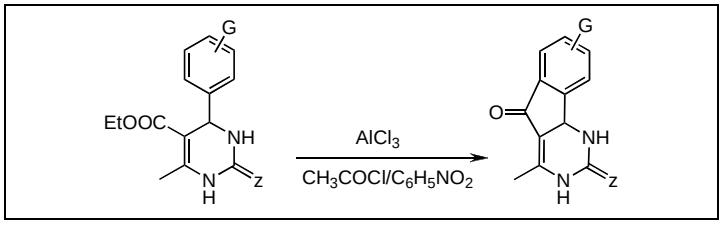

1*H*-Indeno[1,2-*d*]pyrimidine-2,5(3*H*,9b*H*)-dione derivatives **2(a-i)** and 2,3-dihydro-2-thioxo-1*H*-indeno[1,2-*d*]pyrimidine-5(9b*H*)-ones **2(j-q)** were synthesized *via* an intramolecular Friedel-Crafts reaction between the aryl and ester group of ethyl 6-methyl-4-aryl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylates **1a-i**, and their thioxo analogs using $AlCl_3$ and acetyl chloride in nitrobenzene. Yields of the products, after washing with THF, were of the order of 45-69%. IR and NMR spectroscopy together with elemental analysis were used for identification of these compounds.

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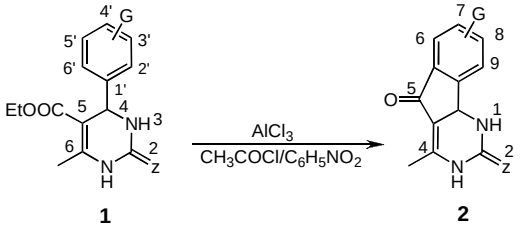
INTRODUCTION

Recently the Biginelli reaction has been extended for preparation of a large number of pyrimidines [1]. It is well documented that various compounds containing pyrimidine ring, are associated with diverse pharmacological activities such as antitumor [2], antiviral [3], anticancer [4], anti-inflammatory [4], antifolate [5], antimicrobial [6], antifungal [7] and antiproliferative [8] activities. Over the past decade many pyrimidines with appropriate functional groups have emerged as antihypertensive agents [9-12] and potent calcium channel blockers [13-14]. In addition, several marine alkaloids with interesting biological activities contain the dihydropyrimidine-5-carboxylate moiety [10]. The most convenient preparation method of simple pyrimidines include one-pot reaction of β-ketoester or β-diketone, arylaldehyde and (thio)urea using a photochemical [15] or microwave irradiation [16-18]. However, because of the incessant interest in this field new efficient synthesis of novel pyrimidine derivatives are still sought [19-22]. In view of these reports and continuation of our studies on the synthesis of pyrimidines [15-18,23,24] we wish to report the synthesis of some fused pyrimidines using an intramolecular electrophilic substitution reaction.

RESULTS AND DISCUSSION

Starting ethyl 6-methyl-4-aryl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylates **1a-i** and their thioxo analogs were synthesized using literature procedures [15-18]. Intramolecular Friedel-Crafts acylation of **1** in the presence of $AlCl_3$ and acetyl chloride afforded fused oxo- and thioxopyrimidine derivatives as shown in Scheme 1. The Lewis acid, $AlCl_3$, is used as a catalyst in this reaction to produce the electrophilic species (acylium ion). However the role of acetyl chloride in this reaction is unknown. It is documented that the presence of an excess amount of acetyl chloride increases the yield of product considerably [25]. However the experiment was unsatisfactory in the absence of the acetyl chloride. We suggest that acetyl chloride, as cocatalyst, may interact with the EtO^- group and makes it as a better leaving group.



1a) G= H, Z=O
1b) G= 3-Cl, Z=O
1c) G= 2-NO_2, Z=O
1d) G= 3-NO_2, Z=O
1e) G= 4-Br, Z=O
1f) G= 4-F, Z=O
1g) G= 4-Cl, Z=O
1h) G= 4-CH_3, X=O
1i) G= 4-$(CH_3)_2CH$, X=O

1j) G= H, Z=S
1k) G= 3-Cl, Z=S
1l) G= 2-NO_2, Z=S
1m) G= 3-NO_2, Z=S
1n) G= 4-Br, Z=S
1o) G= 4-F, Z=S
1p) G= 4-Cl, Z=S
1q) G= 4-CH_3, X=S

Scheme 1

The ^{1}H NMR spectra of **2a-i** and **2j-q** are very similar to each other due to structural similarity. The ^{1}H NMR spectra data of all synthesized compounds are consistent

with their structures. For example the ^{1}H NMR spectrum of **2a** shows singlets at 2.31, 5.38, 8.18 and 9.44 ppm, which attributed to the resonance of the methyl group, CH of pyrimidine ring and two NH protons respectively. These signals are shifted downfield with respect to those of starting material due to loss of the electron releasing OEt group. Also absence of the ^{1}H NMR signals related to the OEt group resonance is a good support to the expected reaction.

EXPERIMENTAL

Melting points were determined on an electrothermal digital melting point apparatus.^{1}H NMR spectra were recorded on a Bruker (300 MHz) Spectrometer. The IR spectra were recorded on Galaxy FT-IR 500 Spectrometer. Reactions were monitored by thin layer chromatography. All materials were used as they received. Starting pyrimidine compounds **1a-q** were synthesized using literature procedures [15-18].

General procedure for the synthesis of 2. A mixture of 6-methyl-4-aryl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate or 6-methyl-4-aryl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (0.001 mol), **1**, anhydrous aluminum chloride (0.005 mol), acetyl chloride (0.005 mol) and nitrobenzene (2 ml) was heated at 90 °C for 4-5 h. The solution was poured into 15-20 ml of ice-cooled water, 5 ml hydrochloric acid (37%) and 15 ml petroleum ether were added, and stirred for 3 h. The precipitate was collected by filtration and washed with THF to give pure product **2**.

4-Methyl-1*H*-indeno[1,2-*d*]pyrimidine-2,5(3*H*,9b*H*)-dione (2a). Yield 45%, mp 230-231 °C; IR (KBr): ν= 3345, 3284, 1700, 1685, 1630cm^{-1}; ^{1}H NMR (DMSO-d$_6$): δ (ppm) = 2.31 (s, 3H, CH$_3$), 5.38 (s, 1H, CH), 7.48-7.87 (m, 4H, H$_{arom}$), 8.18 (bs, 1H, NH), 9.44 (bs, 1H, NH); ^{13}C-NMR (DMSO-d$_6$):δ (ppm) = 13.9, 51.3, 107.2, 121.9, 124.5, 128.0, 132.6, 139.1, 146.1, 146.2, 154.8, 186.6. *Anal* Calcd. for $C_{12}H_{10}N_2O_2$: C, 67.29; H, 4.67; N, 13.08%. Found: C, 67.51; H, 4.79; N, 12.85%.

8-Chloro-4-methyl-1*H*-indeno[1,2-*d*]pyrimidine-2,5(3*H*,9b*H*)-dione (2b). Yield 51%, mp 273-274 °C; IR (KBr): ν= 3345, 3284, 1707, 1680, 1628 cm^{-1}; ^{1}H NMR (DMSO-d$_6$): δ (ppm) = 2.29 (s, 3H, CH$_3$), 5.38 (s, 1H, CH), 7.50-8.21 (m, 3H, H$_{arom}$), 8.27 (bs, 1H, NH), 9.62 (bs, 1H, NH); ^{13}C-NMR (DMSO-d$_6$):δ (ppm) = 13.9, 51.0, 106.9, 123.7, 124.8, 128.4, 137.2, 137.9, 146.8, 147.9, 154.5, 185.2. *Anal* Calcd. for $C_{12}H_9N_2O_2Cl$: C, 57.72; H, 4.01; N, 11.22%. Found: C, 57.50; H, 4.16; N, 11.53%**.**

4-Methyl-9-nitro-1*H*-indeno[1,2-*d*]pyrimidine-2,5(3*H*,9b*H*)-dione (2c). Yield 51%, mp 295-296 °C; IR (KBr): ν= 3320, 3202, 3049, 2931, 1706, 1696, 1670 cm^{-1}; ^{1}H NMR (DMSO-d$_6$): δ (ppm) = 2.27 (s, 3H, CH$_3$), 5.78 (s, 1H, CH), 7.31-7.87 (m, 3H, H$_{arom}$), 8.23 (bs, 1H, NH), 9.29 (bs, 1H, NH); ^{13}C-NMR (DMSO-d$_6$): δ (ppm) = 16.9, 48.9, 107.5, 117.5, 123.2, 127.8, 128.0, 128.2, 129.1, 133.3, 138.6, 183.8. *Anal* Calcd. for $C_{12}H_9N_3O_4$: C, 55.60; H, 3.47; N, 16.21%. Found: C, 55.49; H, 3.67; N, 16.30%**.**

4-Methyl-8-nitro-1*H*-indeno[1,2-*d*]pyrimidine-2,5(3*H*,9b*H*)-dione (2d). Yield 60%, mp 282-283 °C. IR (KBr): ν= 3229, 3126, 3050, 2937, 1710, 1699 cm^{-1}; ^{1}H NMR (DMSO-d$_6$): δ (ppm) = 2.26 (s, 3H, CH$_3$), 5.28 (s, 1H, CH), 7.63-8.15 (m, 3H, H$_{arom}$), 8.22 (bs, 1H, NH), 9.26 (bs, 1H, NH); ^{13}C-NMR (DMSO-d$_6$): δ (ppm) = 18.3, 53.9, 99.4, 121.3, 122.7, 130.6, 133.4, 147.3, 148.2, 149.2, 152.5, 167.4. *Anal* Calcd. for $C_{12}H_9N_3O_4$: C, 55.60; H, 3.47; N, 16.21%. Found: C, 55.71; H, 3.78; N, 16.51%.

7-Bromo-4-methyl-1*H*-indeno[1,2-*d*]pyrimidine-2,5(3*H*, 9b*H*)-dione (2e). Yield 51%, mp 265-266 °C; IR (KBr): ν= 3248, 3109, 2933, 1707, 1641, 1529 cm^{-1}; ^{1}H NMR (DMSO-d$_6$): δ (ppm) = 2.13 (s, 3H, CH$_3$), 5.34 (s, 1H, CH), 7.77-7.87 (m, 3H, H$_{arom}$), 8.26 (bs, 1H, NH), 9.262 (bs, 1H, NH); ^{13}C-NMR (DMSO-d$_6$): δ (ppm) = 15.5, 51.6, 107.5, 122.2, 125.2, 127.4, 135.8, 141.9, 145.7, 148.0, 155.2, 185.6. *Anal* Calcd. for $C_{12}H_9N_2O_2Br$: C, 49.15; H, 3.07; N, 9.56%. Found: C, 49.43; H, 3.32; N, 9.21%.

7-Fluoro-4-methyl-1*H*-indeno[1,2-*d*]pyrimidine-2,5(3*H*, 9b*H*)-dione (2f). Yield 52%, mp 283-284 °C; IR (KBr): ν= 3248, 3200, 3138, 2964, 1716, 1680, 1612 cm^{-1}; ^{1}H NMR (DMSO-d$_6$): δ (ppm) = 2.30 (s, 3H, CH$_3$), 5.35 (s, 1H, CH), 7.38-7.98 (m, 3H, H$_{arom}$), 8.27 (bs, 1H, NH), 9.62 (bs, 1H, NH); Anal Calcd. for $C_{12}H_9N_2O_2F$: C, 62.07; H, 3.88; N, 12.07%. Found: C, 62.34; H, 3.49; N, 11.77%.

7-Chloro-4-methyl-1*H*-indeno[1,2-*d*]pyrimidine-2,5(3*H*, 9b*H*)-dione (2g). Yield 55%, mp 280-281 °C; IR (KBr): ν= 3327, 3294, 3021, 2981, 1711, 1693, 1621 cm^{-1}; ^{1}H NMR (DMSO-d$_6$): δ (ppm) = 2.19 (s, 3H, CH$_3$), 5.32 (s, 1H, CH), 7.21-7.63 (m, 3H, H$_{arom}$), 8.20 (bs, 1H, NH), 9.87 (bs, 1H, NH). *Anal* Calcd. for $C_{12}H_9N_2O_2Cl$: C, 57.95; H, 3.62; N, 11.27. Found: C, 57.32; H, 3.79; N, 11.01%.

4,7-Dimethyl-1*H*-indeno[1,2-*d*]pyrimidine-2,5(3*H*,9b*H*)-dione (2h). Yield 65%, mp 273-274 °C; IR (KBr): ν= 3310, 3335, 3290, 3005, 1712, 1686, 1615 cm^{-1}; ^{1}H NMR (DMSO-d$_6$): δ (ppm) = 1.90 (s, 3H, CH$_3$), 2.28 (s, 3H, CH$_3$), 5.30 (s, 1H, CH), 7.46-8.17 (m, 3H, H$_{arom}$), 8.18 (bs, 1H, NH), 9.47 (bs, 1H, NH); ^{13}C-NMR (DMSO-d$_6$): δ (ppm) = 15.1, 21.0, 51.6, 107.5, 122.2, 129.4, 137.2, 138.8, 142.9, 145.7, 147.0, 155.2, 187.2. *Anal* Calcd. for $C_{13}H_{12}N_2O_2$: C, 68.42; H, 5.26; N, 12.28. Found: C, 68.16; H, 5.55; N, 12.53%.

7-Isopropyl-4-methyl-1*H*-indeno[1,2-*d*]pyrimidine-2,5(3*H*, 9b*H*)-dione (2i). Yield 69%, mp 263-264 °C; IR (KBr): ν= 3290, 3190, 2995, 1720, 1690, 1610 cm^{-1}; ^{1}H NMR (DMSO-d$_6$): δ (ppm) = 1.49 (s, 6H, 2 x CH$_3$ ipropyl), 1.84 (s, 3H, CH$_3$), 2.7 (m, 1H, CH, isopropyl), 5.10 (s, 1H, CH), 6.45-6.83 (m, 3H, H$_{arom}$), 9.28 (bs, 1H, NH), 10.05 (bs, 1H, NH). *Anal* Calcd. for $C_{15}H_{16}N_2O_2$: C, 70.31; H, 6.25; N, 10.94. Found: C, 70.52; H, 6.03; N, 11.15%.

2,3-Dihydro-4-methyl-2-thioxo-1*H*-indeno[1,2-*d*]pyrimidine-5(9b*H*)-one (2j). Yield 49%, mp 225-226 °C; IR (KBr): ν= 3300, 3250, 1700, 1680, 1620 cm^{-1}; ^{1}H NMR (DMSO-d$_6$): δ (ppm) = 2.31 (s, 3H, CH$_3$), 5.37 (s, 1H, CH), 7.24-8.04 (m, 4H, H$_{arom}$), 10.10 (bs, 1H, NH), 10.67 (bs, 1H, NH); ^{13}C-NMR (DMSO-d$_6$): δ (ppm) = 15.1, 52.5, 67.4, 123.7, 126.8, 128.9, 130.3, 132.8, 135.7, 147.4, 158.6, 186.7. *Anal* Calcd. for $C_{12}H_{10}N_2OS$: C, 62.61; H, 4.35; N, 12.17%. Found: C, 62.79; H, 4.60; N, 11.96%.

8-Choloro-2,3-dihydro-4-methyl-2-thioxo-1*H*-indeno[1,2-*d*]pyrimidine-5(9b*H*)-one (2k). Yield 47%, mp 254-255 °C; IR (KBr): ν= 3420, 3140, 3050, 2972, 1662, 1560 cm^{-1}; ^{1}H NMR (DMSO-d$_6$): δ (ppm) = 2.27 (s, 3H, CH$_3$), 5.23 (s, 1H, CH), 7.23-7.71 (m, 3H, H$_{arom}$), 7.86 (bs, 1H, NH), 8.23 (bs, 1H, NH). *Anal* Calcd. for $C_{12}H_9N_2OSCl$: C, 54.44; H, 3.40; N, 10.59%. Found: C, 54.26; H, 3.15; N, 10.75%.

2,3-Dihydro-4-methyl-9-nitro-2-thioxo-1*H*-indeno[1,2-*d*]-pyrimidine-5(9b*H*)-one (2l). Yield 45%, mp 289-290 °C; IR (KBr): ν= 3491, 3306, 1674, 1523 cm^{-1}; ^{1}H NMR (DMSO-d$_6$): δ (ppm) = 2.28 (s, 3H, CH$_3$), 5.91 (s, 1H, CH), 7.49-7.77 (m, 3H,

H_{arom}), 7.91 (bs, 1H, NH), 8.25 (bs, 1H, NH); *Anal* Calcd. for $C_{12}H_9N_3O_3S$: C, 52.36; H, 3.72; N, 15.27%. Found: C, 52.03; H, 3.49; N, 15.57%.

2,3-Dihydro-4-methyl-8-nitro-2-thioxo-1*H*-indeno[1,2-*d*]-pyrimidine-5(9b*H*)-one (2m). Yield 45%, mp 262-263 °C; IR (KBr): ν= 3345, 3412, 2922, 2850, 1655, 1525 cm^{-1}; ^1H NMR (DMSO-d_6): δ (ppm) = 2.31 (s, 3H, CH_3), 5.31 (s, 1H, CH), 7.65-8.26 (m, 3H, H_{arom}), 9.47 (bs, 1H, NH), 10.59 (bs, 1H, NH). ^{13}C-NMR (DMSO-d_6): δ (ppm) = 21.2, 57.4, 104.5, 125.0, 126.0, 126.7, 133.9, 134.4, 137.0, 149.5, 170.7, 178.5. *Anal* Calcd. for $C_{12}H_9N_3O_3S$: C, 52.36; H, 3.72; N, 15.27%. Found: C, 52.66; H, 3.51; N, 15.46%.

7-Bromo-2,3-dihydro-4-methyl-2-thioxo-1*H*-indeno-[1,2-*d*]pyrimidine-5(9b*H*)-one (2n). Yield 47%, mp 349-350 °C; IR (KBr): ν= 3350, 3248, 3109, 2933, 1703, 1641, 1529 cm^{-1}; ^1H NMR (DMSO-d_6): δ (ppm) = 2.27 (s, 3H, CH_3), 5.12 (s, 1H, CH), 7.18-7.75 (m, 3H, H_{arom}), 9.72 (bs, 1H, NH), 10.31 (s, 1H, NH). *Anal* Calcd. for $C_{12}H_9N_2OSBr$: C, 46.60; H, 2.91; N, 9.06%. Found: C, 46.41; H, 3.26; N, 9.29%.

7-Fluoro-2,3-dihydro-4-methyl-2-thioxo-1*H*-indeno[1,2-*d*]-pyrimidine-5(9b*H*)-one (2o). Yield 47%, mp 342-343 °C; IR (KBr): ν= 3245, 3120, 3040, 2940, 1705 cm^{-1}; ^1H NMR (DMSO-d_6): δ (ppm) = 2.27 (s, 3H, CH_3), 5.13 (s, 1H, CH), 7.15-7.44 (m, 3H, H_{arom}), 9.62 (bs, 1H, NH), 10.31 (bs, 1H, NH). *Anal* Calcd. for $C_{12}H_9N_2OSF$: C, 58.06; H, 3.63; N, 11.29%. Found: C, 58.27; H, 3.39; N, 11.44%.

7-Chloro-2,3-dihydro-4-methyl-2-thioxo-1*H*-indeno[1,2-*d*]-pyrimidine-5(9b*H*)-one (2p). Yield 46%, mp 319-320 °C; IR (KBr): ν= 3245, 3120, 3040, 2940, 1760, 1705 cm^{-1}; ^1H NMR (DMSO-d_6): δ (ppm) = 2.11 (s, 3H, CH_3), 5.12 (s, 1H, CH), 7.21-7.89 (m, 3H, H_{arom}), 8.32 (bs, 1H, NH), 9.21 (bs, 1H, NH); ^{13}C-NMR (DMSO-d_6): δ (ppm) = 21.5, 50.2, 104.2, 115.5, 115.8, 125.3, 128.8, 129.0, 135.4, 143.3, 172.4, 178.3. *Anal* Calcd. for $C_{12}H_9N_2OSCl$: C, 54.44; H, 3.40; N, 10.59. Found: C, 54.13; H, 3.65; N, 10.82%.

2,3-Dihydro-4,7-dimethyl-2-thioxo-1*H*-indeno[1,2-*d*]pyrimidine-5(9b*H*)-one (2q). Yield 53%, mp 259-260 °C; IR (KBr): ν= 3371, 3190, 3040, 2940, 1685 cm^{-1}; ^1H NMR (DMSO-d_6): δ (ppm) = 2.30 (s, 3H, CH_3), 2.39 (s, 3H, CH_3), 5.32 (s, 1H, CH), 7.49-7.90 (m, 3H, H_{arom}), 10.09 (bs, 1H, NH), 10.67 (bs, 1H, NH); ^{13}C-NMR (DMSO-d_6): δ (ppm) = 14.7, 21.3, 52.8, 108.7, 123.3, 126.2, 135.1, 139.3, 139.8, 143.6, 144.0, 179.4, 186.2. *Anal* Calcd. for $C_{13}H_{12}N_2OS$: C, 63.93; H, 4.91; N, 11.48. Found: C, 64.27; H, 4.85; N, 11.53%.

CONCLUSION

Some novel fused oxo- and thiopyrimidines including 1*H*-indeno[1,2-*d*]pyrimidine-2,5(3*H*,9b*H*)-dione and 2,3-dihydro-2-thioxo-1*H*-indeno[1,2-*d*]pyrimidine-5(9b*H*)-one derivatives were synthesized *via* an intramolecular Friedel-Crafts reaction under mild conditions. The Lewis acid, $AlCl_3$, was used as a catalyst in this reaction to produce the acylium ion.

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