New Approach for the Synthesis of Pyrido[1,2-a]pyrimidines

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A novel method was successfully demonstrated towards the synthesis of pyrido[1,2-a]pyrimidines having chloroethyl as an intractable side chain, through dihydrofuranone intermediates. The dihydrofuranone intermediates were synthesized by condensation of 2-aminopyridines with α-acetyl-γ-butyrolactone, which upon cyclization using phosphorus oxychloride or ethanol in sodium ethoxide furnished the pyrido[1,2-a]pyrimidines in good yield.

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1. Introduction

Heterofused pyrimidines exhibit promising antiviral [1], antibacterial [2], anti-AIDS [3], and antinociceptive [4] activities. Fused pyrimidines are extensively used in neurology, particularly in the treatment of neurodegenerative disorders such as Parkinson’s disease [5], antianxiety disorders [6], and depression [7]. Fused pyrimidines are selective inhibitors for multidrug resistance (MDR) [8]. Folate metabolism has long been recognized as an attractive target for cancer chemotherapy because of indispensable role of fused pyrimidines’ antifolates as antitumor agents [9]. Resperdone is a derivative of pyrido[1, 2-a]pyrimidines [10, 11]. These compounds showed antipsychotic activity [12] were used as α2 antagonists [13–15]. They exhibits high affinity for α2-adrenoceptor with high selectivity versus the α-receptor and possesses potent in vivo central activity [16, 17]. Wamhoff and Korte [18, 19] have reported the synthesis of pyrido[1,2-a]pyrimidines by using 2-aminothiocarboxylic compounds and α-acetyl-γ-butyrolactone by refluxing dioxane or using PPA, and observed the formation of mixture of products. After the successful study of the reactions of α-formyl-γ-butyrolactone with 2-aminothioketones [20], our interest was sparked to synthesize old compounds by new method with improved yields.

2. Results and Discussion

In the present communication, we have developed new methodology towards the synthesis of pyrido[1,2-a]pyrimidines with 3-hydroxy or 3-chloroethyl side chain. Our method gives single product with high yield. Thus, 2-aminopyridines 1 and α-acetyl-γ-butyrolactone 2 were refluxed in toluene in presence of catalytic amount of PTSA using Dean-Stark apparatus (furnished dihydrofuranone intermediate 3) in 80–85% yield. Here, PTSA is selective catalyst for protonation of carbonyl carbon to make NH2 attack more favorable. In presence of other acid catalysts such as conc. HCl, conc. H2SO4 yields mixture of products as these acids also protonated the ester carbonyl.

The present work gives single product with high yield. Thus, 2-aminopyridines 1 and α-acetyl-γ-butyrolactone 2 were refluxed in toluene in presence of catalytic amount of PTSA using Dean-Stark apparatus (furnished dihydrofuranone intermediate 3) in 80–85% yield. Here, PTSA is selective catalyst for protonation of carbonyl carbon to make NH2 attack more favorable. In presence of other acid catalysts such as conc. HCl, conc. H2SO4 yields mixture of products as these acids also protonated the ester carbonyl. The structure of furanone 3 was characterized by spectral and analytical methods. The IR of 3a showed the bands at 3200 and 1690 cm⁻¹ for NH and lactone CO stretching frequencies; the lactone carbonyl frequency lowered by 30–40 cm⁻¹ due to hydrogen bonding between CO and NH groups. The 1H NMR of 3a showed two triplets at δ 2.92 and 4.31 for –CH2 and –OCH2 protons, respectively, with J = 7 Hz. The singlet appeared at δ 2.50 for CH3 protons as it is attached to olefinic carbon.

The broad singlet appeared at δ 10.50 corresponding to NH proton (exchangeable with D2O). The 13C NMR spectrum of this compound exhibits peaks at δ19.48, 26.26, 65.43, 93.62, 113.9, 117.51, 137.86, 147.83, 152.41, 153.52, and 173.72. Further, the structure of 3a was assigned by HMBC and COSY experiments, which is in agreement with the structure proposed. We have also tried the above reactions in ammonium acetate without using any solvent. Thus, 2-aminopyridine and α-acetyl-γ-butyrolactone were fused in ammonium acetate at 120°C, and the reaction mixture was stirred in water to remove excess of ammonium
acetate and unreacted pyridine if any to yield 4 in 56% yield. Interestingly, it was observed that in ammonium acetate the 3-hydroxyethyl side chain gets acylated to yield 3-acetyl derivative 4. The ammonium acetate worked as acetylating agent, and reaction demonstrates green approach for the synthesis of pyrido[1,2-a]pyrimidines. Similarly, the structure of compound 4 was characterized by spectral and analytical methods. The IR of 4 shows peaks at 1670 (amide) and 1740 (ester) cm$^{-1}$. The $^1$H NMR spectrum of 4 showed two singlets at $\delta$ 2 and 2.52 for OCH$_3$ and CH$_3$ protons, and two triplets at $\delta$ 3.03 and 4.27 for –CH$_2$ and –OCH$_2$ protons, respectively. The multiplet appeared at $\delta$ 7.07–8.90 corresponding to 4 aromatic protons. The $^{13}$C NMR spectrum of this compound exhibits peaks at $\delta$ 21.06, 22.72, 26.58, 62.59, 111.46, 114.98, 125.83, 127.06, 135.30, 148.79, 157.87, 162.62, and 171.06. Further, the structure of 4 was assigned by HMBC and COSY experiments, which is in agreement with the structure proposed. The compound 4 was cyclized to target pyrido[1,2-a]pyrimidines 5 having 3-hydroxyethyl side chain by refluxing in weak or strong base like NH$_3$ or NaOH, or was cyclized to 6 having 3-chloroethyl side chain in refluxing in POCl$_3$. These cyclized compounds 5 and 6 were characterized by IR, $^1$H NMR, and elemental analysis. Thus, in the IR spectrum of 5, the bands at 1690 and 3050 cm$^{-1}$ due to lactone and NH stretching in 3 disappeared, and new bands at 1670 and 3260 cm$^{-1}$ for OH and C=O groups were observed. The $^1$H NMR showed the peaks at $\delta$ 3.01 and 3.92 with $J$ = 7 Hz for –CH$_2$ and –OCH$_2$ protons. The additional broad singlet that appeared at 4.60$\delta$ is due to OH group.

The compound 3 or 5 when refluxed in POCl$_3$ to finish the pyridopyrimidines 6 having 3-chloroethyl side chain. These compounds were characterized by comparing their mp and spectroscopic data with those of literature-reported compounds [19]; further the product was confirmed by preparing its azido derivative 7, with NaN$_3$ in DMF, which shows characteristic stretching frequency at 2100 cm$^{-1}$ for N$_3$. The elimination of terminal halide in 6 was carried out using strong base to yield pyrido[1,2-a]pyrimidine 8 with 3-ethylene side chain. Compound 8 shows IR stretching at 1630 cm$^{-1}$ for carbon-carbon double bond. Two doublets were observed at $\delta$ 5.63 with $J$ = 16 Hz and at $\delta$ 6.55 with $J$ = 8 Hz for two olefinic protons due to cis- and trans-coupling. The =C–H proton appears as triplet at 6.83 with $J$ = 16 and 8 Hz.

3. Conclusion
The work demonstrates the new methodology towards the synthesis of pyrido[1,2-a]pyrimidines having intractable chloroethyl side chain using cyclic $\beta$-ketoesters and 2-aminopyridines. The new methods furnish improved yields up to 80–85% of dihydrofuranone intermediate and 70–75% of cyclized pure product.

4. Experimental
Melting points were determined in open capillary tubes on a Gallenkamp melting point apparatus (Mod. MFB-595 in open capillary tubes), and were uncorrected. Infrared spectra were recorded on a Perkin-Elmer 298 spectrophotometer in potassium bromide pellets unless otherwise stated. $^1$H NMR spectra were recorded on a Varian Gemini 200 (200 MHz) spectrometer. $^{13}$C NMR spectra were recorded on a Buler AM 360 (90 MHz) spectrometer. The chemical shifts are reported in $\delta$ units relative to internal standard tetramethylsilane. Elemental analyses were performed on Fisons EA 1108 C,H,N-automatic analyzer, and were within ±0.3 of the theoretical percentage. All reactions were monitored by thin layer chromatography on 0.2 mm silica gel 60 F$_{254}$ (Merck, Mumbai, India) plates using UV light (254 and 365 nm) for detection. All the reagents were used as received from commercial sources. The solvents were dried over the 40 nm molecular sieves.

4.1. General Procedure for the Synthesis of dihydrofuranone-2-(3H)-one (3a–3c). The mixture of 2-aminopyridines 1 (10 mmol) and $\alpha$-acetyl $\gamma$-butyrolactone 2 (10 mmol, 1.28 g, or 1.10 mL) was refluxed in toluene (30 mL) for 24 hours in presence of catalytic amount of PTSA (0.02 g). The water separator was attached between the reaction flask and the water condenser. The separation of equivalent amount of water indicates the completion of reaction. The solid obtained after cooling the reaction mixture was filtered and washed with toluene and then recrystallized in suitable solvent.

4.2. 3-[1-(Pyridin-2-ylamino)ethylidene]dihydrofuran-2(3H)-one (3a, C$_{11}$H$_{12}$N$_2$O$_2$). Yield 1.69 g (83%), mp 87–88°C (ligroin, colorless flakes); IR: (KBr): 3200 (b, NH), 1610, 1560, 1480, 1450, cm$^{-1}$; $^1$H NMR: (CDCl$_3$) $\delta$ = 2.50 (3H, CH$_3$), 2.92 (t, $J$ = 7 Hz, 2H, CH$_2$), 4.31 (t, $J$ = 7 Hz, 2H, OCH$_2$), 6.75 (dd, $J$ = 2.1 & 8.1 Hz, 1H, C$_2$-H), 6.83 (dt, $J$ = 2.4 & 8.3 Hz, 1H, C$_3$-H), 8.25 (dd, $J$ = 2.4 & 8.3 Hz, 1H, C$_5$-H), 10.5 (bs, 1H, NH) ppm. $^{13}$C NMR (CDCl$_3$): 19.48 (CH$_3$), 26.26 (CH$_2$), 65.43 (CH$_2$O), 93.62 (C=O), 113.99 (C$_3$), 117.91 (C$_4$); 137.86 (C$_6$); 147.83 (C$_2$); 153.22 (C$_3$); 173.72 (C=O) ppm.

4.3. 3-[1-[(6-Methylpyridin-2-yl)amino]ethylidene] dihydrofuran-2(3H)-one (3b, C$_{11}$H$_{14}$N$_2$O$_2$). Yield 1.74 g (80%), mp 105–106°C (cyclohexane, colorless flakes); IR (KBr): 3200 (NH), 2920, 2860(w, CH$_3$), 1690 (C=O lactone), 1640, 1600(C=C), 1570, 1500, 1480, 1410 cm$^{-1}$; $^1$H NMR (CDCl$_3$): $\delta$ = 2.40 (s, 3H, CH$_3$), 3.95 (t, $J$ = 7 Hz, 2H, CH$_2$), 4.42 (t, $J$ = 7.1 Hz, 2H, OCH$_2$), 6.63 (dd, $J$ = 7.8 & 2.8 Hz, 1H, C$_3$–H), 7.45 (t, $J$ = 7.8 & 7.8 Hz, 1H, C$_5$–H), 6.70 (d, $J$ = 7.9 & 2.9 Hz, 1H, C$_7$–H); 10.55 (bs, 1H, NH) ppm.

4.4. 3-[1-[(4-Methylpyridin-2-yl)amino]ethylidene] dihydrofuran-2(3H)-one (3c, C$_{11}$H$_{14}$N$_2$O$_2$). Yield 1.85 g (85%), mp 72–73°C (cyclohexane, colorless flakes); IR (KBr): 3200 (b, NH), 2910, 2880(w, CH$_3$), 1780 (s, C=O lactone), 1650 (s, NH), 1610, 1560, 1480, 1450 cm$^{-1}$; $^{13}$C NMR (CDCl$_3$) 62.33 (s, 3H, CH$_3$), 2.52 (s, 3H, CH$_3$), 2.92 (t, $J$ = 7 Hz, 2H, CH$_2$), 4.36 (t, $J$ = 7 Hz, 2H, OCH$_2$), 6.75 (s, 1H, C$_3$–H), 7.07–7.17 (s, 1H, C$_7$–H).
6.75 (d, J = 8 Hz, 1H, C5–H); 8.19 (d, J = 8 Hz 1H, C6–H); 10.52 (bs, 1H, NH) ppm.

4.5. General Procedure for Synthesis of 2-(2-methyl-4-oxo-4H-pyrido[1,2-a]pyrimidin-3-yl)ethyl acetate (4C13H14N2O3). The mixture of 2-aminopyridine (10 mmol) and α-acetyl γ-butyrolactone (20 mmol, 1.28 g, or 1.10 mL) was heated in ammonium acetate (0.05 mol, 3.85 g) at 100–120°C for 1–2 hours. The reaction mixture was stirred in water, and extracted with chloroform. The chloroform extract was dried over anhydrous sodium sulfate, and evaporated to furnish anhydrous sodium sulphate, and evaporated to furnish a colorless solid, and the compounds were recrystallized in suitable solvent. The furanone (10 mmol) was dissolved in ethanol (20 mL), and the solution was refluxed with sodium ethoxide (0.23 g of freshly cut sodium metal was dissolved in 10 mL of dry ethanol) for 1 hour. The solvent was removed under reduced pressure. The residue was dissolved in cold water (100 mL) and extracted with chloroform. The chloroform was dried over sodium sulfate and evaporated to yield 5 in good yield.

4.5.1. Method A. The furanone 3 (10 mmol) was dissolved in ethanol (20 mL), and the solution was refluxed with sodium ethoxide (0.23 g of freshly cut sodium metal was dissolved in 10 mL of dry ethanol) for 1 hour. The solvent was removed under reduced pressure. The residue was dissolved in cold water (100 mL) and extracted with chloroform. The chloroform was dried over sodium sulfate and evaporated to yield 5 in good yield.

4.5.2. Method B. The compound 4 (10 mmol) was refluxed in ethanol in presence of catalytic amount of sodium hydroxide for about 3–4 hours (TLC check). The excess of solvent was removed under reduced pressure, and the obtained precipitate was dissolved in water and stirred for 1 hour. Then, it was filtered, washed with water, dried, and recrystallized from proper solvent to afford 5 in good yield.

4.6. General Procedure for the Synthesis of 3-(2-hydroxyethyl)-2-methyl-4H-pyrido[1,2-a]pyrimidin-4-one (5a, 5b)

4.6.1. Method A. The furanone 3 (10 mmol) was dissolved in ethanol (20 mL), and the solution was refluxed with sodium ethoxide (0.23 g of freshly cut sodium metal was dissolved in 10 mL of dry ethanol) for 1 hour. The solvent was removed under reduced pressure. The residue was dissolved in cold water (100 mL) and extracted with chloroform. The chloroform was dried over sodium sulfate and evaporated to yield 5 in good yield.

4.6.2. Method B. The compound 4 (10 mmol) was refluxed in ethanol in presence of catalytic amount of sodium hydroxide for about 3–4 hours (TLC check). The excess of solvent was removed under reduced pressure, and the obtained precipitate was dissolved in water and stirred for 1 hour. Then, it was filtered, washed with water, dried, and recrystallized from proper solvent to afford 5 in good yield.

4.7. 3-(2-hydroxyethyl)-2-methyl-4H-pyrido[1,2-a]pyrimidin-4-one (5a, C11H13N2O2). Yield 1.42 g (70%), mp 158–159°C (cyclohexane, colorless needles); IR (KBr): 3260 (b, OH), 2880 (w, CH3), 1670 (s, C=O lactone), 1640(s, C=O lactone), 1640(s, NH), 1530, 1470, 1440, cm⁻¹; 1H NMR (CDCl3): δ = 2.62 (s, 3H, CH3), 2.93 (t, J = 7.1 Hz, 2H, CH2), 3.96 (t, J = 7.1 Hz 2H, CH2), 7.17 (m, 1H, C8-H), 7.43 (bs, 1H, OH); 7.65 (d, J = 8.1 Hz 1H, C7-H); 9.09 (d, J = 8.3 Hz, 1H, C3-H) ppm.

4.8. 3-(2-hydroxyethyl)-2,6-dimethyl-4H-pyrido[1,2-a]pyrimidin-4-one (5b, C12H16N2O2). Yield 1.54 g (71%), mp 175–176°C (ligroin, colorless needles); IR (KBr): 3250 (b, OH), 2920, 2880(w, CH3), 1670 (s, C=O lactone), 1640, 1570, 1520,1470, cm⁻¹; 1H NMR (CDCl3): δ = 2.46 (s, 3H, CH3), 2.52 (s, 3H, CH3), 3.06 (t, J = 7 Hz, 2H, CH2), 3.52 (s, 1H, OH), 3.92 (t, J = 7 Hz, 2H, OCH2), 6.96 (dd, J = 8.3 Hz, 1H, C3–H) ppm.
C–H); 7.22 (d, J = 8.1 Hz, 1H, C6–H); 8.89 (d, J = 8 Hz, 1H, C7–H) ppm.

4.9. General Procedure for the Synthesis of 3-(2-chloroethyl)-2-methyl-4H-pyrido[1,2-a]pyrimidin-4-one (6a–6c). Compound 3 or 5 (10 mmol) was refluxed in phosphorus oxychloride (20 mL) for 1 hour (TLC check). After completing the reaction, the excess of phosphorus oxychloride was removed under reduced pressure. The residue obtained was stirred in ice-cold water (100 mL) for 30 minutes, then neutralized with solid sodium carbonate and further stirred overnight. The solid precipitated was filtered and washed with water and recrystallized from suitable solvent.

4.10. 3-(2-chloroethyl)-2-methyl-4H-pyrido[1,2-a]pyrimidin-4-one (6a, C13H11ClN2O). Yield 1.60 g (72%), mp 140–142°C (cyclohexane, colorless needles); IR (KBr): 2950 (w, CH3), 1670 (s, C=O lactone), 1540, 1480, 1430 cm−1; 1H NMR (CDCl3): δ = 2.55 (s, 3H, CH3); 3.23 (t, J = 7 Hz, 2H, CH2); 3.88 (t, J = 7 Hz, 2H, CH2Cl); 7.15 (t, J = 8 Hz 1H, C8–H); 7.69 (d, J = 8 Hz 1H, C7–H); 7.78 (t, J = 8Hz 1H, C7–H); 9.07 (d, 1H, C=O–H) ppm.

4.11. 3-(2-chloroethyl)-2,6-dimethyl-4H-pyrido[1,2-a]pyrimidin-4-one (6b, C12H13ClN2O). Yield 1.65 g (70%), mp 113–114°C (cyclohexane, colorless needles); IR (KBr): 2980 (w, CH3), 1670 (s, C=O lactone), 1640, 1580, 1540, 1470, 1440 cm−1; 1H NMR (CDCl3): δ = 2.55 (s, 3H, CH3); 3.09 (s, 3H, CH3); 3.18 (t, J = 7 Hz, 2H, CH2); 3.78 (t, J = 7 Hz, 2H, CH2Cl); 7.38 (t, J = 8.3 Hz, 1H, C8–H); 6.66 (d, J = 8 Hz, 1H, C7–H) ppm.

4.12. 3-(2-chloroethyl)-2,8-dimethyl-4H-pyrido[1,2-a]pyrimidin-4-one (6c, C12H13ClN2O). Yield 1.77 g (75%), mp 116–117°C (cyclohexane, colorless needles); IR (KBr): 1670 (s, C=O lactone), 1640, 1560, 1530, 1470, 1440 cm−1; 1H NMR (CDCl3): δ = 2.42 (s, 3H, CH3); 2.53 (s, 3H, C6–H); 3.24 (t, J = 7 Hz, 2H, CH2); 3.89 (t, J = 7 Hz, 2H, CH2Cl); 6.92 (d, J = 8.2 Hz, 1H, C7–H); 8.96 (d, J = 8Hz, 1H, C6–H); 7.41 (d, J = 2.8 Hz, 1H, C8–H) ppm.

4.13. General procedure for the synthesis of 3-(2-azidoethyl)-2-methyl-4H-pyrido[1,2-a]pyrimidin-4-one (7, C13H11N3O). To a stirred solution of 6 (10 mmol) in DMF/H2O (9:1), the sodium azide (2.60 g, 40 mmol) was added and temperature was raised slowly to 80°C. The mixture was kept at this temperature for about 2 hours until TLC showed no more starting material. The temperature was raised to 110°C for 1 hour, and then the solvent was removed under reduced pressure; an oily residue was poured in ice-cold water and stirred for 1 hour; the solid obtained was filtered, washed with water, dried, and recrystallized from the proper solvent to afford 7 in good yield.

Yield 1.60 g (70%), mp 65–66°C (cyclohexane, colorless solid); IR (KBr): 2980(w, CH3), 2100(N3), 1670(s, C=O lactone), 1640, 1560, 1530, 1470, cm−1.

4.14. General procedure for the synthesis of 2-methyl-3-vinyl-4H-pyrido[1,2-a]pyrimidin-4-one (8, C12H10N2O). To a stirred solution of 6 (10 mmol) in ethanol, the solution of sodium ethoxide (prepared by reacting 0.27 gms, 11.5 mg atoms of sodium in 50 mL of absolute ethanol) was added. The reaction mixture was refluxed for about 10–11 hours. (TLC check). Then, the excess of solvent was removed under reduced pressure, and the obtained solid was filtered, washed with ethanol, dried, and recrystallized from the proper solvent to afford 8 in good yield.

Yield 1.20 g (60%), mp 110–111°C (cyclohexane, colorless solid) IR (KBr): 3020(w, CH3), 1780(s, C=O lactone), 1640, 1560, 1530, 1470, cm−1; 1H NMR (CDCl3): δ = 2.61 (s, 3H, CH3); 5.63 & 6.55 (dd, J = 16, 8 Hz, 2H, CH2H); 6.83 (t, J = 16, 8 Hz, 1H, C=H); 7.13 (dt, J = 8.3 Hz, 1H, C8–H); 7.66 (dt, J = 8.3 Hz, 1H, C7–H); 7.75 (dt, J = 8.3 Hz, 1H, C6–H); 9.12 (dd, J = 8.3 Hz, 1H, C8–H) ppm.

Table 1: Elemental analysis.

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<td>4</td>
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References


