Synthesis of 2,4,6-Tri-Substituted Pyridine Derivatives in Aqueous Medium *via* Hantzsch Multi-Component Reaction Catalyzed by Cerium (IV) Ammonium Nitrate

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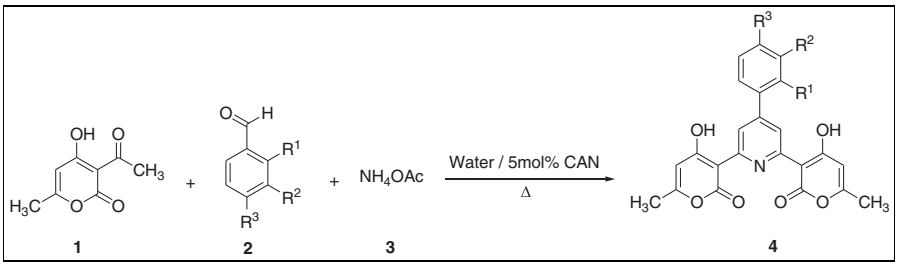

2,4,6-Tri-substituted pyridine derivatives have been efficiently synthesized in excellent yields using catalytic amounts of cerium (IV) ammonium nitrate in aqueous medium *via* one-pot, multi-component reaction. Condensation of dehydroacetic acid with aldehydes and ammonium acetate afforded corresponding tri-substituted pyridine derivatives in excellent yields by using Hantzsch pyridine synthesis. The structures of all the newly synthesized compounds were confirmed from their analytical and spectral data.

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INTRODUCTION

With regard to green chemistry, development of selective and atom-efficient transformations of easily available starting materials into complex functionalized heterocyclic building blocks become increasingly important to both pharmaceuticals and organic chemists. In this context, multi-component reactions (MCRs) involving domino processes "in which more than three different reactants directly get converted into their products by one-pot reaction" [1]. MCRs play an important role in modern organic chemistry, because they generally exhibit higher atom economy, selectivity, molecular complexity and diversity as well as produce fewer by-products compared to classical multistep synthesis [2]. The first MCR was described in 1850 by Strecker [3] and thereafter many such reactions have been reported in the literature [4].

Designing of MCRs in water is another attractive area in chemistry, [5] because water is a cheap, safe, environmentally benign solvent, and isolation of the organic products can be performed by simple phase separation. The significant enhancement in the rate of reaction has been attributed to hydrophobic packing, solvent polarity, hydration, and hydrogen bonding [6]. There are beneficial effects of aqueous solvents on rates and selectivities of important organic transformations, for example, Diels–Alder reactions, aldol reactions, and Michael additions [7,8].

Pyridine is one of the most prominent heterocycles and is widely distributed in many biologically important molecules, it was found in drugs, such as nifedipine and niguldipine undergo redox processes during their metabolism catalyzed by cytochrome P-450 in the liver [9].

Recently, cerium (IV) ammonium nitrate (CAN) is the most notable one-electron oxidant and has been utilized extensively for a broad variety of oxidative transformations in organic chemistry [10–15]. Additionally, many advantages such as excellent solubility in water, inexpensiveness, eco-friendly nature, uncomplicated handling, and high reactivity make CAN a potent catalyst in organic syntheses. Besides this, CAN is able to catalyze various organic transformations on the basis of the Lewis acidic property. These facts inspired us to use this catalyst (CAN) for the synthesis of 2,4,6-tri substituted pyridine derivatives.

A literature survey revealed that there are numerous routes reported for the synthesis of highly substituted pyridines according to modified Hantzsch pyridine synthesis [16]. These can be prepared by reaction of aldehyde, 2 moles of β-keto ester and ammonium acetate. However, these methods suffers more drawbacks such as complex work-up and purification, lower the overall yields, longer reaction times, and high boiling solvents, which are a threat to the environment. Therefore, the development of a new catalytic system to overcome these shortcomings and fulfill the criteria of a simple, efficient and environmentally benign protocol for the synthesis of highly substituted pyridines at ambient temperature is an important task for organic chemists.

On the basis of the aforementioned observations and as a part of our research program in the synthesis of novel heterocyclic systems [17–21], we wish to report in this article the

modified Hantzsch condensation of 2,4,6-tri-substituted pyridine derivatives catalyzed by CAN *via* multi-component approach in aqueous medium.

RESULTS AND DISCUSSIONS

Reaction of 3-acetyl-4-hydroxy-6-methyl-2*H*-pyran-2-one (2 mmol) with 4-nitrobenzaldehyde (1mmol) and ammonium acetate (1.5 mmol) using catalytic amount of CAN at ambient temperature in aqueous medium gave 2,4,6-tri-substituted pyridine derivatives in good yields (Scheme 1). We have carried out the reactions without any catalyst. In these cases, the 2,4,6-tri-substituted pyridine derivatives were isolated in poor yields (30–40%). Although using CAN (3 mol%) as catalyst, the reaction gave a yellow product in 90%, yield in 3 h. By changing the amount of the catalyst from 3 to 5 and 10 mol%, the reaction resulted in the formation of **4a** in 95 and 85% yields, respectively. Thus, the use of just 5 mol% of CAN was chosen as a quantitative catalyst to push the reaction forward with maximum yield of the product. We then continued to optimize the model process by detecting the efficiency of several classic solvents chosen as the medium for comparison (Table 1). Among the tested solvents, such as ethanol, acetonitrile, chloroform, and aqueous medium, the latter gave the best result.

The structures of newly prepared compounds were confirmed from their analytical and spectral data. For example, the IR spectrum of compound **4a** showed three strong absorption peaks at 3243, 1715, and 1597 cm^{-1} for OH, lactone carbonyl, and C=N, respectively. The ^1H NMR spectrum of compound **4a** showed sharp singlets at δ 2.05 and δ 2.12 for methyl groups of pyran moiety. Two singlets observed at δ 9.75 and δ 9.79 for pyridine ring protons. Like this, the remaining spectral data confirms the newly prepared compounds structures (**4a–k**).

CONCLUSION

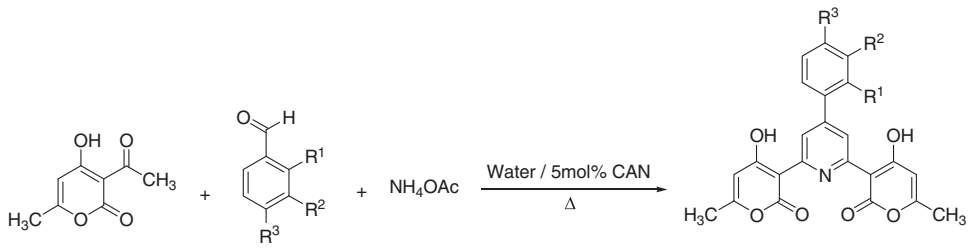
We have developed a simple, efficient, and green method for the synthesis of a variety of 2,4,6-tri-substituted pyridine derivatives *via* an improved Hantzsch reaction catalyzed by small amount of CAN. The reaction conditions are mild, and the reaction gave excellent yields of products. This method does not involve the use of volatile organic solvents and thus is an environmentally friendly process. The biological activity of these compounds is under investigation.

EXPERIMENTAL

General. All the reagents and solvents were pure, purchased from commercial sources and were used without further purification unless otherwise stated. Melting points were determined in open capillaries with a "Cintex" melting point apparatus Mumbai, India and were uncorrected. CHNS analysis was carried out by Carlo Erba EA 1108 automatic elemental analyzer (Italy). The purity of the compounds was checked using TLC plates (E. Merck Mumbai, India). IR spectra (KBr) were recorded on a Bruker WM-4(X) spectrometer (577 models). ^{1}HNMR spectra were recorded on a Bruker WM-400 spectrometer in δ ppm using TMS as standard. Mass spectra (EI-MS) were determined on Perkin Elmer (SCIEX API-2000, ESI) at 12.5 eV.

General procedure for the synthesis of compounds 4a–k. In a typical experimental procedure, dehydroacetic acid (3-acetyl-4-hydroxy-6-methyl-2H-pyran-2-one) **1** (2 mmol), aldehyde **2** (1 mmol), ammonium acetate **3** (1.5 mmol), and CAN (0.05 mmol) were taken in a 3 ml of water, the resultant mixture was refluxed for 3 h. After an appropriate time, the solid obtained was cooled, filtered, and washed with water.

Scheme 1. One-pot synthesis of 2,4,6 tri-substituted pyridine derivatives in aqueous medium.



Compd	R^1	R^2	R^3
4a	H	H	NO_2
b	H	OCH_3	OCH_3
c	H	H	Br
d	H	H	Cl
e	H	H	CH_3
f	H	H	OCH_3

Compd	R^1	R^2	R^3
g	H	H	$N(CH_3)_2$
h	H	H	OH
i	OH	H	H
j	H	NO_2	H
K	2-hydroxy-1-naphthaldehyde		

Table 1

4a. Reaction of 2-mmole dehydroacetic acid, 1-mmol 4-nitrobenzaldehyde, and 1.5-mmol ammonium acetate: effect of solvent and temperature and yields of products in water for various synthesized compounds (**4a–4k**).

Entry	Solvent	Temperature (°C)	Time (min)	Yield (%)
1	C_2H_5OH	Reflux	180	86
2	CH_3CN	Reflux	180	67
3	$CHCl_3$	Reflux	180	53
4	DMF	Reflux	180	70
4a	H_2O	Reflux	180	95
4b	H_2O	Reflux	180	86
4c	H_2O	Reflux	180	81
4d	H_2O	Reflux	180	92
4e	H_2O	Reflux	180	90
4f	H_2O	Reflux	180	87
4g	H_2O	Reflux	180	92
4h	H_2O	Reflux	180	87
4i	H_2O	Reflux	180	80
4j	H_2O	Reflux	180	86
4k	H_2O	Reflux	180	83

The crude product was purified by recrystallization from absolute ethanol.

4-Hydroxy-3-(6-(4-hydroxy-6-methyl-2-oxo-2H-pyran-3-yl)-4-(4-nitrophenyl)pyridin-2-yl)-6-methyl-2H-pyran-2-one (4a). Color: light yellow solid; mp 210–212°C; IR (potassium bromide): 3243 (–OH), 1715 (O-C=O), 1597 (C=N). 1H NMR (dimethyl sulfoxide d6): δ 2.05 (s, 3H, CH_3), 2.12 (s, 3H, CH_3), 5.73 (s, 1H, pyran), 5.82 (s, 1H, pyran), 7.23 (d, 2H, J=8.4 Hz, ArH), 7.84 (d, 2H, J=8.4 Hz, ArH), 9.75 (s, 1H, pyridine), 9.79 (s, 1H, pyridine), 12.06 (s, 1H, OH), 12.14 (s, 1H, OH). ^{13}C NMR (dimethyl sulfoxide d6): 20.4, 110.9, 130.0, 131.9, 132.0, 132.7, 133.1, 133.5, 133.8, 135.6, 152.5, 166.4, 183.1. ESI-MS 449 $[M+H]^+$; *Anal*. calcd. for $C_{23}H_{16}N_2O_8$: C, 61.61; H, 3.60; N, 6.25. Found: C,61.69; H, 3.51; N, 6.20.

4-Hydroxy-3-(6-(4-hydroxy-6-methyl-2-oxo-2H-pyran-3-yl)-4-(3,4-dimethoxyphenyl) pyridine-2-yl)-6-methyl-2H-pyran-2-one (4b). Color: orange solid; mp 287–289°C; IR (potassium bromide): 3220 (–OH), 1721 (O-C=O), 1596 (C=N); 1H NMR (dimethyl sulfoxide d6): δ 2.06 (s, 3H, CH_3), 2.09 (s, 3H, CH_3), 3.81 (s, 6H, OCH_3), 5.67 (s, 1H, pyran), 5.74 (s, 1H, pyran), 7.19 (d, 1H, J=6.8 Hz, ArH), 7.37–7.41 (m, 1H, ArH), 7.68–7.72 (m, 1H, ArH), 9.73 (s, 1H, pyridine), 9.74 (s, 1H, pyridine), 11.95 (s, 1H, OH), 12.15 (s, 1H, OH). ESI-MS 464 $[M+H]^+$; *Anal*. calcd. for $C_{25}H_{21}NO_8$: C, 64.79; H, 4.57; N, 3.02. Found: C, 64.71; H, 4.51; N, 3.14.

3-(4-(4-Bromophenyl)-6-(4-hydroxy-6-methyl-2-oxo-2H-pyran-3-yl)pyridin-2-yl)-4-hydroxy-6-methyl-2H-pyran-2-one (4c). Color: yellow solid; mp 252–254°C; IR (potassium bromide): 3217 (–OH), 1712 (O-C=O), 1598 (C=N); 1H NMR (dimethyl sulfoxide d6): δ 1.92 (s, 3H, CH_3), 2.07 (s, 3H, CH_3), 5.67 (s, 1H, pyran), 5.77 (s, 1H, pyran), 7.13 (d, 2H, J=8.8 Hz, ArH), 7.78 (d, 2H, J=8.8 Hz, ArH), 9.74 (s, 1H, pyridine), 9.78 (s, 1H, pyridine), 12.00 (s, 1H, OH), 12.12 (s, 1H, OH). ^{13}C NMR (dimethyl sulfoxide d6): 19.1, 107.3, 128.0, 128.7, 129.0, 129.2, 130.1, 130.5, 131.2, 131.6, 153.9, 162.1, 192.3. *Anal*. calcd. for $C_{23}H_{16}BrNO_6$: C, 57.28; H, 3.34; N, 2.90. Found: C, 57.21; H, 3.38; N, 2.85.

3-(4-(4-Chlorophenyl)-6-(4-hydroxy-6-methyl-2-oxo-2H-pyran-3-yl)pyridin-2-yl)-4-hydroxy-6-methyl-2H-pyran-2-one (4d). Color: yellow solid; mp 217–219°C; IR (potassium bromide): 3243 (–OH), 1715 (O-C=O), 1601 (C=N); 1H NMR (dimethyl sulfoxide d6): δ 2.06 (s, 3H, CH_3), 2.15 (s, 3H, CH_3), 5.77 (s, 1H, pyran), 5.80 (s, 1H, pyran), 7.24 (d, 2H, J=8.4 Hz, ArH), 7.61 (d, 2H, J=7.6 Hz, ArH), 9.72 (s, 1H, pyridine), 9.73 (s, 1H, pyridine), 12.11 (s, 1H, OH), 12.20 (s, 1H, OH). *Anal*. calcd. for $C_{23}H_{16}ClNO_6$: C, 63.09; H, 3.68; N, 3.20. Found: C, 63.14; H, 3.61; N, 3.24.

4-Hydroxy-3-(6-(4-hydroxy-6-methyl-2-oxo-2H-pyran-3-yl)-4-p-tolylpyridin-2-yl)-6-methyl-2H-pyran-2-one (4e). Color: yellow solid; mp 243–245°C; IR (potassium bromide): 3246 (–OH), 1715 (O-C=O), 1598 (C=N); 1H NMR (dimethyl sulfoxide d6): δ 2.05 (s, 3H, CH_3), 2.10 (s, 3H, CH_3), 2.24 (s, 3H, CH_3), 5.71 (s, 1H, pyran), 5.79 (s, 1H, pyran), 6.84 (d, 2H, J=8.8 Hz, ArH), 7.30 (d, 2H, J=8.8 Hz), 9.71 (s, 1H, pyridine), 9.78 (s, 1H, pyridine), 11.85 (s, 1H, OH), 12.10 (s, 1H, OH). *Anal*. calcd. for $C_{24}H_{19}NO_6$: C, 69.06; H, 4.59; N, 3.36. Found: C, 68.94; H, 4.52; N, 3.31.

4-Hydroxy-3-(6-(4-hydroxy-6-methyl-2-oxo-2H-pyran-3-yl)-4-(4-methoxyphenyl)pyridin-2-yl)-6-methyl-2H-pyran-2-one (4f). Color: yellow solid; mp >300°C; IR (potassium bromide): 3252 (–OH), 1727 (O-C=O), 1596 (C=N); 1H NMR (dimethyl sulfoxide d6): δ 2.06 (s, 3H, CH_3), 2.10 (s, 3H, CH_3), 3.82 (s, 3H, OCH_3), 5.71 (s, 1H, pyran), 5.79 (s, 1H, pyran), 7.12 (d, 2H, J=8.0 Hz, ArH), 7.61 (d, 2H, J=7.6 Hz, ArH), 9.72 (s, 1H, pyridine), 9.75 (s, 1H, pyridine), 12.06 (s, 1H, OH), 12.12 (s, 1H, OH). *Anal*. calcd. for $C_{24}H_{19}NO_7$: C, 66.51; H, 4.42; N, 3.23. Found: C, 66.55; H, 4.38; N, 3.18.

3-(4-(4-(Dimethylamino)phenyl)-6-(4-hydroxy-6-methyl-2-oxo-2H-pyran-3-yl)pyridin-2-yl)-4-hydroxy-6-methyl-2H-pyran-2-one (4g). Color: yellow solid; mp 264–266°C; IR (potassium bromide): 3243 (–OH), 1731 (O-C=O), 1595 (C=N); 1H NMR (dimethyl sulfoxide d6): δ 2.09 (s, 3H, CH_3), 2.13 (s, 3H, CH_3), 3.04 (s, 6H, CH_3), 5.67 (s, 1H, pyran), 5.78 (s, 1H, pyran), 6.79 (d, 2H, J=8.8 Hz, ArH), 7.68 (d, 2H, J=8.8 Hz, ArH), 9.67 (s, 1H, pyridine), 9.72 (s, 1H, pyridine), 12.07 (s, 1H, OH), 12.15 (s, 1H, OH). ^{13}C NMR (dimethyl sulfoxide d6): 19.1, 42.0, 111.9, 125.9, 126.2, 126.8, 127.0, 127.7, 128.0, 129.6, 130.9, 151.3, 162.1, 183.2. *Anal*. calcd. for $C_{25}H_{22}N_2O_6$: C, 67.26; H, 4.97; N, 6.27. Found: C, 67.21; H, 4.91; N, 6.20.

4-Hydroxy-3-(6-(4-hydroxy-6-methyl-2-oxo-2H-pyran-3-yl)-4-(4-hydroxyphenyl)pyridin-2-yl)-6-methyl-2H-pyran-2-one (4h). Color: yellow solid; mp 231–232°C; ir (potassium bromide): 3235 (–OH), 1717 (O-C=O), 1594 (C=N); 1H NMR (dimethyl sulfoxide d6): δ 2.06 (s, 3H, CH_3), 2.14 (s, 3H, CH_3), 5.77 (s, 1H, pyran), 5.83 (s, 1H, pyran), 7.14 (d, 2H, J=8.8 Hz, ArH), 7.52 (d, 2H, J=8.8 Hz, ArH), 9.58 (s, 1H, pyridine), 9.60 (s, 1H, pyridine), 12.07 (s, 1H, OH), 12.15 (s, 1H, OH), 12.30 (s, 1H, OH). *Anal*. calcd. for $C_{23}H_{17}NO_7$: C, 65.87; H, 4.09; N, 3.34. Found: C, 65.81; H, 4.12; N, 3.28.

4-Hydroxy-3-(6-(4-hydroxy-6-methyl-2-oxo-2H-pyran-3-yl)-4-(2-hydroxyphenyl)pyridin-2-yl)-6-methyl-2H-pyran-2-one (4i). Color: orange yellow solid; mp 225–227°C; IR (potassium bromide): 3212 (–OH), 1721 (O-C=O), 1595 (C=N); 1H NMR (dimethyl sulfoxide d6): δ 1.96 (s, 3H, CH_3), 2.04 (s, 3H, CH_3), 5.67 (s, 1H, pyran), 5.79 (s, 1H, pyran), 7.31 (t, 1H, J=7.6 Hz, ArH), 7.49 (d, 1H, J=8.8 Hz, ArH), 7.61 (d, 1H, J=8.0 Hz, ArH), 9.72 (s, 1H, pyridine), 9.75 (s, 1H, pyridine), 12.10 (s, 1H, OH), 12.21 (s, 1H, OH), 12.27 (s, 1H, OH). *Anal*. calcd. for $C_{23}H_{17}NO_7$: C, 65.87; H, 4.09; N, 3.34. Found: C, 65.81; H, 4.12; N, 3.30.

4-Hydroxy-3-(6-(4-hydroxy-6-methyl-2-oxo-2H-pyran-3-yl)-4-(3-nitrophenyl)pyridin-2-yl)-6-methyl-2H-pyran-2-one (4j). Color: orange yellow solid; mp 287–289°C; IR (potassium bromide): 3224 (–OH), 1713 (O-C = O), 1597 (C = N); 1H NMR (dimethyl sulfoxide d6): δ 2.08 (s, 3H, CH_3), 2.11 (s, 3H, CH_3), 5.73 (s, 1H, pyran), 5.75 (s, 1H, pyran), 7.38 (d, 1H, J = 8.8 Hz, ArH), 7.61–7.67 (m, 3H, ArH), 9.72 (s, 1H, pyridine), 9.73 (s, 1H, pyridine). *Anal*. calcd. for $C_{23}H_{16}N_2O_8$: C, 61.61; H, 3.60; N, 6.25. Found: C, 61.68; H, 3.54; N, 6.21.

4-Hydroxy-3-(6-(4-hydroxy-6-methyl-2-oxo-2H-pyran-3-yl)-4-(2-hydroxynaphthalen-1-yl) pyridine −2-yl)-6-methyl-2H-pyran-2-one (4k). Color: orange yellow solid; mp 234–236°C; IR (potassium bromide): 3246 (–OH), 1715 (O-C = O), 1598 (C = N); 1H NMR (dimethyl sulfoxide d6): δ 1.90 (s, 3H, CH_3), 2.06 (s, 3H, CH_3), 5.74 (s, 1H, pyran), 5.81 (s, 1H, pyran), 7.33–7.37 (m, 2H, ArH), 7.43 (d, 1H, J = 8.0 Hz, ArH), 7.52 (d, 2H, J = 8.8 Hz, ArH), 7.70 (d, 1H, J = 7.6 Hz, ArH), 9.81 (s, 1H, pyridine), 9.83 (s, 1H, pyridine), 12.11 (s, 1H, OH), 12.13 (s, 1H, OH), 12.16 (s, 1H, OH). *Anal*. calcd. for $C_{27}H_{19}NO_7$: C, 69.08; H, 4.08; N, 2.98. Found: C, 69.12; H, 4.12; N, 2.92.

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