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AN EFFICIENT SYNTHESIS OF 2, 4, 6 TRI ARYL PYRIDINES USING AMMONIUM CARBONATE IN WATER UNDER SEALED CONDITIONS

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ABSTRACT

Krohnke 2, 4, 6-Triarylpyridines (TAPs) are efficiently synthesized by using various reactants with ammonium carbonate in water under sealed conditions. Using this protocol, Krohnke pyridines (4a-4q) are prepared in higher yields and purities than with other methodologies without the use of a catalyst or an organic solvent.

KEYWORDS

Sealed conditions, Ammonium carbonate, Water and 2, 4, 6-Triarylpyridines.

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INTRODUCTON

Organic transformations in water without using hazardous reagents or solvents are of considerable interest, because of its environmental acceptability, abundance and low cost[1]. Pyridines derivatives represent an important class of six-membered heterocycles widespread in a number of biologically active natural products[2] and pharmaceutical drugs[3]. They have noticeable applications in many fields of chemistry[4]. In particular 2, 4, 6-triarylpyridine is of immense interest because of its unique position in medicinal chemistry[5], such as topoisomerase I and II inhibitory activity, cytotoxicity[6] against several human cancer cell lines[7] antitumor activity[8]. Recent studies providing impetus for further studies in

utilizing this scaffold in new therapeutic drug classes[9].

In addition, the excellent thermal stabilities of these pyridines have instigated a growing interest for their use as monomeric building blocks useful in the development of thin film vortex fluidic device[10], building blocks for the preparation of chiral ligands[11]. TAPs show promising potential as scintillators that will allow liquid scintillation counting to be carried out at high efficiency in strongly acidic solution and new materials with important photo-or electrochemical properties[12]. Some examples are used as pharmaceuticals, dyes, additives, agrochemicals, and also in qualitative and quantitative analyses[13]. Moreover, they are prominent synthons in supramolecular chemistry, with their π-stacking ability along with directional H-bonding capacity[14]. In addition, the excellent thermal stabilities of these pyridines have gained considerable interest for their use as monomeric building blocks in thin films and organometallic polymers[15].

Traditionally TAPs have been synthesized using the reaction of N-phenacylpyridinium salts with α, β - unsaturated ketones in the presence of NH_4OAc[16]. Recently, several new and improved methods and procedures have been developed for the synthesis of TAP's all of these methods use NH_4OAc as a source of ammonia which include arylation of methylthiopyridines via Ni-induced Grignard reactions reactions of phenacylidene dimethylsulfurane with chalcones and NH_4OAc,[17] pyrolysis of 1-vinyl-1, 2-dihydropyridines[18], reactions of a-ketoketene dithioacetals with methyl ketones in the presence of NH_4OAc[19], additions of lithiated b-enaminophosphonates to chalcones[20], reactions of a-benzotriazolyl ketones with a, b-unsaturated ketones and NH_4OAc[21], and solvent-free reactions[22a,b] between acetophenones, benzaldehydes, and NH_4OAc in the presence of sodium hydroxide[22], or without a catalyst under microwave irradiation[23], Ultrasound-mediated[24], However, most of these syntheses of TAPs are multistep, low to moderate yielding processes.

Among all these methods, even the well-established protocol also uses NH_4OAc (one pot reaction between acetophenones, aryl aldehydes, and NH_4OAc) for the synthesis of tri-aryl pyridines using NaOH in PEG-400[25], There have been plethora of catalysts used for this reactions such as PEG-300 along with NaOH[26], catalytic amount of acetic acid[27], $HClO_4$- SiO_2[28], preyssler type hetero poly acid $H_{14}[NaP_5W_30O_{11}0]$[29], wet 2, 4, 6-trichloro-1, 3, 5-triazine (TCT)[30], 3-methyl-1-(4-sulfonylbutyl) imidazolium hydrogen sulfate $[HO_3S(CH_2)_4MIM]$ $[HSO_4]$ and a Bronsted acidic ionic liquid[31], Bismuth triflate[32], But, most of these protocols are having one or more drawbacks, thus leaving room for further improvements.

EXPERIMENTAL

General procedure for the preparation of 2, 4, 6-triarylpyridines

A mixture of the acetophenone (2.1mmol), aromatic aldehyde (1.2mmol) and anhydrous ammonium carbonate (2mmol) in water was heated in a sealed tube at 150°C for 4 h. The reaction was monitored by TLC (Thin layer chromatography) n-hexane-EtOAc (6:4). After completion of the reaction, reaction mixture was cooled to room temperature and the residue was eluted by using n-hexane-EtOAc (5:1) through column chromatography. The residue was recrystallized from absolute EtOH.

RESULTS AND DISCUSSION

We were interested in studying synthesis of 2, 4, 6 tri aryl pyridines using ammonium carbonate in aqueous media using ammonium carbonate with the aim to develop an operationally simple method for the synthesis of a large range of TAPs.

Ammonium carbonate is a low melting (58°C) and less toxic (LD50 = 1497mg/kg) solid. In aqueous media it decomposes to produce two moles of ammonia. Under solvent-free conditions the reaction proceeded in a considerably lower yield due to sublimation of ammonium carbonate. There was no significant change on the results observed using high equiv (0.5-1) of ammonium carbonate, suggests that hydrogen bonding, mild buffered pH of the reaction media and the assistance of water to break down $(NH_4)_2CO_3$ may all be responsible for acceleration of the reaction rate.

Although the preparation of 2, 4, 6 tri aryl pyridines has been known there is no report on the effect of the ammonia source on this reaction (Scheme No.1). Thus, we studied a model four-component condensation of acetophenone, benzaldehyde and an ammonium salt (mole ratio = 2:1:1) in water under different conditions (Table No.1). We were pleased to find that among the conditions screened, the corresponding TAPs was obtained quantitatively with $(NH_4)_2CO_3$ at 140-150ºC in water (entry 10) in the absence of any catalyst. This process is economically viable than the previously reported procedures.

[a]Isolated yields

The optimized conditions required heating with 35 mol % of ammonium carbonate in water for four hours at 140-150ºC under the sealed conditions. In order to study the scope and generality of the ammonium carbonate-catalyzed 2, 4, 6 tri aryl pyridines synthesis in water, a series of TAPs were synthesized from the substituted aromatic aldehydes, and aromatic ketones (Scheme No.2). In all cases, the desired products were isolated in excellent yields (Table No.2).

The optimized reaction conditions further extended to the condensation of other aldehydes with aromatic ketone (Scheme No.2, 4a-4q), chalchone with aromatic ketone (Scheme No.3), chalchone and ammonium carbonate (Scheme No.4), at 80-150ºC. Aromatic aldehydes bearing both electron-deficient and electron-rich substituent have afforded the desired TAPs in excellent yields.

SPECTRAL DATA

2, 4, 6-Triphenylpyridine (4a)

White solid, M.P. 135-137ºC, IR (KBr, cm^{-1}): 3069, 1597, 1552, 1494, 1440, 1398, 1178, 1074, 1027, 867, 759, 692. ^{1}H NMR (300 MHz, DMSO-d_6): δ (ppm): 8.21(2H, d, *J* = 7.2 Hz, H Ar); 7.93(2H, s, H Ar); 7.79(2H, d, *J* = 7.2 Hz, H Ar); 7.53(2H, d, *J* = 7.4 Hz, H Ar); 7.40-7.34 (9H, m, H Ar). ^{13}C NMR (75 MHz, $CDCl_3$): δ (ppm): 157.2; 150.0; 139.8; 139.2; 129.8; 129.3; 128.9; 127.7; 127.2; 117.8. HRMS $[M+H]^+$: 308.1214; Found, %: C 89.78; H 5.51; N 4.50. $C_{23}H_{17}N$. Calculated, %: C 89.87; H 5.57; N 4.56.

4-(4-Chlorophenyl)-2, 6-diphenylpyridine (4b)

White solid, M.P. 127-128ºC, IR (KBr, cm^{-1}): 3061, 1599, 1543, 1489, 1449, 1414, 1384, 1237, 1090, 1013, 825, 773, 692. ^{1}H NMR (300 MHz, DMSO-d_6): δ (ppm) 8.59 (2H, d, *J* = 7.2 Hz, H Ar); 8.53 (2H, d, *J* = 7.8 Hz, H Ar); 8.14 (2H, s, H Ar); 7.84 (2H, d, *J* = 7.8 Hz, H Ar); 7.66 (2H, d, *J* = 7.8 Hz, H Ar); 7.56-7.52 (6H, m, H Ar). ^{13}C NMR (75 MHz, DMSO-d_6): δ (ppm):157.8; 149.0; 139.0; 136.1; 134.6; 129.8; 129.6; 129.5; 129.0; 117.0. HRMS $[M+H]^+$: 342.4899, Found, %: C 80.32; H 4.55; N 4.01. $C_{23}H_{16}ClN$. Calculated, %: C 80.81; H 4.72; N 4.10.

4-(2-Fluorophenyl)-2, 6-diphenylpyridine (4c)

White solid, M.P. 118-119ºC, IR (KBr, cm^{-1}): 3031, 1591, 1544, 1490, 1451, 1395, 1288, 1209, 1204, 1114, 1074, 1025, 878, 759, 694. ^{1}H NMR (300 MHz, DMSO-d_6), δ (ppm):8.13 (4H, d, *J* = 7.5 Hz, H Ar); 7.94 (4H, d, *J* = 7.5 Hz, H Ar); 7.34-7.33 (4H, s, H Ar); 7.09-7.04 (4H, d, *J* = 7.9 Hz, H Ar). ^{13}C NMR (75 MHz, DMSO-d_6): δ(ppm): 160.0; 157.4; 149.2; 139.3; 130.9; 129.2; 127.9; 126.9; 125.2; 124.0; 118.1; 116.4; HRMS $[M+H]^+$: 338.3823, Found, %: C 84.53; H 4.75; N 4.23. $C_{23}H_{16}FN$. Calculated, %: C 84.90; H 4.96; N 4.30.

4-(3-Methoxyphenyl)-2, 6-diphenylpyridine (4d)

White solid, M.P. 122-124ºC, IR (KBr, cm^{-1}): 3034, 2936, 1596, 1547, 1486, 1444, 1398, 1285, 1255, 1204, 1171, 1037, 872, 775, 692. ^{1}H NMR (300 MHz, DMSO-d_6), δ (ppm): 8.15 (2H, d, *J* = 7.8 Hz, H Ar); 8.08 (2H, d, *J* = 7.6 Hz, H Ar); 8.01 (2H, s, H Ar); 7.45-7.38 (8H, m, H Ar); 7.32 (1H, d, *J* = 7.4 Hz, H Ar); 6.78 (1H, t, *J* = 7.4 Hz, H Ar); 3.65 (3H, s, OCH_3). ^{13}C NMR (75 MHz, DMSO-d_6): δ (ppm): 162.3; 158.3; 152.3; 138.8; 135.2; 132.2; 128.5; 129.2; 128.1; 119.0; 116.9; 114.8; 113.2; 54.9. HRMS $[M+H]^+$: 338.3823, Found, %: C 85.34; H 5.55; N 4.10. $C_{24}H_{19}NO$. Calculated, %: C 85.43; H 5.68; N 4.15.

4-(4-Methylphenyl)-2, 6-diphenylpyridine (4e)

White solid, M.P. 122-123ºC, IR (KBr, cm^{-1}): 3034, 2936, 1598, 1548, 1442, 1398, 1286, 1254, 1203, 1170, 1036, 871, 775, 691. ^{1}H NMR (300 MHz, DMSO-d_6), δ (ppm): 7.93 (4H, d, *J* = 7.2 Hz, H Ar); 7.43 (4H, d, *J* = 7.2 Hz, H Ar); 7.36 (2H, d, *J* = 6.0 Hz, H Ar), 7.32-7.27 (4H, m, H Ar); 7.13 (2H, d, *J*

= 6.3 Hz, H Ar); 2.35 (3H, s, CH_3). ^{13}C NMR (75 MHz DMSO-d_6): δ (ppm):157.0; 149.9; 139.8; 135.4; 130.3; 129.8; 129.3; 127.5; 127.1; 116.4; 21.5. HRMS $[M+H]^+$: 322.1726, Found, %: C 89.52; H 5.78; N 4.28. $C_{24}H_{19}N$. Calculated, %: C 89.68; H 5.96; N 4.36.

4-(4-Methoxyphenyl)-2, 6-diphenylpyridine (4f)

White solid, M.P. 98-100ºC, IR (KBr, cm^{-1}): 3035, 2936, 1596, 1547, 1486, 1444, 1398, 1285, 1255, 1204, 1171, 1037, 750, 691. ^{1}H NMR (300 MHz, DMSO-d_6), δ (ppm): 8.01 (4H, d, *J* = 6.9 Hz, H Ar); 7.87 (2H, d, *J* = 7.2 Hz, H Ar); 7.35 (4H, d, *J* = 6.9 Hz, H Ar); 7.319-7.286 (2H, s, H Ar); 6.88 (4H, d, *J* = 7.2 Hz, H Ar); 3.76 (3H, s, OCH_3). ^{13}C NMR (75 MHz, DMSO-d_6): δ, ppm: 160.4; 157.1; 150.7; 139.9; 139.4; 130.3; 129.8; 129.3; 127.1; 120.9; 116.9; 115.4; 113.4; 53.5. HRMS $[M+H]^+$: 338.1501, Found, %: C 85.12; H 5.24; N 4.02. $C_{24}H_{19}NO$. Calculated, %: C 85.43; H 5.68; N 4.15.

***N*, *N*-Dimethyl-4-(2, 6-diphenylpyridin-4-yl) benzenamine (4g)**

Yellow solid, M.P. 137-139ºC, IR (KBr, cm^{-1}): 3037, 2936, 1598, 1525, 1489, 1442, 1398, 1352, 1233, 1199, 1168, 1066, 1023, 818, 773, 695. ^{1}H NMR (300 MHz, DMSO-d_6), δ (ppm): 8.02 (2H, s, H Ar); 7.77 (4H, d, *J* = 7.2 Hz, H Ar); 7.52 (2H, d, *J* = 7.2 Hz, H Ar); 7.22-7.11(6H, m, H Ar); 6.8 (2H, d, *J* = 7.2 Hz, H Ar); 2.95 (6H, s, $N(CH_3)_2$). ^{13}C NMR (75 MHz, DMSO-d_6): δ (ppm): 155.4; 152.2; 150.5; 136.2; 129.9; 129.0; 128.7; 127.0; 118.8; 114.4; 42.2. HRMS $[M+H]^+$: 338.1501, Found, %: C 85.24; H 6.21; N 7.87. $C_{25}H_{22}N_2$. Calculated, %: C 85.68; H 6.33; N 7.99.

2, 6-Bis (4-chlorophenyl)-4-phenylpyridine (4h)

White solid, M.P. 177-178ºC, IR (KBr, cm^{-1}): 3052, 1598, 1544, 1490, 1449, 1413, 1384, 1239, 1174, 1091, 1012, 829, 761, 694. ^{1}H NMR (300 MHz, DMSO-d_6), δ (ppm): 8.22 (4H, d, *J* = 8.1 Hz, H Ar); 7.79 (2H, s, H Ar); 7.64 (4H, d, *J* = 8.1 Hz, H Ar); 7.32-7.26 (5H, m, H Ar). ^{13}C NMR (75 MHz, DMSO-d_6) δ (ppm): 158.0; 150.3; 138.2; 129.7; 129.3; 128.6; 125.3; 117.9. HRMS $[M+H]^+$: 338.1501, Found, %: C 73.12; H 3.98; N 3.63. $C_{23}H_{15}Cl_2N$. Calculated, %: C 73.42; H 4.02; N 3.72.

4-(4-pyridinel)-2, 6-diphenylpyridine (4i)

Colorless crystals, M.P. 187-188ºC, IR (KBr, cm^{-1}): 3050, 1562, 1544, 1450, 1413, 1384, 1239, 1174, 1078, 1015, 829, 678. ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm): 8.76 (2H, d, *J* = 4.7 Hz, 2CH), 8.18 (4H, d, *J* = 7.5 Hz, 4CH), 7.84 (2H, s, 2CH), 7.61 (2H, d, *J* = 7.4 Hz, 2CH), 7.51 (4H, d, *J* = 7.4 Hz, 4CH), 7.45 (2H, d, *J* = 7.4 Hz, 2CH),. ^{13}C NMR (75 MHz, $CDCl_3$): 157.91; 150.52; 147.31; 146.49; 139.00; 129.37; 128.78; 127.09; 121.65; 116.58. HRMS $[M+H]^+$: 309.5263, Found, %: C 85.23; H 5.09; N 8.98. $C_{22}H_{16}N_2$. Calculated, %: C 85.69; H 5.23; N 9.08.

4-(Furan-2-yl)-2, 6-diphenylpyridine (4j)

Light-brown solid, M.P. 167-169ºC, IR (KBr, cm^{-1}): 3058, 1606, 1541, 1487, 1454, 1414, 1244, 1158, 1073, 1010, 868, 772, 690. ^{1}H NMR (300 MHz, DMSO-d_6), δ (ppm): 8.30 (2H, d, *J* = 7.6 Hz, H Ar); 8.20 (2H, d, *J* = 7.5 Hz, H Ar); 8.14 (2H, s, H Ar); 7.96 (1H, s, H Ar); 7.57-7.47 (7H, m, H Ar); 6.75 (1H, d, *J* = 8.1 Hz, H Ar). ^{13}C NMR (75 MHz, DMSO-d_6): δ (ppm): 157.0; 151.4; 145.2; 139.6; 130.0; 129.8; 129.2; 127.2; 113.1; 113.0; 110.9. HRMS $[M+H]^+$: 298.4825, Found, %: C 84.65; H 4.98; N 4.36. $C_{21}H_{15}NO$. Calculated, %: C 84.82; H 5.08; N 4.71.

2, 6-Bis (4-Methylphenyl)-4-phenylpyridine (4k)

White solid, M.P. 158-159ºC, IR (KBr, cm^{-1}): 3052, 2928, 1602, 1543, 1512, 1489, 1426, 1381, 1291, 1247, 1177, 1088, 1011, 824. ^{1}H NMR (300 MHz, DMSO-d_6), δ (ppm): 7.97 (4H, d, *J* = 6.3 Hz, H Ar); 7.86 (2H, s, H Ar); 7.22-7.42 (5H, m, H Ar); 7.14 (4H, d, *J* = 6.3 Hz, H Ar); 2.23 (6H, s, CH_3). ^{13}C NMR (75 MHz, DMSO-d_6): δ (ppm): 158.1; 150.7; 138.2; 137.2; 132.3; 129.8; 129.3; 128.5; 126.9; 117.6; 19.15. HRMS $[M+H]^+$: 336.1666, Found, %: C 89.28; H 6.17; N 4.02. $C_{25}H_{21}N$. Calculated, %: C 89.51; H 6.31; N 4.18.

4-(4-Nitrophenyl)-2, 6-diphenylpyridine (4l)

Colorless crystals, M.P. 198-199ºC, IR ((KBr, cm^{-1}): 3052, 2926, 1602, 1543, 1512, 1489, 1426, 1380, 1290, 1245, 1175, 1088, 1011, 824. ^{11}H NMR (300 MHz, $CDCl_3$): d = 7.39 (t, *J* = 7.2 Hz, 2H, 2CH), 7.45 (t, *J* = 7.5 Hz, 4H, 4CH), 7.78 (s, 2H, 2CH), 7.79 (d, *J* = 8.6 Hz, 2H, 2CH), 8.11 (d, *J* = 7.3 Hz, 4H, 4CH), 8.29 (d, *J* = 8.6 Hz, 2H, 2CH). ^{13}C NMR

(125.8 MHz, $CDCl_3$): d = 116.9, 124.3, 127.1, 128.1, 128.8, 129.4 (6CH), 139.0, 145.4, 147.8, 148.2. HRMS $[M+H]^+$: 353.3627, Found, %: C 78.05; H 4.36; N 7.83. $C_{23}H_{16}N_2O_2$. Calculated, %: C 78.39; H 4.58; N 7.95.

2, 6-Bis (4-4-methylphenyl)-4-(4-chlorophenyl) pyridine (4m)

White solid, M.P. 198-200ºC, IR (KBr, cm^{-1}): 3062, 2932, 1595, 1546, 1490, 1460, 1411, 1383, 1265, 1211, 1176, 1089, 1012, 833, 787. ^1H NMR (300 MHz, DMSO-d_6), δ (ppm): 8.40 (2H, d, *J* = 7.5 Hz, H Ar); 8.31 (2H, d, *J* = 7.4 Hz, H Ar); 8.20 (2H, s, H Ar); 7.62 (2H, d, *J* = 7.6 Hz, H Ar); 7.55-7.52 (4H, m, H Ar); 7.42 (1H, d, *J* = 7.3 Hz, H Ar); 7.05 (1H, d, *J* = 7.4, H Ar); 2.88 (6H, s, CH_3). ^{13}C NMR (75 MHz, DMSO-d_6): δ, (ppm): 161.2; 155.2; 152.0; 138.9; 134.3; 132.9; 130.3; 129.3; 129.0; 119.7; 118.0; 114.8; 111.1; 15.8. $[M+H]^+$: 390.2140, Found, %: C 81.02; H 5.39; N 3.69. $C_{25}H_{20}ClN$. Calculated, %: C 81.18; H 5.45; N 3.79.

4-(4-Nitrophenyl)-2, 6-bis (4-methylphenyl) pyridine (4n)

Colorless crystals, M.P. 143-144ºC, IR (KBr, cm^{-1}): 3062, 2932, 1595, 1545, 1490, 1460, 1411, 1380, 1265, 1210, 1175, 1085, 1011, 830, 785; ^1H NMR (300 MHz, DMSO-d_6): 8.31 (d, *J* = 8.4 Hz, 2H), 7.88 (d, *J* = 7.2 Hz, 4H), 7.76 (2H, s, H Ar), 7.61(d, *J* = 8.4 Hz, 2H), 6.92 (d, *J* = 7.2 Hz, 4H), 2.86 (6H, s, CH_3). ^{13}C NMR (125.8 MHz, DMSO-d_6): 150.2, 146.3, 139.2,129.2, 129.0, 128.1,127.4, 124.1,19.2 HRMS $[M+H]^+$: 381.6270, Found, %: C 78.78; H 5.17; N 7.22 $C_{23}H_{16}N_2O_2$. Calculated, %: C 78.93; H 5.30; N 7.36.

2, 6-Bis (4-methylphenyl)-4-(4-methoxylphenyl) pyridine (4o)

White solid, M.P. 153-154ºC, IR (KBr, cm^{-1}): 3062, 2932, 1595, 1546, 1490, 1460, 1411, 1383, 1265, 1211, 1176, 1089, 1012, 833, 787. ^1H NMR (300 MHz, DMSO-d_6), δ (ppm): 7.98 (4H, d, *J* = 7.5, H Ar); 7.81(2H, s, H Ar); 7.62 (4H, d, *J* = 7.5, H Ar); 7.36 (2H, m, H Ar); 6.93 (2H, d, *J* = 7.4, H Ar); 3.58 (3H, s, OCH_3) 2.88 (6H, s, CH_3). ^{13}C NMR (75 MHz, DMSO-d_6): δ, ppm: 161.8; 157.2; 149.6; 136.9; 136.3; 129.1; 129.7; 127.0; 119.7; 118.0; 116.8; 113.1; 52.3,19.8. $[M+H]^+$: 366.2893, Found, %: C 85.20; H 6.23; N 3.59. $C_{26}H_{23}NO$. Calculated, %: C 85.45; H 6.34; N 3.83.

4, 4', 4''-(pyridine-2, 4, 6-triyl) triphenol (4p)

Yellow solid, M.P. 283-284ºC, IR (KBr, cm^{-1}): 3294, 1708, 1603, 1513, 1393, 1234, 1175, 831; ^1H NMR (300MHz, DMSO-d_6): δ (ppm) 9.82 (s, 1H, OH), 9.73 (s, 2H, OH), 8.12(d, *J* = 8.7 Hz, 4H), 7.87 (s, 2H, H Ar), 7.44 (d, *J* = 8.4 Hz, 2H), 6.90–6.84 (m, 6H); ^{13}C NMR (75MHz, DMSO-d_6): δ (ppm) 158.2, 158.1, 156.4, 148.9, 130.1, 128.9, 128.6, 128.0, 115.9, 115.4, 113.5; $[M+H]^+$: 356.1196, Found, %: C 77.35; H 4.55; N 3.68. $C_{23}H_{17}NO_3$. Calculated, %: C 77.73; H 4.82; N 3.94.

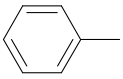
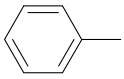
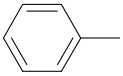
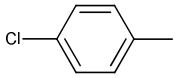
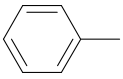
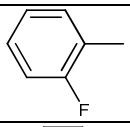
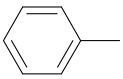
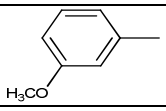
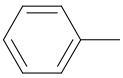
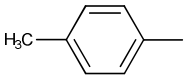
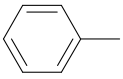
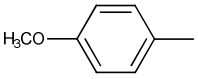
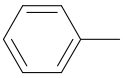
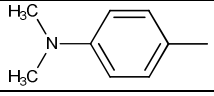
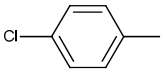
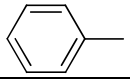
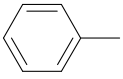
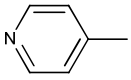
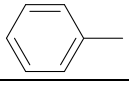
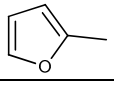
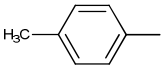
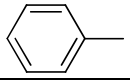
4-(2, 6-diphenylpyridin-4-yl) phenol (4q)

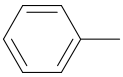
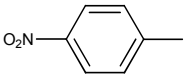
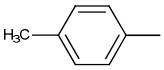
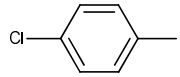
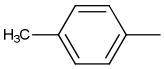
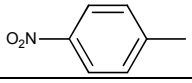
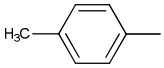
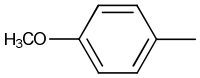
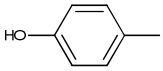
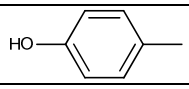
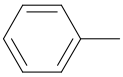
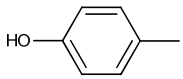
Yellow solid, M.P. 206-208ºC, IR (KBr, cm^{-1}): 3426, 2358, 1560, 1512, 1393, 835, 685. ^1H NMR (300MHz, DMSO-d_6): δ (ppm): 9.87 (s, 1H, OH), 8.31 (d, *J* = 6.9 Hz, 4H), 8.12 (s, 2H, H Ar), 7.86 (d, *J* = 7.2 Hz, 2H), 7.57-7.42 (m, 6H), 6.92 (d, *J* = 6.9 Hz, 2H); ^{13}C NMR (75MHz, DMSO-d_6): δ (ppm) 158.2, 157.3, 150.2, 139.8, 130.4, 129.4, 128.0, 128.4, 127.9, 117.3, 116.2; $[M+H]^+$: 324.0928, Found, %: C 85.23; H 5.15.5; N 4.19. $C_{23}H_{17}NO$. Calculated, %: C 85.42; H 5.30; N 4.33.

Table No.1: Catalyst-free synthesis of 2, 4, 6 Tri aryl pyridines with various ammonium salts in water under sealed conditions

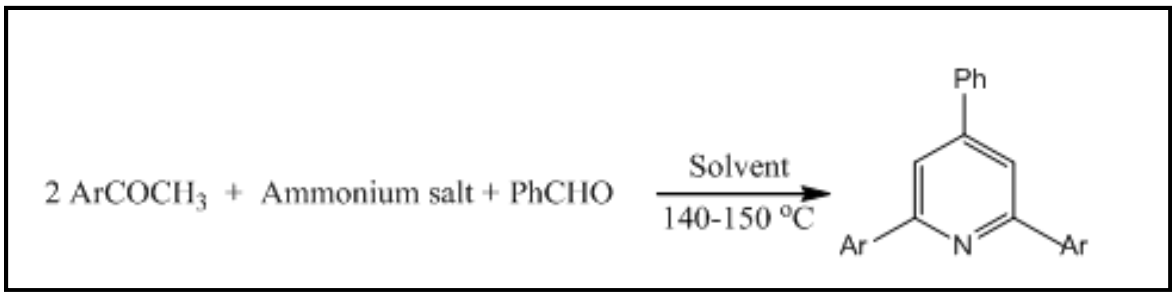
Entry	Ammonium salt	Solvent	Time(h)	Yield[a] (%)
1	NH_4OAc	H_2O	5	85
2	NH_4Cl	H_2O	5	75
3	NH_2CONH_2	H_2O	5	60
4	$(NH4)_2SO_4$	H_2O	5	55
5	NH_4NO_3	H_2O	5	72
6	NH_4VO_3	H_2O	5	74
7	NH_4HCO_3	H_2O	5	68
8	$(NH_4)_6Mo_7O_{24}$	H_2O	5	55
9	$(NH_4)_2[Ce(NO_3)_6]$	H_2O	6	56
10	$(NH_4)_2CO_3$	H_2O	4	95
11	$(NH_4)_2CO_3$	EtOH	6	85
12	$(NH_4)_2CO_3$	H_2O/EtOH (50:50)	5	88
13	$(NH_4)_2CO_3$	None	6	62

Table No.2: Synthesis of 2, 4, 6 Tri aryl pyridines under sealed conditions with ammonium carbonate as source of ammonia

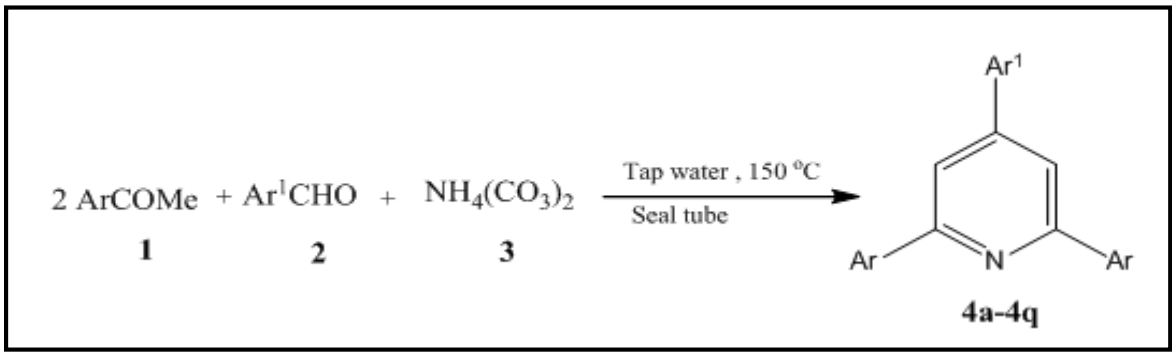
Entry	Ar	Ar[1]	Compound	Yield[a]%
1			4a	97
2			4b	98
3			4c	93
4			4d	95
5			4e	96
6			4f	96
7			4g	95
8			4h	94
9			4i	96
10			4j	92
11			4k	91

12			4l	97
13			4m	98
14			4n	94
15			4o	90
16			4p	98
17			4q	98

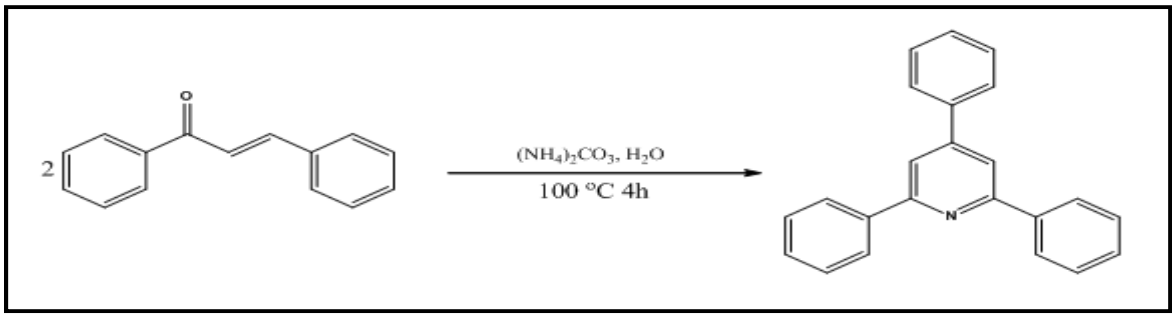
[a] Isolate yields



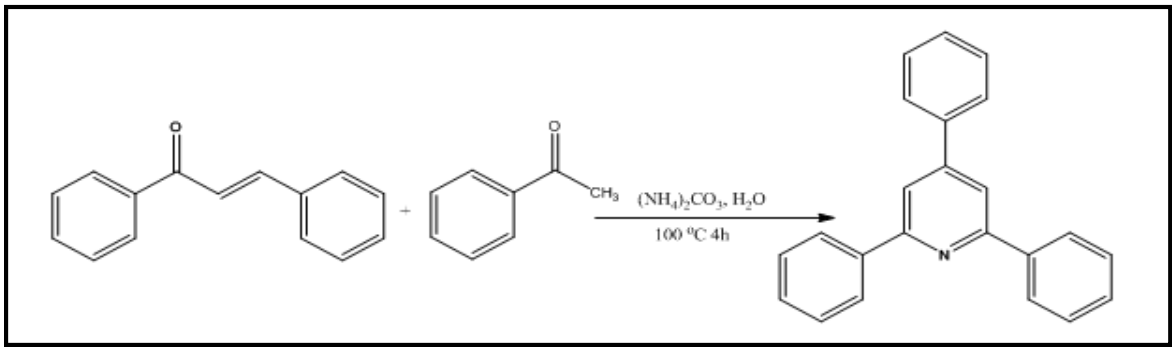
Scheme No.1: Catalyst-free synthesis 2, 4, 6 Triaryl pyridines with various ammonium salts in water under



Scheme No.2: Synthesis of 2, 4, 6 Tri aryl pyridines under sealed conditions with ammonium carbonate as source of ammonia



Scheme No.3: Two component 2, 4, 6 Tri aryl pyridines from chalcone and ammonium carbonate under sealed conditions



Scheme No.4: Three component 2, 4, 6 Tri aryl pyridines from chalcone acetophenone and with ammonium carbonate under sealed conditions

SPCTRAS

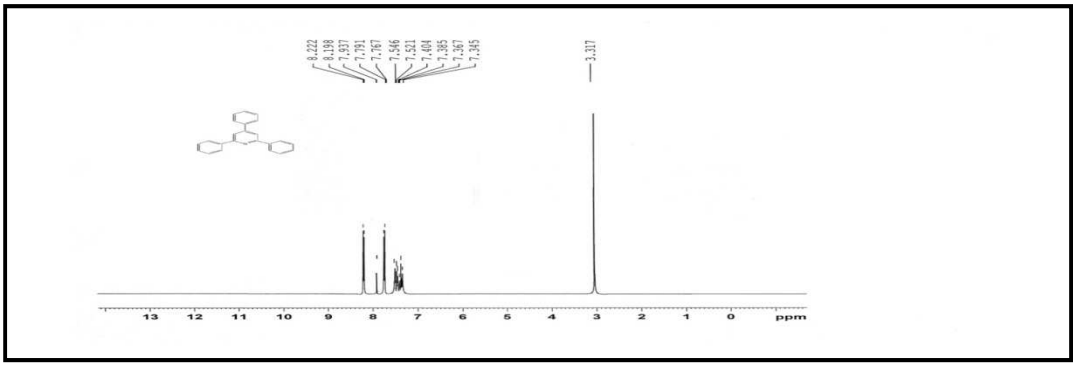


Figure No.1: ^{1}H NMR Spectra of 2, 4, 6-Triphenylpyridine (4a)

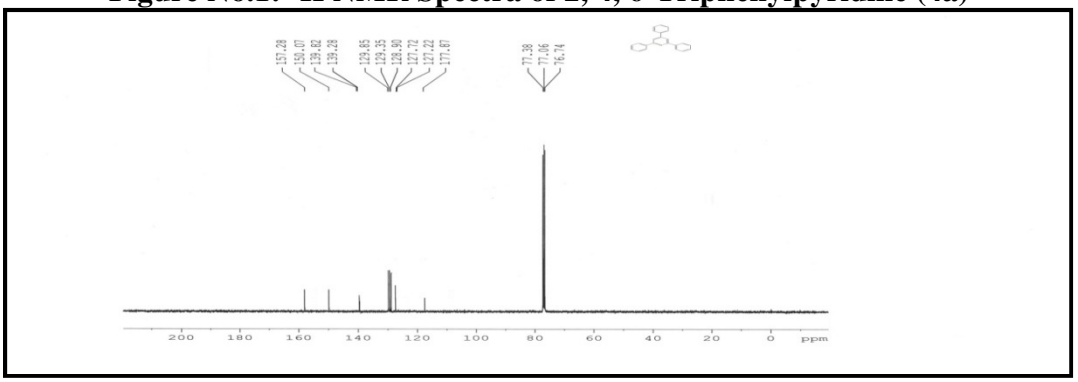


Figure No.2: ^{13}C NMR Spectra of 2, 4, 6-Triphenylpyridine (4a)

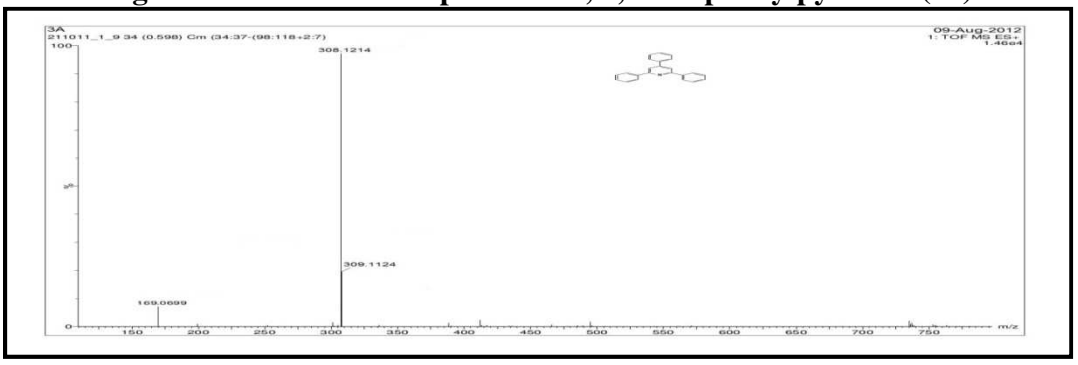


Figure No.3: HRMS Spectra of 2, 4, 6-Triphenylpyridine (4a)

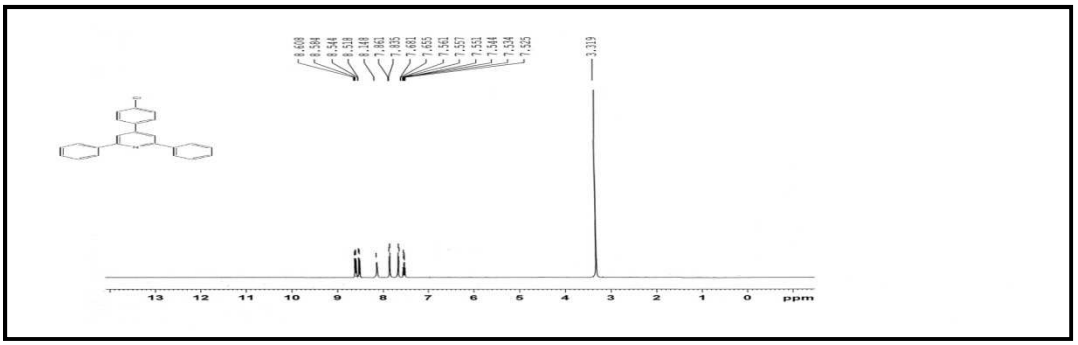


Figure No.4: ^{1}H NMR Spectra of 4-(4-Chlorophenyl)-2, 6-diphenylpyridine (4b)

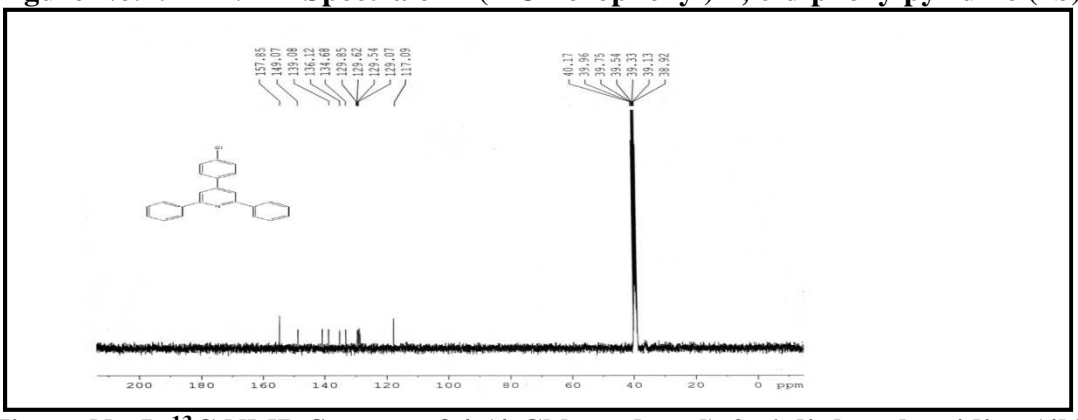


Figure No.5: ^{13}C NMR Spectra of 4-(4-Chlorophenyl)-2, 6-diphenylpyridine (4b)

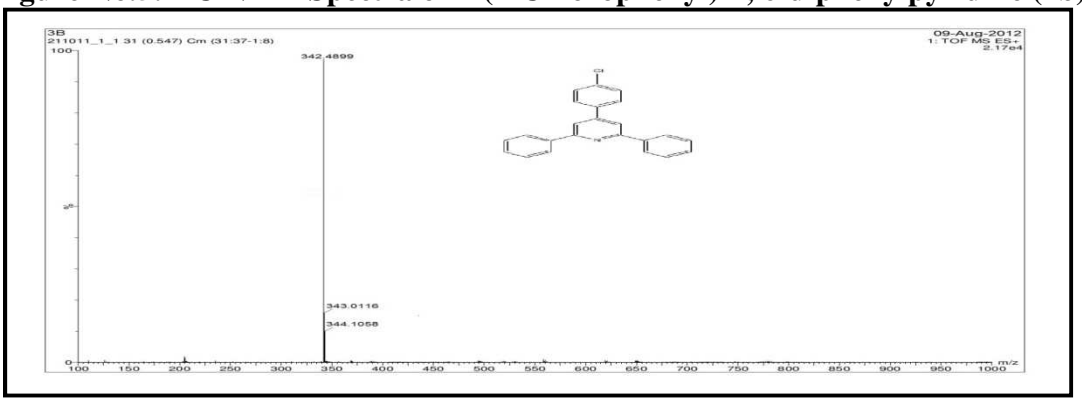


Figure No.6: HRMS Spectra of 4-(4-Chlorophenyl)-2, 6-diphenylpyridine (4b)

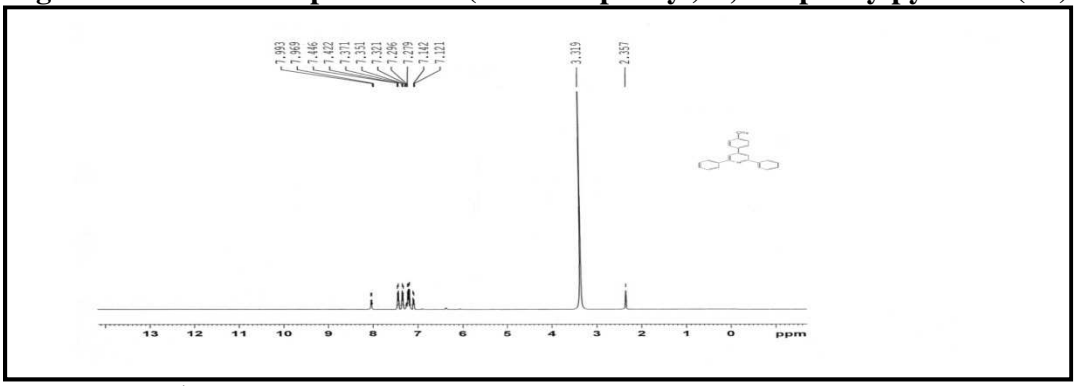


Figure No.7: ^{1}H NMR Spectra of 4-(4-Methylphenyl)-2, 6-diphenylpyridine (4e)

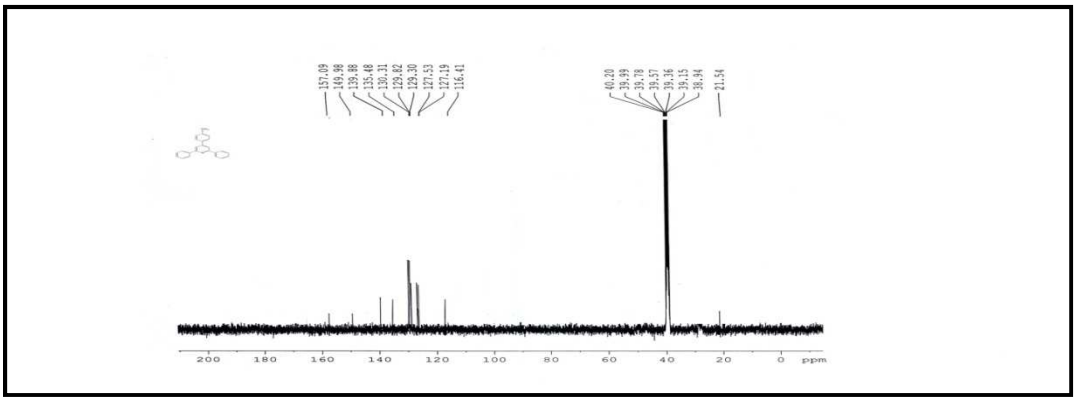


Figure No.8: ^{13}C NMR Spectra of 4-(4-Methylphenyl)-2, 6-diphenylpyridine (4e)

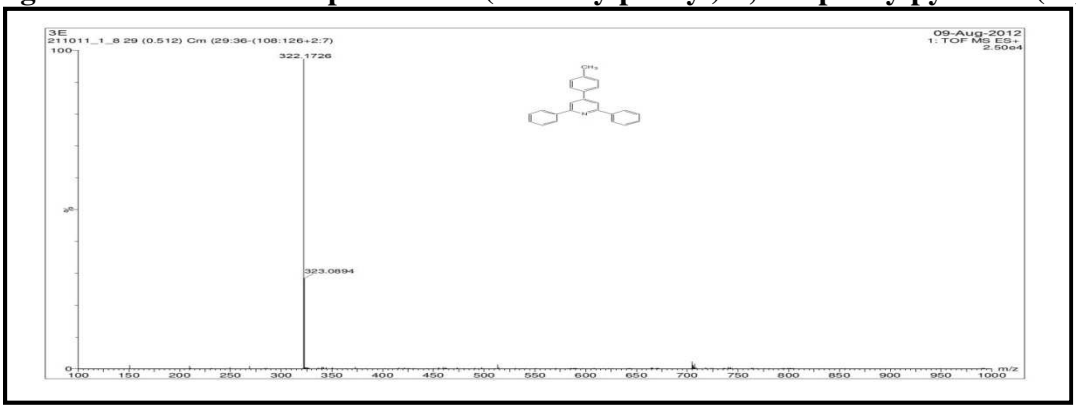


Figure No.9: HRMS Spectra of 4-(4-Methylphenyl)-2, 6-diphenylpyridine (4e)

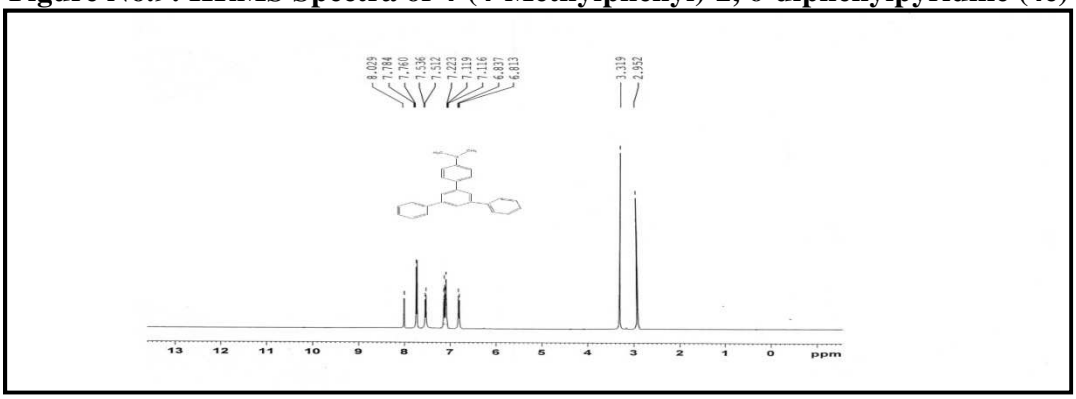


Figure No.10: ^{1}H NMR Spectra of *N*, *N*-Dimethyl-4-(2, 6-diphenylpyridin-4-yl) benzenamine (4g)

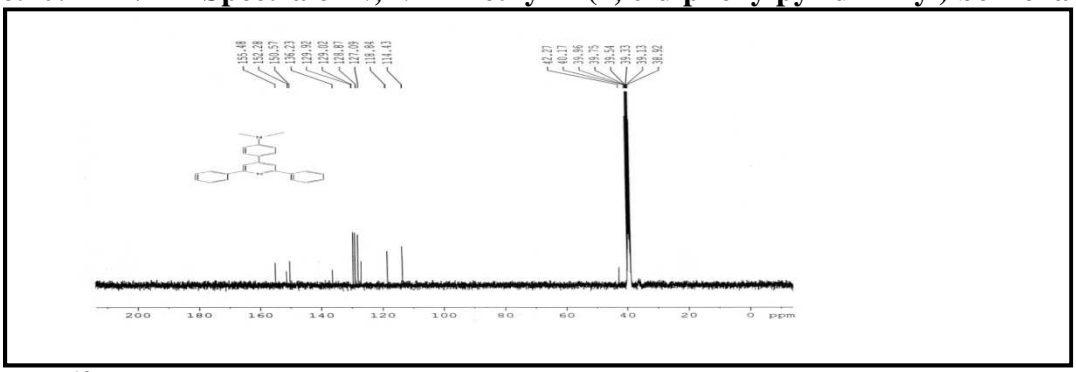


Figure No.11: ^{13}C NMR Spectra of *N*, *N*-Dimethyl-4-(2, 6-diphenylpyridin-4-yl) benzenamine (4g)

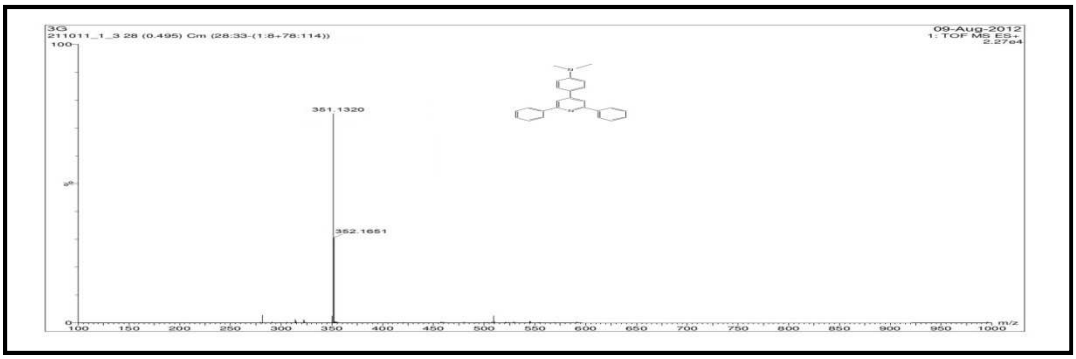


Figure No.12: HRMS Spectra of *N*, *N*-Dimethyl-4-(2, 6-diphenylpyridin-4-yl)benzenamine (4g)

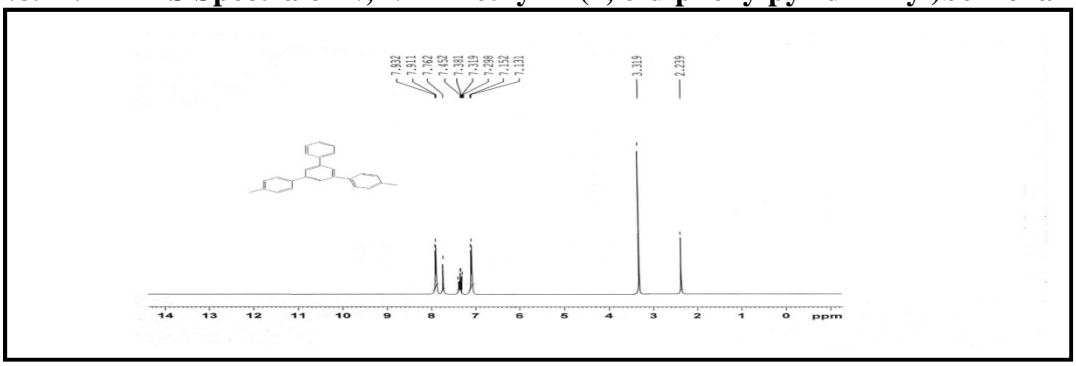


Figure No.13: ^{1}H NMR Spectra of 2, 6-Bis (4-Methylphenyl)-4-phenylpyridine (4k)

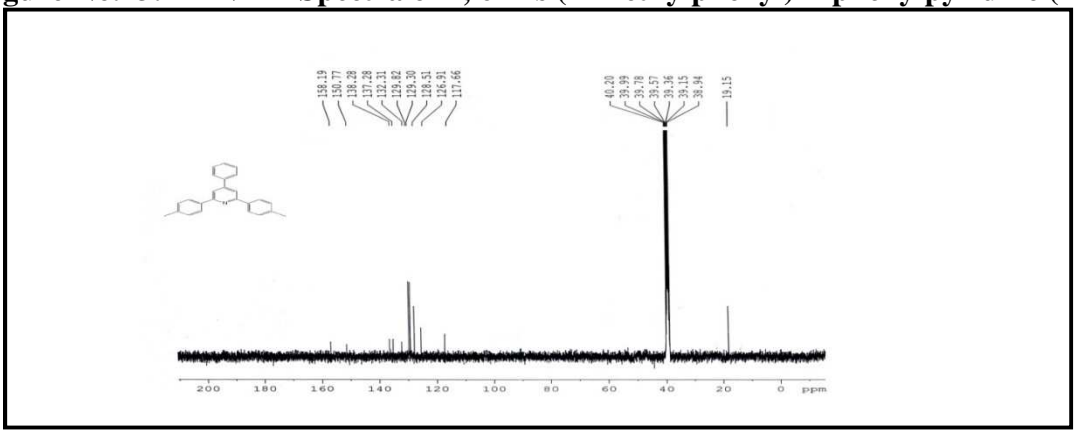


Figure No.14: ^{13}C NMR Spectra of 2, 6-Bis (4-Methylphenyl)-4-phenylpyridine (4k)

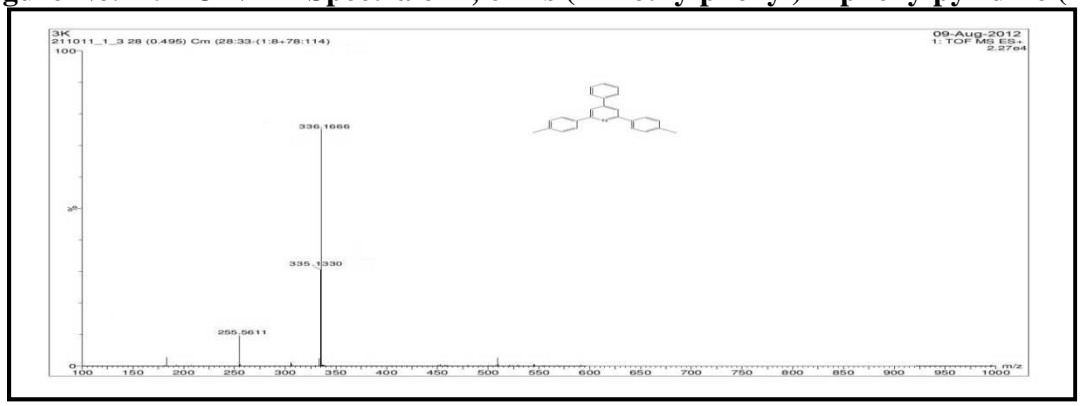


Figure No.15: HRMS Spectra of 2, 6-Bis (4-Methylphenyl)-4-phenylpyridine (4k)

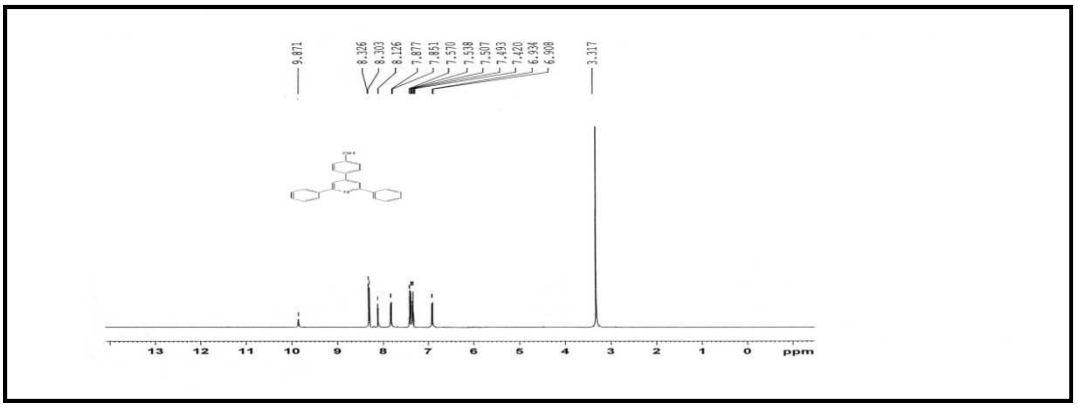


Figure No.16: ^{1}H NMR Spectra of 4-(2, 6-diphenylpyridin-4-yl) phenol (4q)

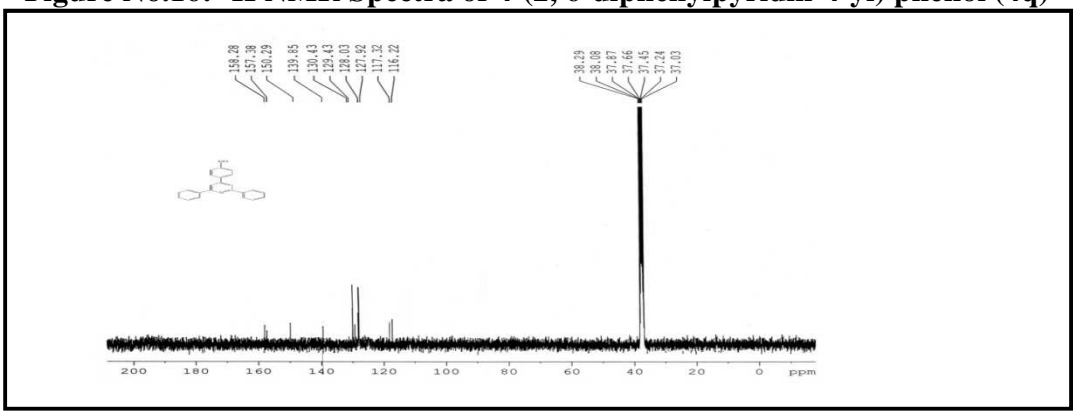


Figure No.17: ^{13}C NMR Spectra of 4-(2, 6-diphenylpyridin-4-yl) phenol (4q)

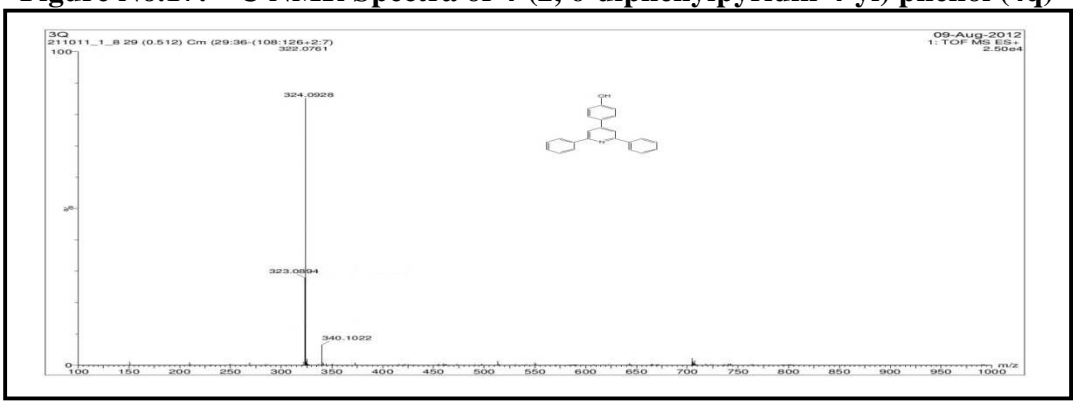


Figure No.18: HRMS Spectra of 4-(2, 6-diphenylpyridin-4-yl) phenol (4q)

CONCLUSION

We have developed an efficient and facile method for the synthesis of 2,4,6 tri arylpyridines. Ammonium carbonate as a source of ammonia, water media, use of simple and readily available starting materials, excellent yields short reactions times are the main advantages of this reaction.

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CONFLICT OF INTEREST

We declare that we have no conflict of interest.

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