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REGULAR ARTICLE

Cerium(IV) carboxymethylcellulose (CMC−Ce^{IV}) as an efficient and reusable catalyst for the one-pot pseudo-four component synthesis of 2,4,6-triphenylpyridines

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Abstract. Cerium(IV) carboxymethylcellulose (CMC−Ce^{IV}) was found to be a highly efficient, eco-friendly and recyclable heterogeneous catalyst for the synthesis of 2,4,6-triarylpyridine derivatives in excellent yields *via* one-pot multicomponent reaction of various benzaldehydes, acetophenones, and ammonium acetate under solvent-free condition. Moreover, the present protocol has the merits of easy work-up, short reaction times, and high yields.

Keywords. Cerium(IV) carboxymethylcellulose; 2,4,6-triphenylpyridine; multicomponent reaction; green synthesis.

1. Introduction

Pyridine ring systems are ubiquitous in various synthetic compounds and natural products such as pyridoxol (Vitamin B6), alkaloids (Nicotine) and NAD nucleotides (Figure 1).[1,2] Many pyridine derivatives possess a wide range of important biological and pharmaceutical activities and are already used as dopamine transporter inhibitors,[3] anti-inflammatory agents,[4] antimicrobial agents[5] and topoisomerase I and II inhibitors.[6] Among pyridines, 2,4,6-triarylpyridines (commonly named as Kröhnke pyridines[7]) are widely used as intermediates in the synthesis of chemosensors,[8] asymmetric catalysts[9] and photosensitizers.[10] In addition, some of them are used in supramolecular chemistry because of their Õ-stacking ability.[11] Due to the significance of these pyridine derivatives, development of efficient synthetic protocols remains a topic of considerable interest in modern organic synthesis.

Since Kröhnke firstly reported the synthesis of 2,4,6-triarylpyridines,[7] a variety of efficiently improved procedures have been reported for the preparation of these kind of pyridines. Among the reports, the most common synthetic route for the synthesis of Kröhnke pyridines involve MCRs of direct condensation of acetophenones, benzaldehydes and ammonium acetate in the presence of various catalytic systems or under microwave irradiation without catalysts.[12] The typical catalysts include Preyssler type heteropolyacid $H_{14}[NaP_5W_{30}O_{110}]$,[13] $HClO_4$-SiO_2,[14] I_2,[15] $Bi(OTf)_3$,[16] $MgAl_2O_4$,[17,18] Brønsted-acidic ionic liquid,[19] $ZrOCl_2$,[20] silica vanadic acid,[21] magnetic Fe_3O_4 supported acidic ionic liquid,[22] pentafluorophenylammonium triflate,[23] cerium (IV) ammonium nitrate,[24] and so on. However, many of these methods suffer from drawbacks such as expensive catalyst, harsh reaction conditions, long reaction times, tedious isolation procedure, low to moderate yields and environmental pollution. Therefore, a need still exists for further development of an efficient, reusable, inexpensive and eco-friendly catalyst for the synthesis of Kröhnke pyridines. Despite advances in pyridine synthesis using homogeneous and/or heterogeneous catalyst, no attention so far has been paid to using biopolymer and its derivatives as a carrier in the preparation of supported catalyst for the synthesis of Kröhnke pyridines.

CMC−Na is a nontoxic, biodegradable and water-soluble cellulose derivative that exhibits unique physical chemical and biological properties. Because of bearing carboxylic groups ($-COO^-Na^+$) on its molecular chain, CMC−Na possess the potential to act as an excellent metallic ion exchange agent, which makes it an excellent support to immobilize metal catalysts. More recently, we have reported the employment of CMC as a support for stabilization of palladium

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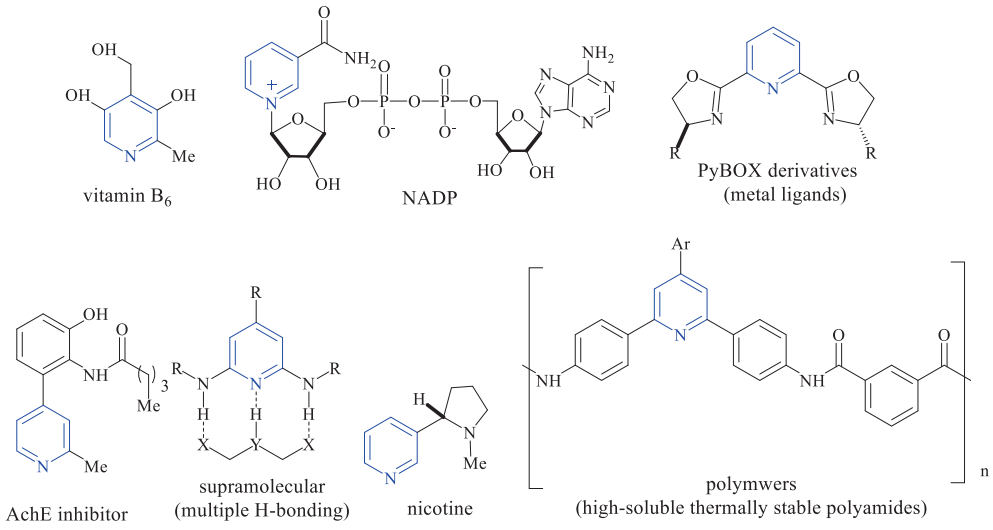


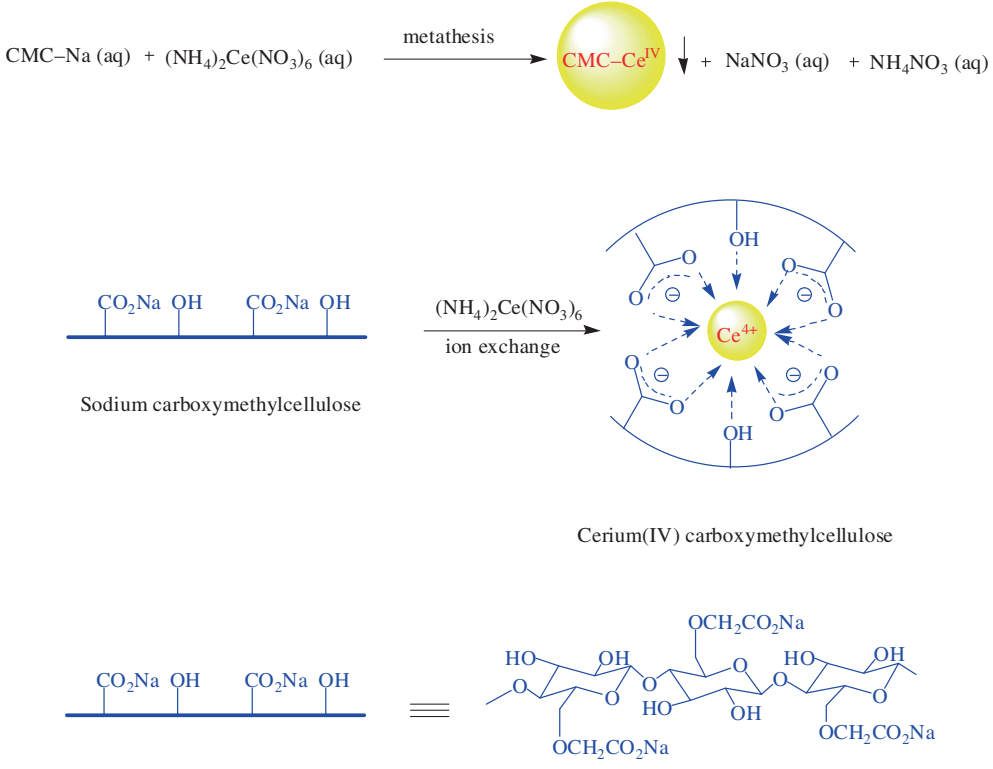
Figure 1. Representative pyridine derivatives.

nanoparticles and its applications in Suzuki–Miyaura and Heck–Mizoroki coupling reactions.[25,26] On the basis of ion-exchange property of CMC−Na, for the first time, we herein report the preparation of the CMC−Ce^{IV} *via* metathesis of Ce^{4+} with CMC−Na in aqueous solution (Scheme 1), and further report its application as an excellent recyclable novel catalyst in pseudo-four component reactions for the synthesis of Kröhnke pyridines under solvent-free condition in air (Scheme 2).

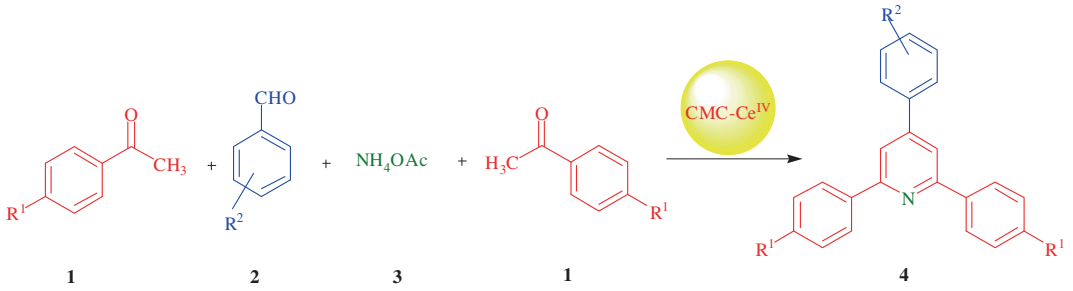
2. Experimental

2.1 *Materials and methods*

All the chemicals used in this study were of commercial grade and were used without further purification. Melting points were measured on a SGW X-4B apparatus in open capillary tubes and uncorrected. ^{1}H NMR spectra were recorded on a Bruker Avance 300 MHz spectrometer using DMSO-d_6 as the solvent containing TMS as an internal standard. HRMS data were recorded using Bruker maXis impact



Scheme 1. Schematic illustration of the preparation and possible structure of CMC–Ce^{IV}.



Scheme 2. CMC–Ce^{IV}-catalyzed pseudo-four component reactions.

detection. FT-IR spectra were recorded on a Nicolet 6700 FT-IR spectrometer using KBr disc. The elemental cerium content of polymeric catalysts was determined by Perkin-Elmer Optima 2000DV ICP-AES. Scanning electron microscopy (SEM) and Energy disperse spectrum (EDX) were conducted with a Philips XL 30ESEM instrument. Progress of the reactions was monitored by thin layer chromatography (TLC) using silica gel G/GF 254 plates.

2.2 *Preparation of cerium(IV) carboxymethylcellulose (CMC−Ce^{IV})*

The 5.5 wt% aqueous solution of cerium(IV) ammonium nitrate (5.5 g dissolved in 94.5 mL H_2O) was slowly added dropwise to an aqueous 1.0 wt% solution of sodium carboxymethylcellulose (1.0 g dissolved in 99 mL H_2O) with constant stirring at room temperature. Yellow solid was precipitated immediately which was left to equilibrate in solution for 12 h. The resulting solid was separated from the solution by suction and washed thoroughly with distilled water, then dried at 60°C to constant weight to provide the CMC−Ce^{IV} as yellow powder. The Ce content was determined to be 2.79 mmol/g by ICP-AES.

2.3 *General procedure for synthesis of 2,4,6-triarylpyridines (Kröhnke pyridines)*

In a 15 mL Schlenk tube, a mixture of acetophenone (2 mmol), ammounium acetate (5 mmol), and aromatic aldehyde (1 mmol) was stirred at 80°C (oil-bath temperature) in the presence of CMC−Ce^{IV} (4.0 mol%) for a specific time. Progress of the reaction was monitored by TLC. Upon the completion of the reaction, the mixture was cooled to room temperature. Then, enough hot ethanol was added and the catalyst was separated by simple suction filtration. The crude product was obtained after removing ethanol, which was further purified by recrystallization with ethanol to afford the pure triarylpyridine.

2.4 *Spectral data of representative compounds*

2.4a *2,4,6-Triphenylpyridine* ***4a***: White solid ; M.p. 134–136°C; IR (KBr): ν = 3058, 1594, 1550, 1494, 1448, 1398, 1177, 1073, 1027, 866, 756, 690 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6, TMS): δ 8.34 (d, J = 7.6 Hz, 4H, ArH), 8.20 (s, 2H, ArH), 8.05 (d, J = 7.6 Hz, 2H, ArH), 7.63–7.47 (m, 9H, ArH).

2.4b *2,6-Diphenyl-4-p-tolylpyridine* ***4c***: Yellow solid; M.p. 118–120°C; IR (KBr): ν = 3034, 2918, 1598, 1544, 1447, 1390, 1251, 1178, 1025, 873, 768, 685 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6, TMS): δ 8.33 (d, J = 7.8 Hz, 4H, ArH), 8.18 (s, 2H, ArH), 7.97 (d, J = 7.8 Hz, 2H, ArH), 7.53 (dt, J = 13.7, 7.0 Hz, 6H, ArH), 7.39 (d, J = 7.8 Hz, 2H, ArH), 2.41 (s, 3H, CH3).

2.4c *2,6-Bis(4-chlorophenyl)-4-(3-nitrophenyl)pyridine* ***4m***: White solid; M.p. 229–231°C; IR (KBr): ν = 3074, 1600, 1547, 1493, 1411, 1385, 1242, 1175, 1089, 1011, 829, 737 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6, TMS): δ 8.87 (s, 1H, ArH), 8.54 (d, J = 7.8 Hz, 1H, ArH), 8.39 (t, J = 11.0 Hz, 7H, ArH), 7.87 (t, J = 8.0 Hz, 1H, ArH), 7.62 (d, J = 8.1 Hz, 4H, ArH).

2.4d *2,6-Bis(4-chlorophenyl)-4-(3-chlorophenyl)pyridine* ***4j***: White solid; M.p. 215–217°C; IR (KBr): ν = 3061, 1594, 1544, 1493, 1425, 1382, 1235, 1176, 1088, 1011, 819, 738 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6, TMS): δ 8.40 (d, J = 8.1 Hz, 4H, ArH), 8.31 (s, 2H, ArH), 8.21 (s, 1H, ArH), 8.06 (s, 1H, ArH), 7.61 (d, J = 8.0 Hz, 6H, ArH). ^{13}C NMR (75 MHz, DMSO-d_6, TMS): δ 155.86, 148.78, 139.99, 137.76, 134.75, 134.50, 131.31, 129.72, 129.29, 129.20, 127.72, 126.66, 117.42. HRMS(ESI) m/z: calcd for $C_{23}H_{14}Cl_3N[M+H]^+$, 410.0265; Found, 410.0267.

2.4e *2,6-Bis(4-chlorophenyl)-4-(3-bromophenyl)pyridine* ***4k***: White solid; M.p. 217–219°C; IR (KBr): ν = 3060, 1594, 1543, 1489, 1424, 1378, 1234, 1174, 1088, 1008, 819, 748 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6, TMS): δ 8.40 (d, J = 8.2 Hz, 4H, ArH), 8.33 (s, 1H, ArH), 8.30 (s, 2H, ArH), 8.09 (d, J = 7.7 Hz, 1H, ArH), 7.72 (d, J = 8.0 Hz, 1H, ArH), 7.61 (d, J = 8.1 Hz, 4H, ArH), 7.53 (t, J = 7.9 Hz, 1H, ArH). ^{13}C NMR (75 MHz, DMSO-d_6, TMS): δ 155.86, 148.76, 140.25, 137.77, 134.75, 132.66, 131.57, 130.51, 129.30, 129.21, 127.07, 123.10, 117.44. HRMS(ESI) m/z: calcd for $C_{23}H_{14}BrCl_2N[M+H]^+$, 453.9759; Found, 453.9751.

2.4f *2,6-Bis(4-methoxyphenyl)-4-(3-chlorophenyl)pyridine* ***4r***: Yellow solid; M.p. 122–125°C; IR (KBr): ν = 3067, 2974, 2838, 1604, 1544, 1383, 1245, 1173, 1112, 1023, 830 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6, TMS): δ 8.30 (d, J = 8.2 Hz, 4H), 8.15 (s, 1H), 8.09 (s, 2H), 8.00 (d, J = 5.4 Hz, 1H), 7.58 (d, J = 5.6 Hz, 2H), 7.09 (d, J = 8.2 Hz, 4H), 3.85 (s, 6H). ^{13}C NMR (75 MHz, DMSO-d_6, TMS): δ 160.30, 156.16, 147.77, 140.12, 133.95, 131.25, 130.78, 128.94, 128.33, 127.09, 126.03, 114.98, 114.02, 55.25. HRMS(ESI) m/z: calcd for $C_{25}H_{20}ClNO_2[M+H]^+$, 402.1255; Found, 402.1256.

3. Results and Discussion

To accomplish the synthesis of Kröhnke pyridines, initially, we prepared and characterized the CMC−Ce^{IV} catalyst. Briefly, the CMC−Ce^{IV} catalyst was prepared by the metathesis from the aqueous solutions of $(NH_4)_2Ce(NO_3)_6$ and CMC−Na at room temperature. The yellow solid precipitated from the solution indicating visually the successful formation of CMC−Ce^{IV} (Scheme 1). The resulting solid was separated by simple suction filtration, washed adequately with distilled water, and dried at 60°C until constant weight was reached. The Ce content in the catalyst was found to be 2.79 mmol/g by ICP-AES.

The FT-IR spectroscopy is an important tool that determines the coordination property of the polymer with metal ions. In order to understand the metal chelation with CMC, we recorded FT-IR spectra of CMC−Na and CMC−Ce^{IV} (Figure 2). The FT-IR spectrum of CMC−Na showed characteristic absorption peaks at 1596 $cm^{-1}$and 1432 cm^{-1} for carboxylate (−COO^-) asymmetric and symmetric stretching vibration. The band at 1045 cm^{-1} is assigned to the ether bond stretches. The broad band ranging of 3100–3700 cm^{-1} is attributed to –OH stretching vibrations. The asymmetric carboxylate peak is negatively shifted to 1421 cm^{-1} in the CMC−Ce^{IV}, revealing the coordination of −COO^- with Ce^{4+}. With respect to metal–carboxylate interactions, the −COO^- group can coordinate cations in four ways; *e.g.*, ionic, monodentate, bidentate and bridging. The wavenumber separation (Δ) in the frequencies between the asymmetric (−COO^-) and symmetric (−COO^-) stretches can be used to identify the type of the interactions. Characteristically, the Δ value falls in the range of 200–320 cm^{-1} for monodentate, 140–190 cm^{-1} for bidentate bridging, and <110 cm^{-1} for chelating bidentate.[27,28] As can be seen from the FT-IR data for Ce(IV) complexes, a Δ value of 33 cm^{-1} (1421–1388 cm^{-1}, as shown in Figure 2b) was obtained, and thus, it is clear that the mode of the carboxylate binding can be correlated to bidentate chelating The stretching vibrations of –OH usually form a broad band in the region of 3100–3700 cm^{-1}. This high-frequency peak shifted from 3415 cm^{-1} in CMC−Na to 3365 cm^{-1} in CMC−Ce^{IV}, This observation indicates that an enhanced coordination is formed between Ce^{4+} with –OH group. The peak at 1060 cm^{-1} is due to –CH_2–O–CH_2– stretching which is slightly shifted to 1043 cm^{-1} for CMC−Ce^{IV}, implying a very weak coordination between –CH_2–O–CH_2–

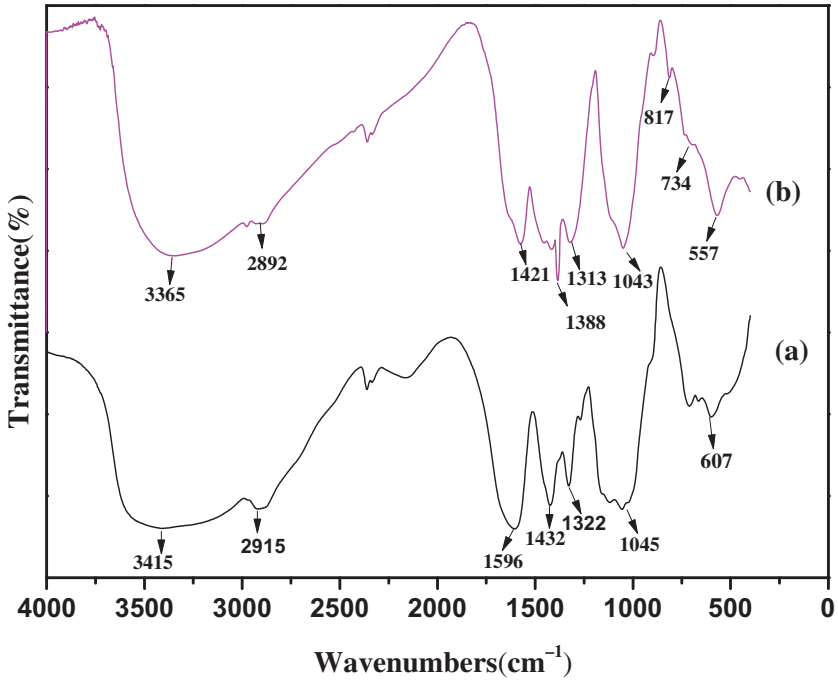


Figure 2. FT-IR of (a) CMC−Na and (b) CMC−Ce^{IV}.

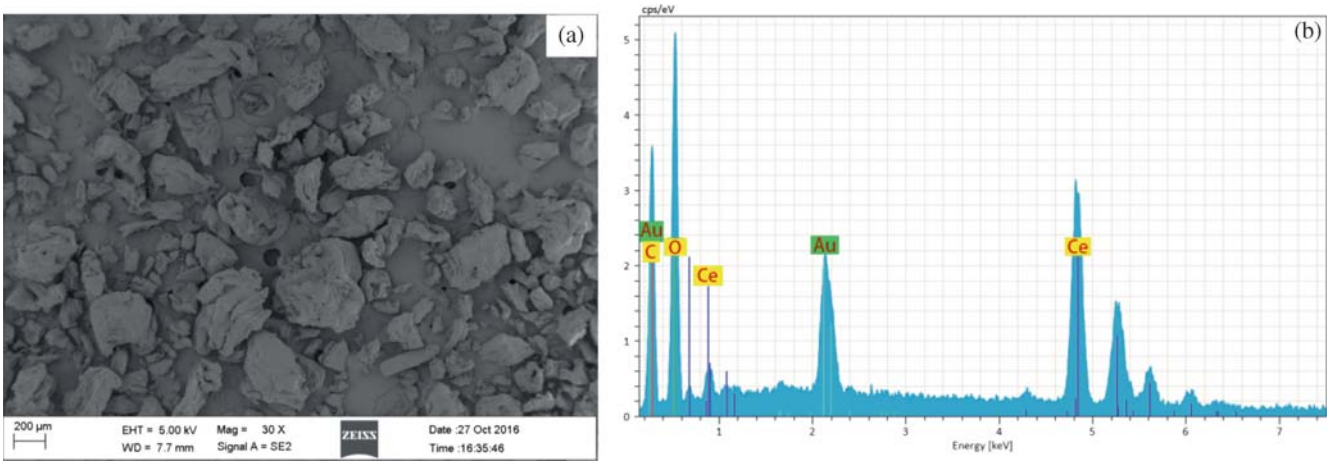


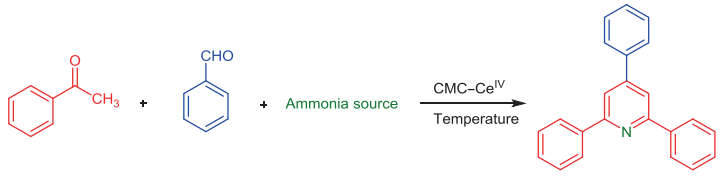
Figure 3. SEM and EDX of fresh CMC−Ce^{IV}.

and Ce(IV). Evidently, CMC can interact with Ce^{4+} *via* both the $-COO^-$ and −OH functional groups, and the possible chemical structure of CMC−Ce^{IV} is suggested in Scheme 1.

Surface morphology and size of the as-synthesized catalyst that play an important role in the catalytic performance were investigated with scanning electron microscopy (SEM) associated with energy-dispersive X-ray spectroscopy (EDX) (Figure 3). SEM images of CMC−Ce^{IV} showed that the catalyst was not smooth and composed of many tiny particles with an average diameter ranging of 50–300 μm (Figure 3a). Also, the presence of carbon, oxygen and cerium in CMC−Ce^{IV} was confirmed by EDX (Figure 3b). The observed Au peak was caused by the contamination from Au sputter. As a result, it is reasonable to assume that Ce^{4+} species are successfully supported on CMC matrix.

In order to optimize the reaction conditions, the reaction of acetophenone (2 mmol), ammonium acetate (5 mmol), and benzaldehyde (1 mmol) were selected

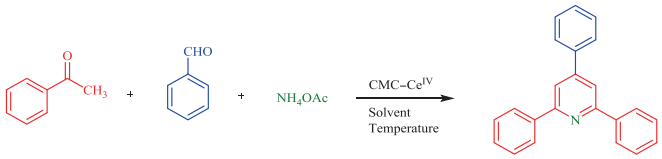
Table 1. Optimization of the reaction conditions.[a]



Entry	Catalyst (mol%)	Temp (°C)	Ammonia source	Time (min)	Yield (%)[b]
1	0	80	NH_4OAc	120	<10
2	1	80	NH_4OAc	120	56
3	2	80	NH_4OAc	120	72
4	3	80	NH_4OAc	90	80
5	4	80	NH_4OAc	60	85 (<10[c])
6	5	80	NH_4OAc	60	84
7	4	r.t.	NH_4OAc	120	<10
8	4	40	NH_4OAc	120	45
9	4	60	NH_4OAc	90	64
10	4	100	NH_4OAc	60	85
11	4	120	NH_4OAc	60	83
12	4	80	NH_4Cl	80	45
13	4	80	NH_4HCO_3	120	nd[d]
14	4	80	NH_4NO_3	120	40

[a]Conditions: acetophenone (2 mmol), benzaldehyde (1 mmol), ammonia source (5 mmol), and catalytic amount of the CMC–Ce^{IV} under solvent-free condition.
[b]Isolated yield.
[c]4.0 mol% of CMC–Na as catalyst.
[d]No detection.

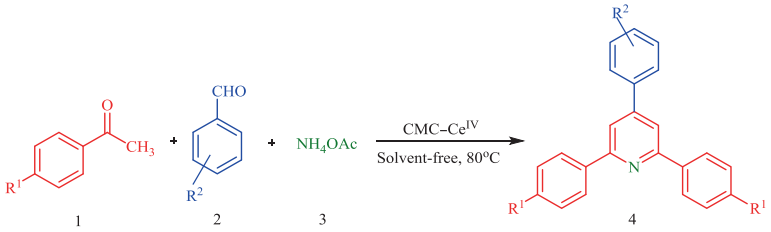
Table 2. Effect of different solvents on the synthesis of **4a**.[a]



Entry	Solvent	Time (min)	Yield (%)[b]
1	Solvent-free	60	85
2	EtOH	60	71
3	H_2O	90	40
4	CH_2Cl_2	90	53
5	CH_3CN	90	68
6	*n*-hexane	90	62
7	Toluene	90	55

[a]Conditions: acetophenone (2 mmol), benzaldehyde (1 mmol), ammonium acetate (5 mmol), and CMC–Ce^{IV} (4.0 mol%) at 80°C.
[b]Isolated yield.

Table 3. Synthesis of 2,4,6-triphenylpridine derivatives.[a]



Entry	**1** (R^1-)	**2** (Ar-)	Product	Yield (%)[b]	Time (min)	M.p. (°C) Found	M.p. (°C) Reported [Ref.]
1	H	C_6H_5	4a	85	60	134–136	133–135[16]
2	H	4-Cl-C_6H_4	4b	84	75	126–128	127–128[17]
3	H	4-$CH_3C_6H_4$	4c	86	60	118–120	118–121[20]
4	H	3-Cl-C_6H_4	4d	81	75	115–117	116–117[29]
5	H	3-CH_3O-C_6H_4	4e	87	60	123–126	124–127[17]
6	Cl	C_6H_5	4f	89	60	176–178	175–178[17]
7	Cl	4-Cl-C_6H_4	4g	85	75	262–264	264–265[15]
8	Cl	4-Br-C_6H_4	4h	87	75	>300	—
9	Cl	4-CH_3-C_6H_4	4i	88	60	202–205	201–203[14]
10	Cl	3-Cl-C_6H_4	4j[c]	82	90	215–217	—
11	Cl	3-Br-C_6H_4	4k[c]	81	90	217–219	—
12	Cl	3-CH_3O-C_6H_4	4l	89	60	159–161	159–160[17]
13	Cl	3-NO_2-C_6H_4	4m	84	90	229–231	230–232[30]
14	OCH_3	C_6H_5	4n	92	90	98–101	98–99[14]
15	OCH_3	4-CH_3-C_6H_4	4o	90	60	154–157	155–156[14]
16	OCH_3	4-CH_3O-C_6H_4	4p	93	60	135–137	135–137[21]
17	OCH_3	4-HO-C_6H_4	4q	87	75	243–245	243–244[31]
18	OCH_3	3-Cl-C_6H_4	4r[c]	88	75	122–125	—
19	H	2-Furyl	4s[d]	83	180	160–161	160–162[17]
20	H	2-Thienyl	4t[d]	85	180	161–163	161–163[16]

[a]Conditions: acetophenone (2 mmol), aldehyde (1 mmol), ammonium acetate (5 mmol), and CMC–Ce^{IV} (4.0 mol%) under solvent-free condition at 80°C.
[b]Isolated yield.
[c]New compound.
[d]Reaction was performed at 100°C.

Table 4. Comparison of the efficiencies of different methods for the synthesis of **4a**.

Entry	Catalyst	Condition	Time (min)	Yield (%)[Ref.]
1	DPAT	Solvent-free,120°C	240	96[23]
2	$H_{14}[NaP_5W_{30}O_{110}]$	Solvent-free,120°C	210	98[13]
3	$Bi(OTf)_3$	Solvent-free,120°C	120	89[16]
4	$[HO_3S(CH_2)_4MIM][HSO_4]$	Solvent-free,120°C	180	88[19]
5	Nanocrystalline $MgAl_2O_4$	Solvent-free,120°C	180	85[17]
6	TCT	Solvent-free,130°C	240	70[32]
7	PFPAT	Solvent-free,120°C	120	89[23]
8	$HClO_4$–SiO_2	Solvent-free,120°C	240	80[14]
9	n-TSA	Solvent-free,110°C	120	93[33]
10	PPA-SiO_2	Solvent-free,120°C	60	82[34]
11	CMC–Ce^{IV}	Solvent-free, 80°C	60	85This work

as a model at 80°C under solvent-free condition. The effect of the amount of catalyst on the reaction yield was examined first. In the absence of the catalyst, the desired product was obtained in <10% yield after 2 h (Table 1, entry 1), which indicated that CMC−Ce^{IV} plays a key role in the reaction. When the amount of the catalyst was increased from 1 to 4 mol%, yield also increased from 56% to 85% (Table 1, entries 2, 3, 4 and 5). However, a higher amount of the catalyst (5 mol%) did not increase the yield noticeably (Table 1, entry 6). Thus, 4 mol% of catalyst is enough for the completion of the model reaction. The effect of various ammonia sources, such as NH_4OAc, NH_4Cl, NH_4HCO_3, NH_4NO_3 on the reaction was also investigated (Table 1, entries 5, 12, 13 and 14). Under similar conditions, when NH_4OAc acted as ammonia source, the product was obtained in highest yield in a short time. It is worth noting that less than 10% of product yield was obtained using catalytic amount of CMC−Na as catalyst (Table 1, entry 5).

It is well known that the reaction medium plays an important role on the reaction. Therefore, the reaction was performed in various solvents and under solvent-free conditions. As shown in Table 2, in comparison with conventional solution methods, the yields of the reaction under solvent-free condition are higher and the reaction times are shorter.

To establish the scope of this method, synthesis of a variety of triarylpyridines from different aldehydes and acetophenones was studied under the optimized reaction conditions. In all cases, aromatic aldehydes with substituents carrying either electron-donating or electron-withdrawing groups reacted successfully and gave the products in high yields (81–93%) in short times (60–90 min). The effect of the nature of

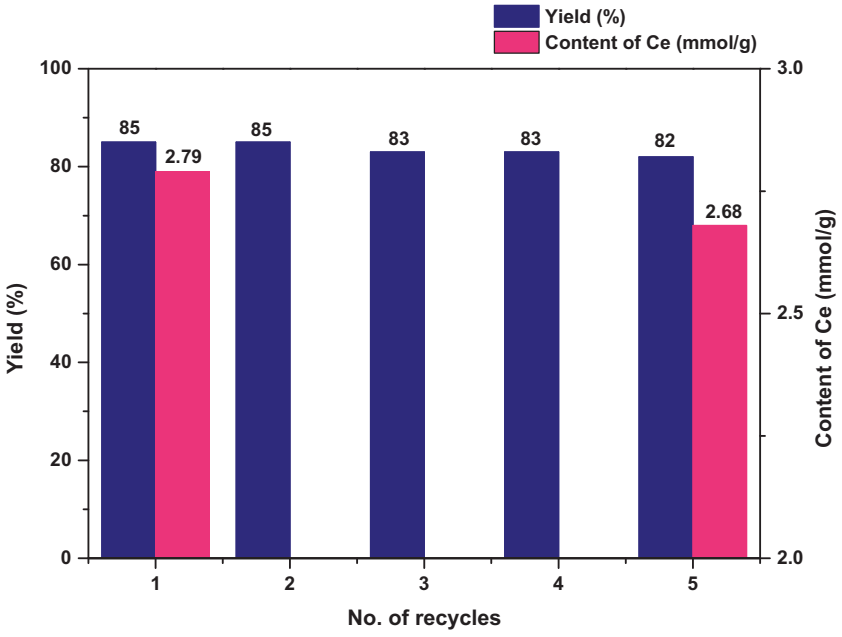


Figure 4. Recycling of CMC–Ce^{IV} catalyst.

substituents on the aromatic ring showed no obvious effect on this conversion. The results are summarized in Table 3.

At last, we compared the efficiency of our work with other reports. As shown in Table 4, our procedure reported herein has more merits than other published procedures; such as, reusable catalyst, excellent yields, short reaction times, simple work-up, lower dose of catalyst, and no use of organic solvents.

Reusability of the catalyst is highly desirable for a catalytic process. In this regard, the recyclability of CMC−Ce^{IV} was investigated in the model reaction of acetophenone, benzaldehyde, and ammonium acetate under the optimized conditions. After completion of the reaction, hot ethanol was added to the reaction mixture and all the catalyst was separated from the mixture simply by use of suction filter process. The recovered catalyst was washed with ethanol, dried at room temperature, and reused for five consecutive runs. As shown in Figure 4, the catalyst could be reused at least five runs without significant loss of its catalytic activity in comparison with the fresh catalyst. To further understand the underlying reason for the excellent recyclability of the CMC−Ce^{IV} catalyst, ICP-AES and SEM along with EDX were employed. Measurement of the Ce content of the catalyst after five consecutive runs using ICP-AES analysis showed an extremely low level of leaching with only 3.9% loss of the initially

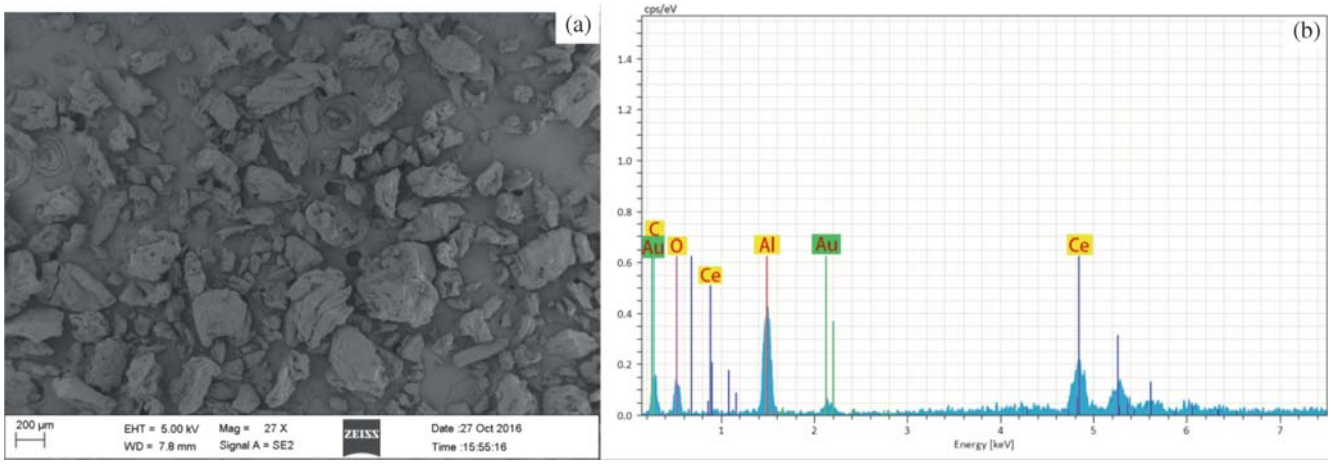
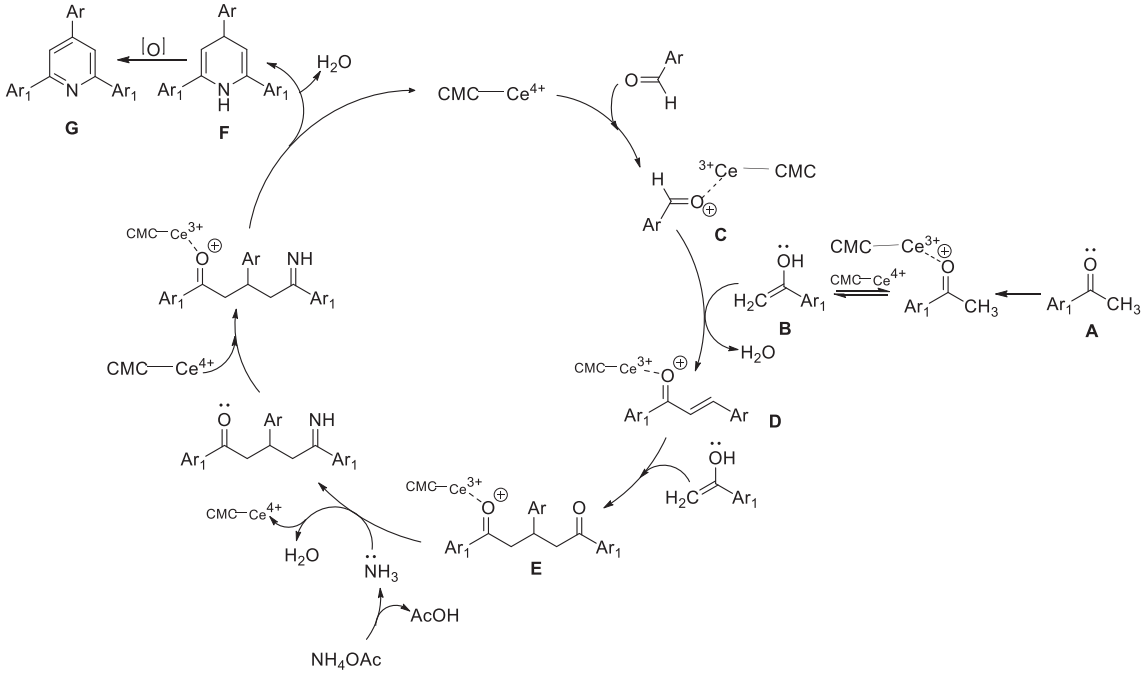


Figure 5. SEM and EDX of used CMC−Ce^{IV} after five runs



Scheme 3. Purposed mechanism for CMC−Ce^{IV} catalyzed formation of 2,4,6-triarylpyridines.

added Ce. The SEM of the reused catalyst after 5th run (Figure 5a) showed that there were no apparent changes in the CMC−Ce^{IV}, and EDX demonstrated that the Ce element was dispersed on the reused catalyst (Figure 5b).

A plausible mechanism for the preparation of 2,4,6-triarylpyridines in the presence of CMC−Ce^{IV} is proposed based on previous reports[15,16,20] (Scheme 3). Initially, acetophenone (A) is converted into its enol form (B) (*via* tautomerization) with the assistance of CMC−Ce^{IV}, which affords nucleophilic addition on the intermediate (C) to give chalcone product (D) (*via* aldol condensation). Then, the second molecule of acetophenone undergoes Michael addition reaction with (D) to form the 1,5-diketone intermediate (E). This 1,5-diketone on reaction with ammonium acetate followed by cyclization and tautomerization gives compound (F). Finally, air oxidation of (F) leads to the formation of corresponding target product (G).

4. Conclusions

In conclusion, a one-pot pseudo-four-component synthesis of 2,4,6-triarylpyridines in the presence of cerium carboxymethylcellulose (CMC−Ce^{IV}) as a novel and reusable catalyst under solvent-free condition has been developed. To the best of our knowledge, this is the first example of preparation of CMC−Ce^{IV} for the synthesis of these pyridine derivatives under solvent-free conditions. Present methodology offers very attractive features such as cleaner reaction profile, shorter reaction times, higher yields, and tolerance of wide scope of substrates which make the process efficient and practical. Further synthetic application of the newly developed catalyst is in progress.

Supplementary Information (SI)

All additional information pertaining to characterization of known compounds using ^{1}H NMR spectra, and characterization of the new compounds using ^{1}H NMR, ^{13}C MNR spectra and HRMS (Figures S1 to S9) are given in the supporting information, available at www.ias.ac.in/chemsci.

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