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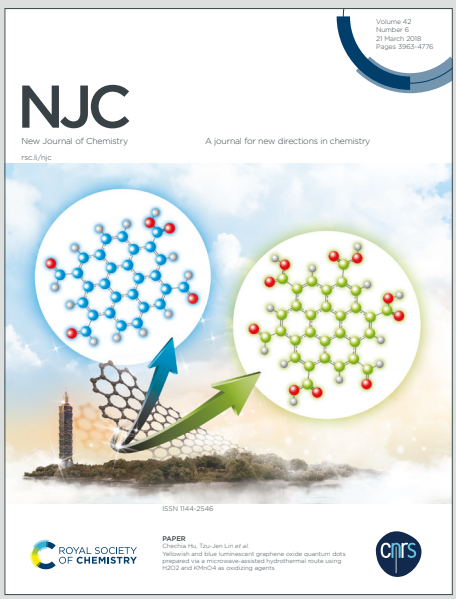

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ARTICLE

One-pot three-component synthesis of 1-amidoalkyl naphthols and polyhydroquinolines using deep eutectic solvent: A green method and mechanistic insight

Received 00th January 20xx,
Accepted 00th January 20xx

DOI: 10.1039/x0xx00000x

www.rsc.org/

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Abstract. The multicomponent synthesis of 1-amidoalkyl naphthols and polyhydroquinolines has been developed as an atom-economic procedure catalyzed by a deep eutectic solvent ([CholineCl][$ZnCl_2$]$_3$). The reactions proceed smoothly at low temperatures for a short reaction time without the use of toxic and volatile organic solvents. Deep eutectic solvents are capable of not only allowing multicomponent reactions to proceed in high yield but also controlling the selectivity towards desired products. The mechanistic insight was examined by HRMS (ESI) to propose a plausible mechanism. Furthermore, [CholineCl][$ZnCl_2$]$_3$ can be recycled up to three consecutive cycles with an insignificant loss of catalytic activity under the optimized conditions.

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Introduction

Deep eutectic solvents (DESs) are a new class of green solvents analogous to ionic liquids**,** which are generally prepared by mixing a hydrogen bond acceptor with a hydrogen bond donor under a certain temperature.[1] The deep eutectic solvents are generally based on readily available and low-cost components such as choline chloride and hydrogen bond donor (urea, ethylene glycol, glycerol, glucose, *etc*.), which makes them more suitable for large-scale applications.[2] These compounds played a vital as catalysts or solvents for organic synthesis reactions.[3, 4] DESs have been used in many areas such as extraction of cellulose,[5] extraction of chitin,[6] extraction of polysaccharides, lipid, and various polarities compounds,[7-10] electrolyte in dye-sensitized solar cells,[11] electrolyte in lithium-Ion battery,[12] and electrolyte in electrochemical double-layer capacitor.[13] Recently, deep eutectic solvent [CholineCl][$ZnCl_2$]$_3$ from choline chloride and zinc chloride emerged as the green catalyst in organic transformation. [CholineCl][$ZnCl_2$]$_3$ has been successfully applied on several reactions such as Friedel-Crafts,[14-17] Paal-Knorr,[18] Diels-Alder,[19] aza Diels–Alder.[20]

Multicomponent reactions allow the rapid synthesis of diverse compounds in a cost- and time-effective manner from abundant and available starting materials. The prominent features of multicomponent reactions are reducing overall steps and minimizing the production of hazardous waste.[21-23] 1-Amidoalkyl-2-naphthols containing 1,3-amino oxygenated functional groups possess many biological characteristics and pharmacological activities such as antitumor,[24, 25] antibiotics,[26] antianginals,[27-29] antimalarial,[30, 31] and HIV protease inhibitors.[32, 33] Various reports have been published regarding the preparation of 1-amidoalkyl-2-naphthol derivatives through the C-N, C-C coupling reaction from 2-naphthol, aldehydes, and amides in the presence of various catalysts such as sulfonated material,[34-36] ionic liquid,[37-41] nano-S_8,[42] Brönsted acid,[43-45] deep eutectic solvents,[46] $ZrO(OTf)_2$,[47] metal salt,[48] oxide metal,[49] thiamine hydrochloride,[50] trityl chloride,[51] I_2,[52] montmorillonite K10 clay,[53] cation-exchanged resins,[54] etc. However, many disadvantages existed in these methods, including prolonged reaction time, low yield, high reaction temperature, and the formation of by-products.

Polyhydroquinolines were important compounds of *N*-heterocycles, which was obtained from the condensation reaction of aldehydes, dimedone, ethyl acetoacetate, and ammonium acetate. These compounds exhibited many critical biological activities and were applied in the pharmaceutical field, such as potential α-amylase and α-glucosidase inhibitors,[55] antitumor,[56] calcium antagonists,[57] antiplasmodial,[58] antibacterial,[59] antiproliferative,[60] antimalarial,[61] antiatherosclerotic,[62] and antidiabetic properties.[63] There were many methods for synthesizing polyhydroquinoline derivatives by using the catalysts such as metal triflates,[64] zeolites,[65] ionic liquids,[66, 67] nanoparticles,[68, 69] heterogeneous nanocatalyst,[70, 71] organic-solid-acid,[72] deep eutectic solvents,[73, 74] enantioselective organocatalysis,[75] Brönsted acids,[76] etc.

In this work, we report the efficient protocol for the multicomponent synthesis of *N*-containing heterocyclics, including the preparation of 1-amidoalkyl-2-naphthols and polyhydroquinolines using [CholineCl][$ZnCl_2$]$_3$ as a green catalyst

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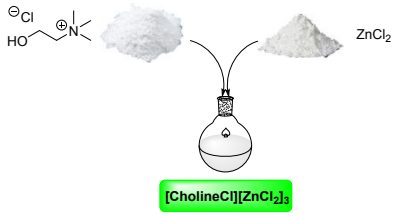
under solvent-free condition. The plausible mechanism was proposed by the mean of HRMS (ESI) analysis.

Results and discussion

Preparation of [CholineCl][$ZnCl_2$]$_3$

[CholineCl][$ZnCl_2$]$_3$ was prepared from previous reports from choline chloride and zinc chloride with a molar ratio of 1:3 (Scheme 1).[14, 18, 77, 78] The reaction was carried out at 100 °C until a clear colorless liquid obtained. The structure of the catalyst was determined by ^{1}H and ^{13}C NMR spectra, FT-IR, HR-MS (ESI), Raman, and TGA. ^{1}H and ^{13}C NMR spectra provided signals consistent with previous literature (*Fig. S1 and S2*†).[79] These results showed that no impurities were detected by the NMR spectrum. The Fig. S3† showed the FT-IR spectra of [CholineCl][$ZnCl_2$]$_3$, and reused [CholineCl][$ZnCl_2$]$_3$ after three cycles. The vibrational band around 3409 cm^{-1} was from the O–H-stretching of the hydroxyl group. Moreover, the bands at 3000~2890 cm^{-1} and 1438~1400 cm^{-1}, indicated the presence of alkyl groups. The peaks at ~1125 and ~1063 cm^{-1} were from the C-N vibration. The signals at ~1045 and ~950 cm^{-1} were found to correlate the C–O and C–C–O asymmetric stretching. These results were compared with published works.[18, 80] As displayed in Fig. S4†, the Raman spectrum indicated the signal at 130 cm^{-1}, assigned to Zn–Cl stretching modes.[81] The absorption peaks of $ZnCl_3^-$ were found at ~280 cm^{-1} and ~340 cm^{-1}.[82] Based on the Thermogravimetric analysis (TGA), the results showed that the catalyst worked stably to 350-370 °C, responding well to reaction conditions (Fig. S5†).



Scheme 1. [CholineCl][$ZnCl_2$]$_3$ synthesis process

Synthesis of 1-amidoalkyl naphthol derivatives

The catalytic activity of [CholineCl][$ZnCl_2$]$_3$ was investigated by the synthesis of *N*-((2-hydroxynaphthalen-1-yl)(phenyl)methyl)benzamide, which performed between 2-naphthol, benzaldehyde, and benzamide. The results were listed in Table S1†. This reaction was conducted to investigate the influence of reaction parameters such as reaction temperature, reaction time, and amount of catalysts. To optimal condition reaction, the C-N coupling reaction was performed at different temperatures such as room temperature, 60, 80, 100, and 120 °C for 40 min, giving the best yield of the main product at 60 °C (Table S1†, entry 2). Then, the reaction time was investigated by varying the time from 10 min to 60 min (Table S1†, entries 6-10). The amount of the catalyst was also evaluated, and 20 mol% of [CholineCl][$ZnCl_2$]$_3$ was chosen to study the reaction between 2-naphthol, benzaldehyde, and benzamide. As can be seen in Table 1, the effect of various catalyst was investigated with metal chlorides ($ZnCl_2$, $FeCl_2$, and $AlCl_3$), choline chloride, methanesulfonic acid, other deep eutectic solvents including [CholineCl][acid oxalic], [CholineCl][urea]$_2$, and [CholineCl][ethyleneglycol], and ionic liquids such as [BMIM]Cl and [EMIM]Cl. No reaction occurred when the reaction was carried out with [CholineCl][ethyleneglycol] or without a catalyst. The best result was observed in the presence of 20 mol% of [CholineCl][$ZnCl_2$]$_3$ at 60 °C for 40 min (Table 1, entry 10). The synthesis of *N*-((2-hydroxynaphthalen-1-yl)(phenyl)methyl)benzamide was also scaled up from 1 mmol scale to 10 mmol scale without a significant drop in yield.

Table 1. The effect of various catalyst for the synthesis of *N*-((2-hydroxynaphthalen-1-yl)(phenyl)methyl)benzamide.[a]

Entry	Catalysts	Isolated yield %
1	$ZnCl_2$	61
2	$FeCl_2$	65
3	$AlCl_3$	74
4	Choline chloride	58
5	MsOH[b]	60
6	[CholineCl][acid oxalic]	70
7	[CholineCl][urea]$_2$	10
8	[CholineCl][ethylene glycol]	NR[c]
9	[CholineCl][lactic acid]$_9$	59
10	[CholineCl][$ZnCl_2$]$_3$	94 (83)[d]
11	[BMIM]Cl	6
12	[EMIM]Cl	8
13	None	NR[b]

[a] Reaction conditions: 2-Naphthol (1.0 mmol), aldehydes (1.0 mmol), and acetamides (1.0 mmol) at 60 °C for 40 min with 20 mol% of catalyst.
[b] MsOH = Methanesulfonic acid
[c] NR: No reaction.
[d] 10 mmol scale.

To compare the present method with previous literature, the reaction of 2-naphthol, benzaldehyde, and benzamide afforded *N*-((2-hydroxynaphthalen-1-yl)(phenyl)methyl)benzamide in excellent yield within 15 min under solvent-free condition (Table 2, entry 8). In general, the availability of the reagents, mild condition, and simple reaction rendered this method more attractive from the viewpoint of green chemistry.

Table 2. Comparison of current work with previous reports for preparation of 1-amidoalkyl naphthols.

Entry	Catalysts	Temp. (°C)	Time (min)	Yield (%)
1	Fe_3O_4@C-SO_3H MNPs Solvent-free	80	160	88[83]
2	[Msim]$FeCl_4$ (5 mol%) Solvent-free	110	3	95[84]
3	IL@MNP (1.5 mol%) Dichloroethane (10 mL)	30	70	62[85]
4	AIL@MNP Solvent-free	90	22	91[86]
5	Fe_3O_4@SiO_2@IL-PVP (0.015 g) Solvent - free	80	15	95[87]
6	nano-Fe_3O_4@ZrO_2-	100	20	97[88]

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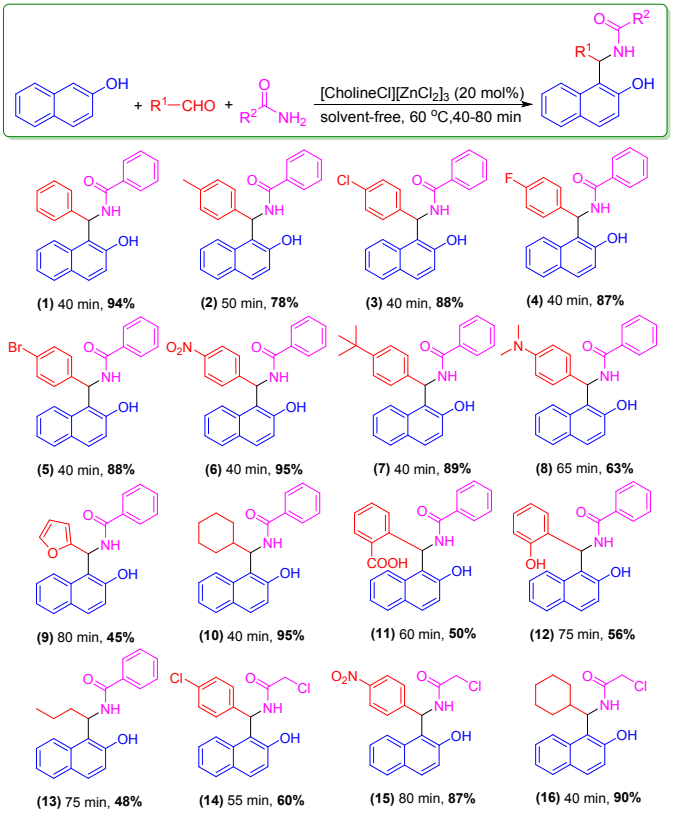
7	H_3PO_4 (0.06 g) $[C_6(MPy)_2][CoCl_4]^{2-}$ (0.2 g) Solvent-free	120	25	75[41]
8	This work: $[CholineCl][ZnCl_2]_3$ (20 mol%) Solvent-free	60	40	94

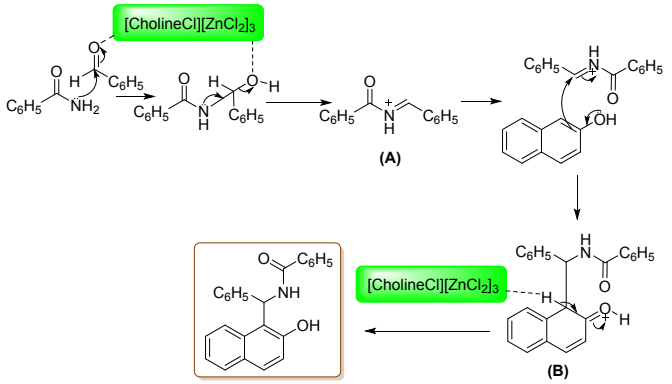
The reaction was then conducted with several amides and aldehydes using $[CholineCl][ZnCl_2]_3$ as the catalyst under the optimal conditions. The results were presented in Table 3. First, the reaction was performed with aldehydes containing the electron-donating groups ($-CH_3$, $-C(CH_3)_3$) and electron-withdrawing groups (-Cl, -F, -Br, $-NO_2$, and $-N(CH_3)_2$) at the *para* position on the benzene ring afforded desired products in high yield (compounds 1-8, Table 3). Next, the reaction was further investigated with furfural; the yield of the product decreased significantly. The five-membered heterocyclic compounds containing the oxygen resulted in lower activity than the benzene ring (compound 9, Table 3). Aldehyde derivatives containing substituents (-COOH, -OH) at the *ortho* position on the benzene ring provided the desired products in low yields due to steric obstacles (compounds 11-12, Table 3). Next, the reaction of 2-naphthol, aldehydes, and 2-chloroacetamide in the presence of $[CholineCl][ZnCl_2]_3$ occurred smoothly with good to excellent yields (compounds 14-16, Table 3).

Table 3. Synthesis of 1-amidoalkyl naphthol using $[CholineCl][ZnCl_2]_3$ as the catalysis.[a]



[a] Reaction condition: 2-naphthol (1.0 mmol), aldehydes (1.0 mmol) and amides (1.0 mmol) in the presence of $[CholineCl][ZnCl_2]_3$ (20 mol%) at 100 °C. Isolated yields.

The reaction mechanism for the preparation of 1-amidoalkyl-2-naphthol was illustrated in Scheme 2. From the result of the HRMS analysis, a plausible reaction mechanism was proposed. Firstly, benzaldehyde was activated by [CholineCl]-$[ZnCl_2]_3$. Then, benzamide attacked to the carbonyl group of the benzaldehyde to give intermediate (A), detected by HRMS (*m/z* 209.0562 $[M+H]^+$, Cacld. *m/z* 209.0840). Further, 2-naphthol was attacked and condensed with intermediate (A). The intermediate (B) was affirmed by HRMS (*m/z* 264.0980 $[M+H]^+$, Cacld. *m/z* 264.1019). Next, the deprotonation of intermediate (B) generated the desired product, which was detected by HRMS (*m/z* 354.1489 [M] Cacld. *m/z* 354.1433).



Scheme 2. The plausible mechanism for preparation of 1-amidoalkyl naphthol.

Synthesis of polyhydroquinoline derivatives

To evaluate the catalytic activity of $[CholineCl][ZnCl_2]_3$, the synthesis of polyhydroquinolines was carried out with aldehydes (1.0 mmol), dimedone (1.0 mmol), ethyl acetoacetate (1.0 mmol), and ammonium acetate (1.0 mmol). The results were described in Table S2†. Initially, the parameter of condensation reaction was studied with different temperatures (RT, 60, 80, 100, and 120 °C), different time (5, 10, 15, 20, 30, and 40 min), and the amount of catalyst (0, 0.5, 1, 2, 5, and 10 mol%). The optimal conditions were observed in the formation of ethyl 2,7,7-trimethyl-5-oxo-4-phenyl-1,4,5,6,7,8-hexahydro-quinoline-3-carboxylate in the concentration of 0.5 mol% of $[CholineCl][ZnCl_2]_3$ for 40 min at 100 °C. The condensation reaction was investigated with various catalysts, and the major product was achieved in the highest yield with $[CholineCl][ZnCl_2]_3$ (entry 7, Table 4).

Table 4. Effects of different catalysts for the preparation of polyhydroquinolines.[a]

Entry	Catalyst	Isolated yield (%)
1	MgO	35
2	Al_2O_3	28
3	$ZnCl_2$	54
4	$AlCl_3$	63
5	$FeCl_3$	32
6	$Cu(NO_3)_2$	32
7	$[CholineCl][ZnCl_2]_3$	87

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[a] Reaction condition: Benzaldehyde (1.0 mmol), dimedone (1.0 mmol), ethyl acetoacetate (1.0 mmol), ammonium acetate (1.0 mmol), and catalysts (0.5 mol%) at 100 °C for 20 min.

The preparation of ethyl 2,7,7-trimethyl-5-oxo-4-phenyl-1,4,5,6,7,8-hexahydro-quinoline-3-carboxylate from benzaldehyde, ethyl acetoacetate, dimedone, and ammonium acetate under the present method was compared with previous reports. From the results of Table 5, the use of [CholineCl][$ZnCl_2$]$_3$ demonstrated some advantages such as short reaction time, the ease of being recycled, and economic and environmental efficiency.

Table 5. Comparing the current method in the reaction of benzaldehyde, dimedone, ethyl acetoacetate, and ammonium acetate with previous literature.

Entry	Catalysts	Temp. (°C)	Time (min)	Yield[a] %
1	$CoFe_2O_4$@Pr (0.04 g) EtOH (2 mL)	reflux	400	91[89]
2	BIL@MNP (0.25 mol%) solvent-free	70	15	92[90]
3	TBA-AMPS (Ionic liquid) (10 mol%) MeOH	reflux	8	97[91]
4	$[(CH_2)_4SO_3HMIM][HSO_4]$ (25 mol%) EtOH (5 mL)	reflux	67	90[92]
5	$[TBA]_2[W_6O_{19}]$ (7 mol%) solvent-free	110	20	95[93]
6	$Hf(NPf_2)_4$ (1 mol%) Perfluorodecalin	60	180	95[66]
7	Ni@IL-OMO (0.5 mol%) solvent-free	15	70	96[94]
8	$[BBMIm][HSO_4]_2$ (2.4 mol%) solvent-free	60	18	92[95]
9	This work: [CholineCl][$ZnCl_2$]$_3$ (0.5 mol%) solvent-free	100	20	87

With the optimized reaction conditions in hand, various starting materials were tested to synthesize polyhydroquinoline derivatives with [CholineCl][$ZnCl_2$]$_3$ under solvent-free conditions. All the condensation reactions were carried out at 100 °C for 20 min. The results were presented in Table 6. In the case of cyclohexanecarboxaldehyde, the condensation reaction was occurred to form ethyl 4-cyclohexyl-2,7,7-trimethyl-5-oxo-1,4,5,6,7,8-hexahydroquinoline-3-carboxylate with good yield (compound 17, Table 6). The catalytic effect of the [CholineCl][$ZnCl_2$]$_3$ was then investigated with aromatic aldehydes containing the electron-withdrawing (e.g., Cl, –F, $–OCH_3$, $–N(CH_3)_2$) and electron-donating groups (e.g., CH_3, $–C(CH_3)_3$) at the *ortho* or *para* position giving the major products were obtained in good yields (compounds 19-25, Table 6). The presence of the substituent at the *ortho* position of the aromatic aldehydes increased the steric hindrances, which gave the desired products in low yields (compounds 26-27, Table 6). The heterocyclic aldehydes containing oxygen showed less reactivity toward condensation reactions for the synthesis of ethyl 4-(furan-2-yl)-2,7,7-trimethyl-5-oxo-1,4,5,6,7,8-hexahydroquinoline-3-carboxylate and their derivatives (compounds 29-34, Table 6).

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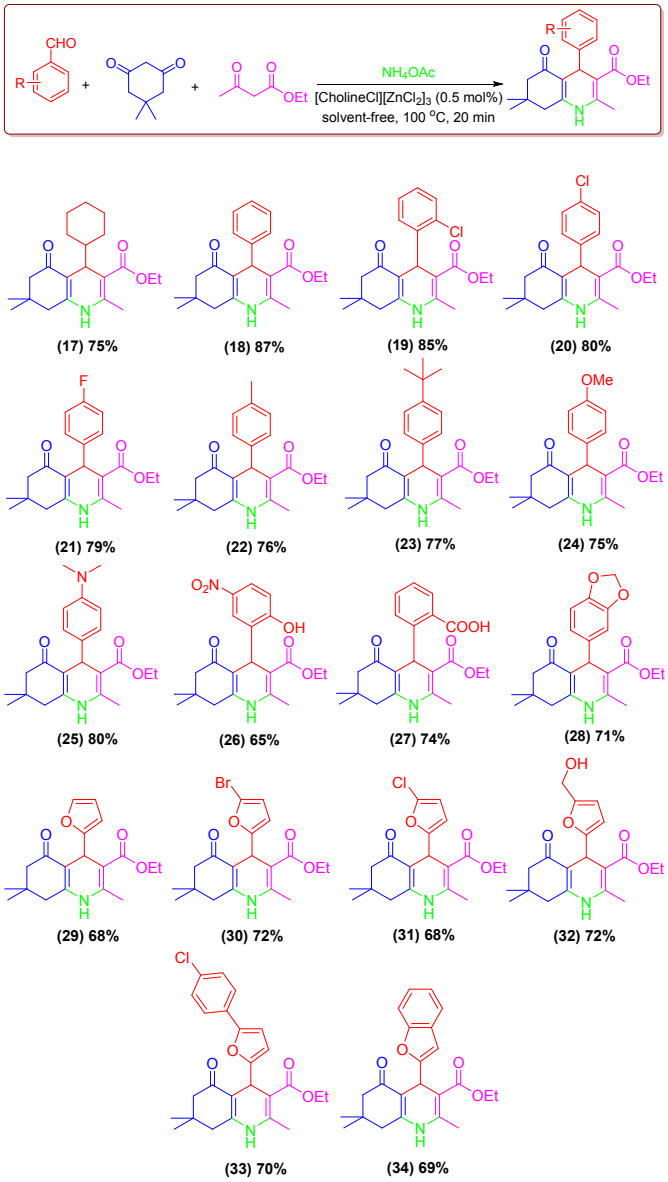
Table 6. [CholineCl][$ZnCl_2$]$_3$-catalyzed for synthesis of poly-hydroquinolines.[a]



[a] Reaction condition: Aldehydes (1.0 mmol), dimedone (1.0 mmol), ethyl acetoacetate (1.0 mmol) and ammonium acetate (1.0 mmol) in the presence of [CholineCl][$ZnCl_2$]$_3$ (0.5 mol%) at 100 °C in 20 min. Isolated yields.

The synthesis of polyhydroquinoline derivatives was carried out through the reaction of benzaldehyde, ethyl acetoacetate, dimedone, and ammonium acetate in the presence of

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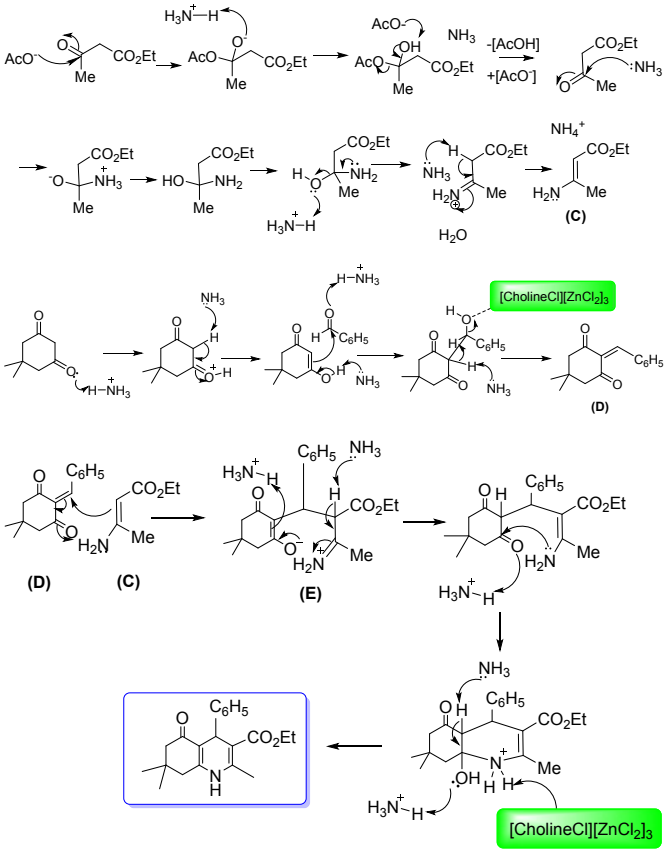
[CholineCl][$ZnCl_2$]$_3$ as the green catalyst (Scheme 3), and the precursors were monitored by HRMS. First, ethyl acetoacetate interacted with ammonium acetate to form the imine (C), detected by HRMS (*m/z* 130.95869). On the other hand, the Knoevenagel condensation occurred between benzaldehyde and the active methylene group of dimedone in the presence of [CholineCl][$ZnCl_2$]$_3$ to generate the *α,β*-unsaturated compound (D) with *m/z* 229.10520. Next, the imine (C) reacted with *α,β*-unsaturated compound (D) to give the intermediate (E), determined by HRMS (*m/z* 358.20593). Finally, the ethyl 2,7,7-trimethyl-5-oxo-4-phenyl-1,4,5,6,7,8-hexahydro-quinoline-3-carboxylate was obtained *via* the cyclization reaction, and this product was also detected by HRMS (*m/z* 340.19134). Besides, some by-products were observed by HRMS with different *m/z* values such as 330.16392 and 350.21205, which could be assigned for diethyl 2,6-dimethyl-4-phenyl-1,4-dihydropyridine-3,5-dicarboxylate (from benzaldehyde and ethyl acetoacetate)[96] and 3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (from benzaldehyde, dimedone, and ammonium acetate),[97] respectively.



Scheme 3. Proposed mechanism for preparation of polyhydroquinolines

To study the reusability of the catalyst, the recovered [CholineCl][$ZnCl_2$]$_3$ was tested in the synthesis of 1-amidoalkyl naphthol and polyhydroquinoline (Fig. 1). The [CholineCl][$ZnCl_2$]$_3$ still remained a good activity after three consecutive runs. The recovered catalyst was recorded by FT-IR spectroscopy. From Fig. S3†, FT-IR spectra of fresh and recovered [CholineCl][$ZnCl_2$]$_3$ catalysts showed similar vibration bands within 3500 cm^{-1}, 1619 cm^{-1}, and 1475 cm^{-1}. The fresh and reused catalyst showed that the structure has remained mostly unchanged.

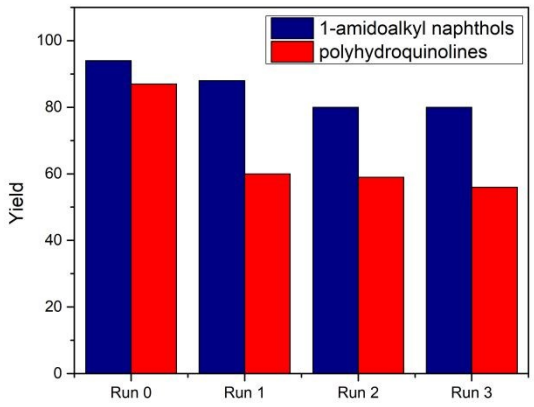


Figure 1. Recycling of [CholineCl][$ZnCl_2$]$_3$

Experimental

General procedure for the preparation of 1-amidoalkyl naphthol derivatives

In a 25 mL round bottom flask, a mixture of 2-naphthol (1.0 mmol, 0.144 g), benzaldehyde (1.0 mmol, 0.106 g), benzamide (1.0 mmol, 0.121 g), and [CholineCl][$ZnCl_2$]$_3$ (20 mol%, 0.109 g) was heated at 100 °C for 40 min. The reaction times were monitored in Table 3 (checked by TLC). After completion, the reaction mixture was allowed to cool to room temperature. The reaction mixture was extracted with ethyl acetate (3 x 30 mL). The ethyl acetate layer was washed with distilled water (3 x 10 mL). The organic layer was dried over Na_2SO_4, and the solvent was recovered by a rotary evaporator. The desired product was purified by crystallization from hot ethanol. The structure and purity of the product were determined by ^{1}H, and ^{13}C NMR, FT-IR, or HRMS (ESI).

General procedure for the preparation of polyhydroquinoline derivatives

In a 25 mL round bottom flask, a mixture of benzaldehyde (1.0 mmol, 0.106 g), dimedone (1.0 mmol, 0.140 g), ethyl acetoacetate (1.0 mmol, 0.027 g), ammonium acetate (1.0 mmol, 0.077 g), and [CholineCl][$ZnCl_2$]$_3$ (0.5 mol%, 0.0027 g) was heated at 100 °C for 20 min. After completion (checked by TLC), the reaction mixture was allowed to cool to room temperature. Ethyl acetate was used to extract the reaction mixture (3 x 30 mL). The ethyl acetate layer was washed with distilled water (3 x 10 mL). The product was purified by crystallization from hot ethanol. The structure and purity of the product were determined by ^{1}H, and ^{13}C NMR, FT-IR, or HRMS (ESI).

Conclusions

In conclusion, we have successfully developed an effective method for the multicomponent synthesis of 1-amidoalkyl naphthols and

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polyhydroquinolines using [CholineCl][$ZnCl_2$]$_3$ as a Lewis acid catalyst. The process was accomplished under mild conditions, and the desired product was isolated easily through the recrystallization from hot ethanol. The prominent features of the present work are an inexpensive catalyst, mild condition, and work-up simplicity. The straightforward multicomponent synthesis of these compounds from available starting materials is more favorable than the two-step procedure in terms of eliminating the isolation and purification of precursors and saving energy, time, and chemicals. The plausible mechanism was proved by HRMS (ESI). The process can be applied to the industry to reduce costs and to prevent environmental pollution.

Acknowledgements

This research is funded by Vietnam National Foundation for Science and Technology Development (NAFOSTED) under grant number 104.01-2019.26.

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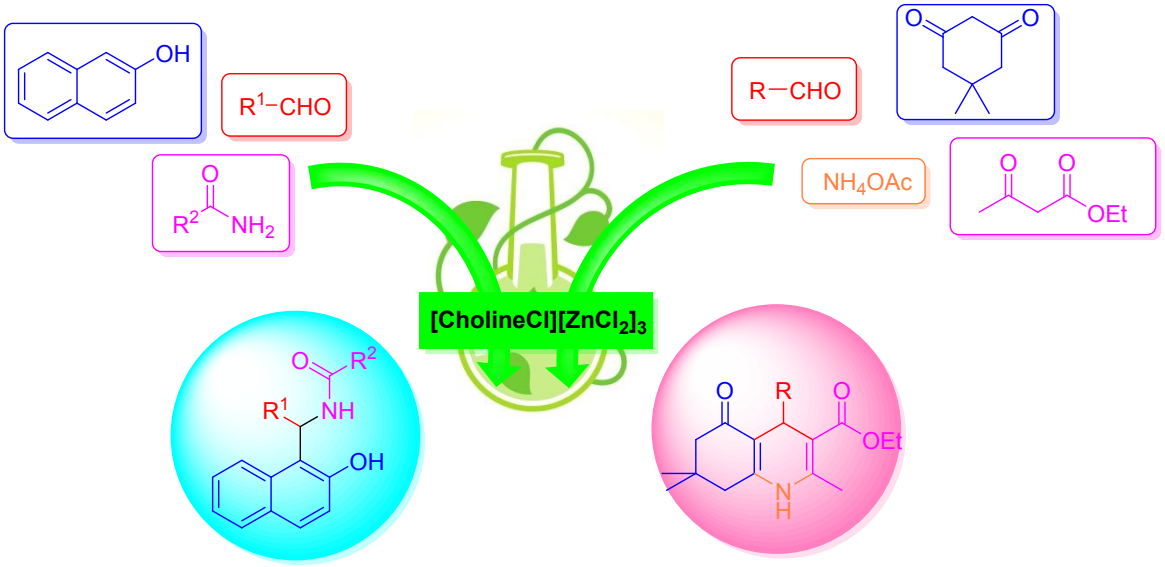

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