Current Chemistry Letters 13 (2024) 483–490

Contents lists available at GrowingScience

Current Chemistry Letters

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Ionic liquid mediated efficient synthesis of 2,4,5-triarylimidazoles via green economical multicomponent reaction

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C H R O N I C L E

Article history:
Received October 4, 2023
Received in revised form January 10, 2024
Accepted March 3, 2024
Available online March 5, 2024

Keywords:
2,4,5-triaryl imidazole
Benzil
1-butyl-3-methyl-imidazolium hexafluoro phosphate
multicomponent reaction

A B S T R A C T

In the present work, a new protocol was developed for the synthesis of 2,4,5-triaryl imidazoles via three component condensation of aryl aldehyde, benzil and ammonium acetate in the presence of 1-butyl-3-methyl-imidazolium hexafluoro phosphate ([BMIM][PF6]) as a catalyst under reflux in ethanol. The present protocol has many beneficial advantages such as excellent yields, easy workup procedure, green catalyst and purification of the targeted molecules without the use of column chromatography which increases the green chemistry value of the present work.

1. Introduction

Multicomponent reactions (MCRs) have great importance in organic synthesis due to the high functional group tolerance and have properties like high atom economy, synthetic efficiency, lower operational cost than multistep synthesis, and easy separation of the products.[1] The synthesis of 2,4,5-triarylimidazole is an important example of multicomponent condensation reactions.[2] The nitrogen containing heterocyclic compounds has great interest to organic chemists for many years due to the broad range of biological activities.[3-5] Among them the 2,4,5-triarylimidazole derivatives possess a crucial spectrum of biological activities like anti-epileptic,[6] anti-inflammatory,[7] inhibitors of P38 MAP kinase,[8] glucagon receptor antagonists,[9] antiviral,[10] and anti-cancer activities.[11] Imidazole is a versatile core which found in many naturally occurring compounds (**Fig.1**).

In the literature, various protocols have been reported for the synthesis of 2,4,5-triarylimidazole by three component condensation reaction of aldehydes, benzil and ammonium acetate in the presence of different catalytic materials such as L-proline,[12] $NiFe_2O_4$/geopolymer,[13] $(CTA)_3$PMo-MMT,[14] Fe_3O_4@PVA–SO_3H,[15] *p*-TSA,[16] chitosan-SO_3H,[17] caffeine-H_3PO_4,[18] citrate trisulfonic acid,[19] silica sulfuric acid,[20] $TiCl_4.SiO_2$,[21] $InCl_3.3H_2O$,[22] Pumice@SO_3H,[23] modified-silica-coated cobalt ferrite nanoparticles with tungstic acid,[24] LADES@MNP,[25] Fe‑DTPMP,[26] Cu_2O/Fe_3O_4@guarana,[27] $Zn(OAc)_2.2H_2O$,[28] trichloromelamine,[29] etc. Also, divergent five-membered heterocycles were synthesized using a universal and effective [3+2] cycloaddition reaction path under thermal condition.[30]

In the recent era, ionic liquids have revolutionized research which played a diversified role in the chemical science. They have been employed as a solvent as well as a catalyst in synthetic organic chemistry due to its significant chemical

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doi: 10.5267/j.ccl.2024.3.001

and physical properties like high thermal stability, lower vapor pressure, excellent solvating capability, enough liquid range, and good ionic conductivity. The diverse ionic liquids have been used for the synthesis of multi-substituted imidazole derivatives such as [BMIM][BF_4],[31] [H-NP][HSO_4],[32] [HNMP][HSO_4],[33] [Et_3NH][HSO_4],[34] [(4-SB)T(4-SPh)][$PHSO_4$],[35] [Hmim][HSO_4],[36] [2-(imm)-4-{b(immh)m}c][HSO_4]$_3$,[37] mesoporous organosilica supported benzotriazolium ionic liquid,[38] etc.

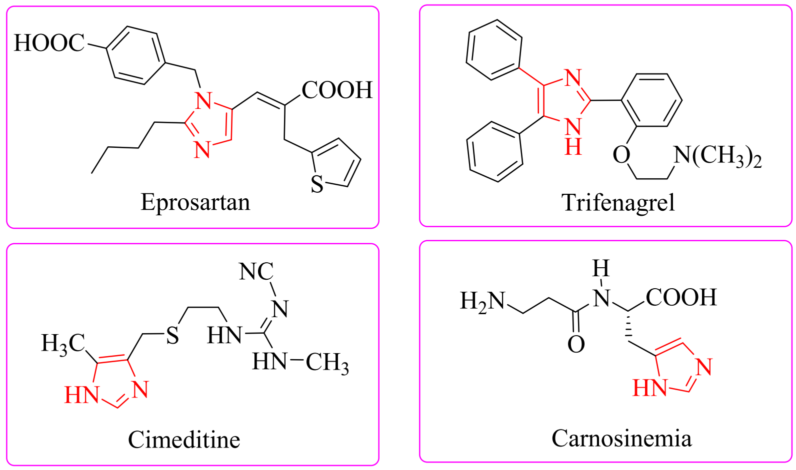
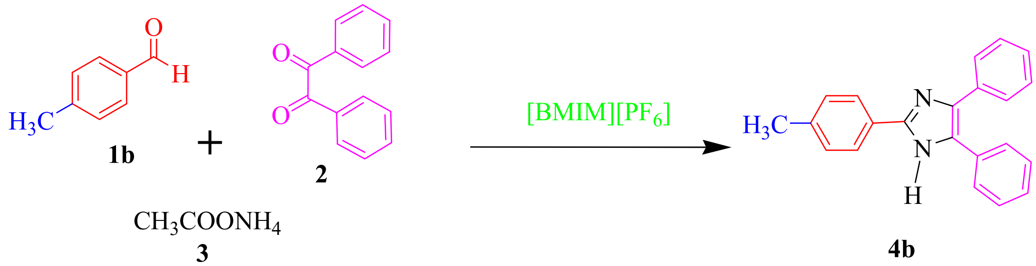


Fig.1. Biologically active drugs containing imidazole core

In consideration of some drawbacks and limitations in front of the researchers such as expensive catalyst, lower yields, lengthy and critical workup procedure etc., herein we report a newer eco-friendly protocol for the one-pot synthesis of 2,4,5-triarylimidazole from aryl aldehyde, benzil and ammonium acetate in the presence of green catalyst 1-butyl-3-methyl-imidazolium hexafluoro phosphate ([BMIM][PF_6]) under reflux condition in ethanol.

2. Result and Discussion

For the selection of optimized reaction conditions, we have chosen three component reactions of *p*-methyl benzaldehyde (**1b**), benzil (**2**), and ammonium acetate (**3**) as a pilot of reaction (**Scheme 1**).



Scheme 1. Pilot reaction for the synthesis of 4,5-diphenyl-2-*p*-tolyl-1*H*-imidazole (**4b**)

At first, we carried out the pilot reaction without catalyst with and without solvent at different temperatures as shown in **Table 1**. The reaction did not proceed to any extent of formation of targeted molecule, which supports the need of the catalyst in the synthesis of multi-substituted imidazoles. Then the reaction mixture was agitated at room temperature in the presence of an ionic liquid [BMIM][PF_6] under solvent-free conditions as well as in combination with solvent like ethanol. But at room temperature, the reaction did not proceed to any extent. Hence, the model reaction was carried out under reflux condition in ethanol. The desired product of the pilot reaction was obtained in 2.5hrs with excellent yield (**Table 1, Entry 7**).

Table 1. Optimization of reaction conditions for the synthesis of 4,5-diphenyl-2-*p*-tolyl-1*H*-imidazole (**4b**)

Entry	Condition	Temperature	Time (min)	Yield (%)
1	Catalyst free / Solvent free	Grinding	30	NR
2	Catalyst free / SF	Heating at 100ºC	30	NR
3	Catalyst free / EtOH	Stirring at RT	60	NR
4	Catalyst free / EtOH	Reflux	60	NR
5	90 mg [BMIM][PF_6] / SF	Grinding	60	NR
6	90 mg [BMIM][PF_6] / EtOH	Stirring at RT	60	NR
7	90 mg [BMIM][PF_6] / EtOH	Reflux	120	96

Reaction condition: **1b** (1mmol), **2** (1mmol), **3** (2mmol), [BMIM][PF_6] (90 mg)

The next step after selection of the optimized condition, the pilot reaction was carried out by changing the amount of catalyst to increase the yield and decrease the time of the reaction (**Table 2**). The best result was obtained with an optimal amount of 90 mg of the catalyst (**Table 2, Entry 4**). The results are summarized in **Table 2**.

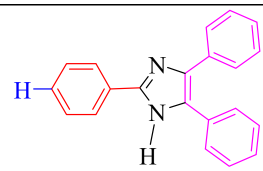
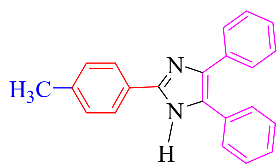
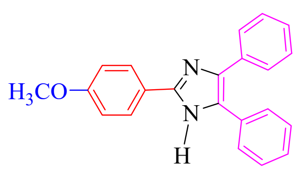
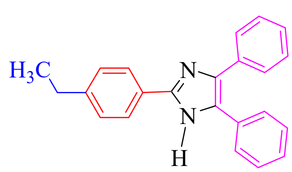
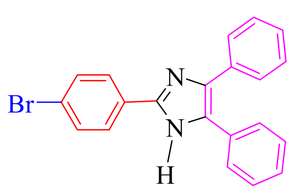
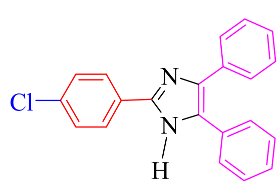
Table 2. Influence of the amount of [BMIM][PF_6] on the synthesis of 4,5-diphenyl-2-*p*-tolyl-1*H*-imidazole (**4b**)

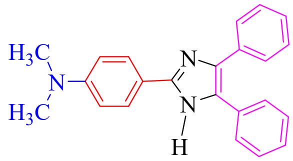
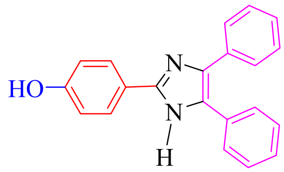
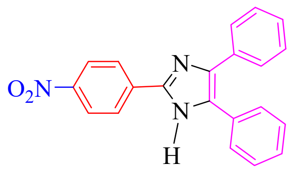
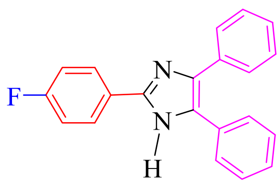
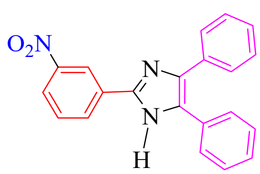
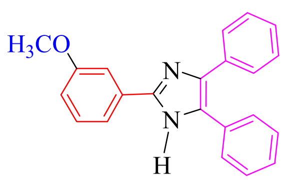
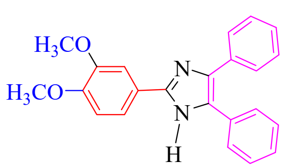
Entry	Amount of Catalyst	Condition	Time (min)	Yield (%)
1	No catalyst/ EtOH	Reflux	60	NR
2	50 mg [BMIM][PF_6] / EtOH	Reflux	120	40
3	75 mg [BMIM][PF_6] / EtOH	Reflux	120	80
4	90 mg [BMIM][PF_6] / EtOH	Reflux	120	96
5	125 mg [BMIM][PF_6] / EtOH	Reflux	120	96

Reaction condition: **1b** (1mmol), **2** (1mmol), **3** (2mmol), Reflux in EtOH

With the standardized condition, the present catalytic protocol worked very well for the synthesis of 2,4.5-triaryl imidazole analogues (**Table 3, Entry 4a-m**) from the aryl aldehydes containing electron donating groups like –CH_3, -CH_2-CH_3, -OH, -OCH_3, -$N(CH_3)_2$ as well as electron withdrawing groups like -F, -Cl, -Br, -NO_2. The excellent results obtained with the current protocol are summarized in **Table 3**. All obtained products were separated by simple filtration and were purified by the simple recrystalization method. While the formation of products was confirmed by analytical spectroscopic methods such as FT-IR, ^{1}H-NMR, ^{13}C-NMR, and Mass and compared with the data available in the literature.

Table 3. Synthesis of 2,4,5-triaryl imidazole derivatives (**4a-m**)

Entry	Imidazole derivative	Color	Time (hrs.)	Yield (%)	M.P. (°C)	
					Found	Lit. (Ref.)
4a		White	2.5	94	272-274	273-276[24]
4b		White	2	96	228-230	226-229[15]
4c		White	2	96	226-228	228-231[24]
4d		White	2.5	92	230-232	230-232[23]
4e		White	2	95	260-262	260-264[24]
4f		Off-white	2.5	90	258-260	259-262[24]

4g		Faint Yellow	2	90	250-252	255-257[15]
4h		White	2.5	90	230-232	231-234[24]
4i		Brown	2.5	92	236-238	232-234[24]
4j		White	2	90	186-188	186-189[24]
4k		Brown	2.5	90	262-266	269-271[24]
4l		White	2	94	258-260	259-260[27]
4m		White	2	90	218-220	220-222[15]

Reaction condition: **1** (1mmol), **2** (1mmol), **3** (2mmol), [BMIM][PF_6] (90 mg), Reflux in EtOH

In order to further authenticate our work, the results obtained in the present work were compared with the literature reported work based on the catalysts, condition, time and yields as summarized in **Table 4**.

Table 4. Comparison of [BMIM][PF_6] catalyzed protocol with other literature reported protocols in the synthesis of Tri-aryl imidazoles

Entry	Catalyst	Condition	Time (min)	Yield (%)	References
1	50 mg Fe_3O_4@PVA-SO_3H	Reflux in EtOH	35-70	75-91	15
2	100 mg Pumice@SO_3H	MW irradiation at 280W	10-15	80-92	23
3	100 mg $CoFe_2O_4$@SiO_2@ (–CH_2)$_3$OWO$_3$H	SF Heating at 110ºC	15-50	72-92	24
4	10 mol % $Zn(OAc)_2.2H_2O$	SF Heating at 70ºC	60-180	74-92	28
5	70 mg Trichloromelamine	SF Heating at 110ºC	60-420	86-94	29
6	90 mg [BMIM][PF_6]	Reflux in EtOH	120-150	90-96	Present work

3. Conclusions

The present work describes the recent development on the synthesis of 2,4,5-triarylimidazoles via three component condensation of aryl aldehyde, benzil and ammonium acetate in the presence of [BMIM][PF_6] as a greener catalyst under reflux condition in ethanol. The multisubstituted imidazole derivatives were isolated in good to excellent yields in optimum time. The present protocol has many beneficial advantages such as excellent yields, easy workup procedure, green catalyst and purification of the targeted molecules without the use of column chromatography technique which reaches the goal and increases the value of green chemistry. Also, this work discussed the current status of ionic liquids as a homogeneous catalytic materials and their efficiency in one-pot synthesis of 2,4,5-triaryl-imidazole derivatives.

Acknowledgement

We are very much thankful to the Yogeshwari Mahavidyalaya, Ambajogai, Beed, Maharashtra (India) for providing infrastructural facilities. We are also thankful to SAIF, Punjab University, Chandigarh for providing the good characterization.

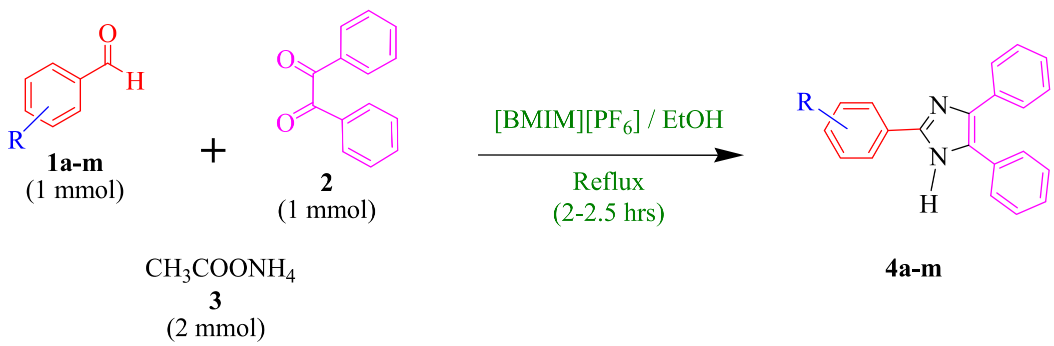
4. Experimental

4.1. Materials and methods

Physical constants were taken in an open capillary and are uncorrected. The PMR and CMR spectra were recorded on a Brucker Avance II 500MHz in $CDCl_3$ using TMS as an internal standard. Mass spectra were recorded on a Finnigan Mass spectrometer. The progress of the reaction was monitored by thin-layer chromatography (TLC) using Aluminum plates pre-coated with silica gel and visualized under UV light.

*4.2. General procedure for the synthesis of 2,4,5-triaryl imidazoles (**4a-m**)*

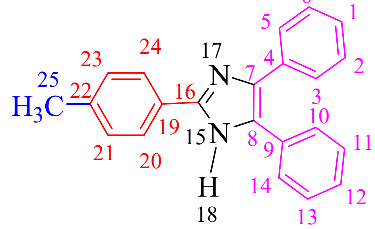
A mixture of aryl aldehyde **1** (1 mmol), benzil **2** (1 mmol), ammonium acetate **3** (2 mmol) and 90 mg [BMIM][PF_6] catalyst was taken in a 100 mL round bottom flask containing 10ml ethanol (**Scheme 2**). The resulting reaction mixture was refluxed for 2-2.5hrs as mentioned in **Table 3**. The progress of the reaction was monitored by TLC which carried out in mixture of n-hexane and ethyl acetate (4:1) as a mobile phase. Once the reaction was completed, the reaction mixture was poured over 100 gm crushed ice. The solid product was separate out which were filtered on suction pump and dried under the IR lamp. The obtained solid product was purified by recrystallization using ethanol as a solvent.



Scheme 2. Synthesis of 2,4,5-triaryl imidazoles (**4a-m**)

4.3. Physical and Spectral data of 2,4,5-trisubstituted imidazole derivatives

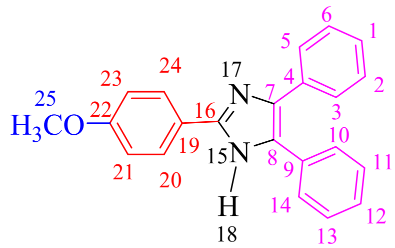
*4,5-diphenyl-2-p-tolyl-1H-imidazole (**4b**):*



Yield 96%; m.p. 228-230°C; ^{1}H NMR (500 MHz, $CDCl_3$) δ (ppm): 2.34 (s, 3H, Ar-CH_3), 7.17 (d, J= 7.3Hz, 2H, Ar-H), 7.25-7.29 (m, 6H, Ar-H), 7.48 (d, J= 5.6Hz, 4H, Ar-H), 7.75 (d, J= 6.7Hz, 2H, Ar-H); ^{13}C NMR (500 MHz, $CDCl_3$) δ (ppm);

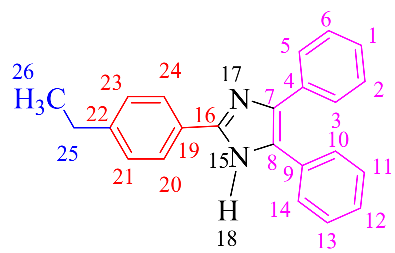
21.47 (C_{25}), 125.39 (C_{20}, C_{24}), 126.88 (C_3, C_5, C_{10}, C_{14}), 127.36 (C_{19}), 127.89 (C_1, C_{12}), 128.49 (C_2, C_6, C_{11}, C_{13}), 129.52 (C_7, C_8), 132.65 (C_{21}, C_{23}), 138.85 (C_4, C_9), 146.31 (C_{22}), 175.16 (C_{16}); MS (ESI):m/z = 311.1889 [M+H]

*2-(4-methoxyphenyl)-4,5-diphenyl-1H-imidazole (**4c**):*



Yield 96%; m.p. 226-228°C; ^{1}H NMR (500 MHz, $CDCl_3$) δ (ppm): 3.82 (s, 3H, Ar-CH_3), 6.91 (d, J= 8.7Hz, 2H, Ar-H), 7.23-7.30 (m, 6H, Ar-H), 7.48-7.51 (m, 4H, Ar-H), 7.82 (d, J= 8.6Hz, 2H, Ar-H); ^{13}C NMR (500 MHz, $CDCl_3$) δ (ppm); 55.36 (C_{25}), 114.26 (C_{21}, C_{23}), 122.16 (C_{19}), 127.06 (C_3, C_5, C_{10}, C_{14}), 127.42 (C_{20}, C_{24}), 127.86 (C_1, C_{12}), 128.53 (C_2, C_6, C_{11}, C_{13}), 129.03 (C_7), 129.92(C_8), 132.45 (C_4), 134.91(C_9), 146.06 (C_{16}), 160.32 (C_{22}); MS (ESI):m/z = 327.1853 [M+H]

*2-(4-ethylphenyl)-4,5-diphenyl-1H-imidazole (**4d**):*



Yield 92%; m.p. 230-232°C; ^{1}H NMR (500 MHz, $CDCl_3$) δ (ppm): 1.25 (t, J= 7.5Hz, 3H, -CH_3), 2.68 (q, J= 7.5Hz, 2H, -CH_2), 7.24-7.28 (m, 4H, Ar-H), 7.30-7.32 (m, 4H, Ar-H), 7.51 (d, J= 7.0Hz, 4H, Ar-H), 7.82 (d, J= 8.2Hz, 2H, Ar-H); ^{13}C NMR (500 MHz, $CDCl_3$) δ (ppm); 15.35 (C_{26}), 28.71 (C_{25}), 125.51 (C_{20}, C_{24}), 126.86 (C_3, C_5, C_{10}, C_{14}), 127.48 (C_{19}), 127.88 (C_{21}, C_{23}), 128.38 (C_1, C_{12}), 128.56 (C_2, C_6, C_7, C_8, C_{11}, C_{13}), 132.55 (C_4, C_9), 145.41 (C_{22}), 146.11 (C_{16}); MS (ESI):m/z = 325.1846 [M+H]

*2-(4-bromophenyl)-4,5-diphenyl-1H-imidazole (**4e**):*

Yield 95%; m.p. 260-262°C; ^{1}H NMR (500 MHz, $CDCl_3$) δ (ppm): 7.26-7.32 (m, 6H, Ar-H), 7.49-7.53 (m, 6H, Ar–H), 7.79 (d, J= 8.5Hz, 2H, Ar–H);

*2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazole (**4f**):*

Yield 90%; m.p. 258-260°C; ^{1}H NMR (500 MHz, $CDCl_3$) δ (ppm): 7.26 (m, 2H, Ar-H), 7.31-7.34 (m, 4H, Ar–H), 7.42 (m, 2H, Ar–H), 7.50 (d, J= 7.2Hz, 4H, Ar–H), 7.81 (m, J= 8Hz, 2H, Ar–H); MS (ESI):m/z = 331.1363 [M+H].

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