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Copper borate (CuB_4O_7)-promoted multi-component green synthesis of 2,4,5-triarylimidazole derivatives and evidence of *in situ* conversion of copper borate (CuB_4O_7) into $Cu(OAc)_2$ in the presence of NH_4OAc†

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A green, efficient, and straightforward methodology for the three-component synthesis of 2,4,5-triarylimidazole has been developed under solvent-free conditions using unconventional CuB_4O_7 as a promoter. This green approach encouragingly provides access to a library of 2,4,5-tri-arylimidazole. Also, we have been able to isolate the compound (**5**) and (**6**) *in situ*, which provides an insight into the direct conversion of CuB_4O_7 into copper acetate in the presence of NH_4OAc under solvent-free condition. The main advantage of this protocol includes an easy reaction procedure, short reaction time, and easy work up of the product without using any tedious separation method.

Introduction

Of late, the design and development of reaction methodology in an environmentally benign, greener, and sustainable way to reduce the use of hazardous chemicals and to explore the alternative reaction conditions or reaction media to bring about the desired chemical transformations by minimizing the byproducts or waste and the exclusion of the conventional organic solvents has gained considerable interest in the field of organic transformation reactions.[1–10] Fortunately, multi-component reactions (MCRs) serve as good example to meet the requirements for the green chemical processes and it may be viewed as an art of performing efficient chemical transformation by coupling three or more reactants in a single pot in single operation, avoiding toxic reagents, solvents, and costly purification techniques. MCR also has an advantage in the development and formulation of novel and cost-effective approaches in synthetic chemistry research, leading to environmental protection and have emerged as powerful tools for drug discovery.[11–19] The MCR technique has gained special attention and drawn considerable interest in modern synthetic chemistry and pharmaceutical chemistry owing to its advantages, such as high atom-economy, high selectivity, operational simplicity, structural diversity, short reaction time, low cost, low energy consumption, high yield, and easy purification process.[20–24] The imidazole scaffold belongs to an important heterocyclic compound due to their richness in various natural products and their widespread use in medicinal fields.[25,26] Imidazole ring system possesses a variety of pharmaceutical properties and plays a fundamental role in numerous biochemical processes.[27] Over the past few years, highly substituted imidazoles have found a special place owing to their biological relevance in many natural products, extensive applications in the area of material science,[28] and potential use as herbicide,[29] fungicide,[30] inhibitors of P38 MAP kinase,[31] B-Raf kinase,[32] antiinflammatory,[33] antitumor,[34] and antithrombotic[35] agents. In addition, some of the highly substituted imidazole compounds serve as a photosensitive material in photography.[36] Clemizole (antihistaminic agent),[37] nocodazole (antinematodal),[38] azathioprine (antirheumatoid arthritis),[39] and dacarbazine (anticancer)[40,41] are commercially available

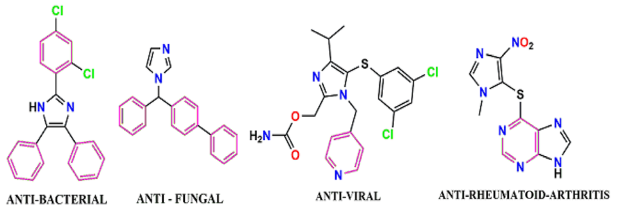

Fig. 1 Representative example of some commercially available drugs containing the imidazole core.

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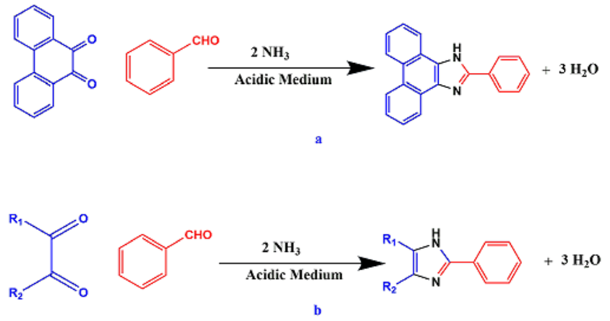
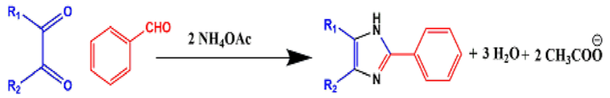


Scheme 1 (a) Japp imidazole synthesis and (b) Radziszewski imidazole synthesis.



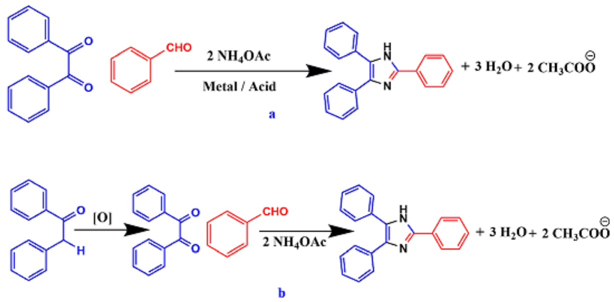
Scheme 2 Synthesis of 2,4,5-triarylimidazole using NH_4OAc as nitrogen source.

drugs that contain the imidazole scaffold and have been used for the treatment of different diseases (Fig. 1).

Japp and Radziszewski independently reported the first synthesis of 2,4,5-triarylimidazole in 1882 by the cyclocondensation of 1,2-dicarbonyl compound with an aldehyde in the presence of ammonia as a nitrogen source in acidic medium[42,43] (Scheme 1a and b).

After the successful synthesis of triarylimidazoles by Radziszewski and Japp, various synthetic modifications have been made in this approach. The most common modification that has been made is the use of ammonium acetate in place of ammonia as a nitrogen source[44] (Scheme 2).

In view of the varied properties and applications of imidazole core, numerous synthetic methodologies have been formulated using various catalytic systems under different reaction conditions for the efficient and rapid synthesis of substituted imidazole derivatives by the three-component cyclocondensation of a 1,2-diketone, α-hydroxyketone, or α-ketomonoxime with an aldehyde and ammonium acetate[45–57] (Scheme 3a). In addition to the use of conventional 1,2-diketone, α-hydroxyketone or α-ketomonoxime, α-methylene ketone has also been used for the synthesis of substituted imidazoles[58,59] (Scheme 3b).



Scheme 3 (a) Triarylimidazole synthesis from cyclocondensation of 1,2-diketone, aldehyde and ammonium acetate and (b) synthesis of tri-substituted imidazole using α-hydroxyketone, aldehyde, and ammonium acetate.

Although these documented procedures offer several beneficial features, yet most of them have serious drawbacks such as the use of expensive catalyst, lower yield, use of toxic solvent, harsh reaction condition, longer reaction time, and tedious work-up procedures.[60–63] Interestingly, there are several reports on the synthesis of 2,4,5-tri-aryl imidazole using different copper salts as catalyst,[64–69] but there are no reports on the study of catalytic activities of copper borate salt so far.

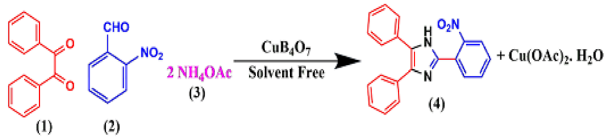
All these facts have encouraged us to find an ecofriendly, environmentally benign, greener, and sustainable method for the synthesis of substituted imidazole and stimulated us to employ copper(II) borate (CuB_4O_7) for the one-pot three-component synthesis of 2,4,5 triarylimidazole derivatives by the condensation of benzil, ammonium acetate, and aromatic/heteroaromatic aldehydes in solvent-free condition.

Results and discussion

Synthesis of 2,4,5 triarylimidazole derivatives

To ascertain the optimized reaction conditions for the designed protocol, benzil (**1**), 2-nitrobenzaldehyde (**2**), and NH_4OAc (**3**) were chosen as the model reactants (Scheme 4).

We investigated the model reaction with various Cu(II) salts including copper borate, and the results are listed in Table 1. We found that product **4** is formed in 60% yield after 120 min in ethanol at 80 °C using $CuSO_4$ is as a catalyst (Table 1, entry 1). Furthermore, we extended the model reaction with other copper salts such as CuO, $CuCl_2$, $CuBr_2$, $Cu(OAc)_2$, and CuB_4O_7 at 80 °C in ethanol medium and obtained 64%, 70%, 71%, 68%, and 86% yields of product **4**, respectively (Table 1, entries 2–6). Thus, from the optimization reaction, we obtained the maximum yield of product **4** in the presence of copper borate.



Scheme 4 Synthetic route for the preparation of 2,4,5-triarylimidazoles.

Table 1 Optimization of the catalyst for the synthesis of 2,4,5-triarylimidazole

Entry	Catalyst[a]	Yield[b] (%)
1	$CuSO_4$	60
2	CuO	64
3	$CuCl_2$	68
4	$CuBr_2$	67
5	$Cu(OAc)_2$	71
6	CuB_4O_7	84

[a] The reaction was performed with benzil **1** (1 mmol), 2-nitrobenzaldehyde **2** (1 mmol), and NH_4OAc **3** in the presence of different copper(II) salts in ethanol at 80 °C for 2 h. [b] Isolated yield.

Table 2 Optimization of catalyst loading for the synthesis of 2,4,5-triarylimidazole

Entry	Catalyst[a] (mol%)	Yield[b] (%)
1	0	12
2	0.5	24
3	1.0	42
4	1.5	70
5	**2.0**	**92**
6	2.5	85
7	3.0	80

[a] The reaction was performed with benzil **1** (1 mmol), 2-nitrobenzaldehyde **2** (1 mmol), and NH_4OAc **3** in the presence of different copper(II) salts in ethanol at 80 °C for 2 h. [b] Isolated yield.

We further investigated the amount of catalyst loading for the designed reaction and set up the model reaction employing different mol% (0, 0.5, 1.0, 1.5, 2.0, 2.5, and 3.0) of copper borate (Table 2). Interestingly, we observed that the reaction goes to completion when the amount of copper borate is 2 mol% at 80 °C (92%, Table 2, entry 5). Also, we carried out the same reaction in the absence of copper borate and observed that only a minimum amount (12%) of product **4** was formed (Table 2 entry 1).

We screened the catalytic efficiency of the catalyst with the model reactants in different solvents, and the result is summarized in Table 3. We found that in more polar solvents such as alcohol and DMF, the yield of the products were good (Table 3, entries 1–3) but the reaction needed more time (2 h) to achieve completion and required a tedious technique for the recovery of the product. Again, by considering the green chemistry principles, we were interested in employing water as a green solvent for the designed reaction, but in this case, we obtained only 32% yield of the product in the stipulated time (Table 3, entry 5). Due to this drawback, we focused our study on solid phase synthesis using silica gel as a reaction medium, and 91% yield of the product **4** was obtained; however, in this case also, we encountered problems during the work-up of the desired product **4** (Table 3, entry 6).

Therefore, these drawbacks prompted us to carry out the desired reaction in neat conditions without using any organic solvents. Encouragingly, the desired product 2,4,5-triarylimidazole (**4**) was obtained in excellent yield in less time (8–10 min) at 80 °C under solvent-free condition (Table 3, entry 7). The green chemistry metric *E*-factor for the designed reaction has been included in the ESI.†

Table 3 Screening of solvents for the synthesis of 2,4,5-triarylimidazole using CuB_4O_7

Entry	Catalyst[a]	Solvent[b]	Yield[c] (%)
1	CuB_4O_7	Methanol	92
2		Ethanol	88
3		DMF	86
4		DMSO	82
5		Water	32
6		Silica gel	91
7		**Neat**	**98**

[a] The reaction was performed with benzil **1** (1 mmol), 2-nitrobenzaldehyde **2** (1 mmol), and NH_4OAc **3** in the presence of different copper(II) salts in different solvent/neat conditions at 80 °C for 2 h except for neat condition. [b] Under neat condition, the reaction completed in 10 min. [c] Isolated yield.

Also, we observed that the yield of the product is maximum when the amount of NH_4OAc is 2.5 mmol with respect to other substrate, and with the decrease or increase in the amount of NH_4OAc, the yield of product **4** did not changed significantly.

Moreover, having recognized the optimized reaction conditions, we subsequently examined the catalytic efficiency and applicability of this protocol by extending this protocol to other aromatic/heteroaromatic aldehydes to prepare 2,4,5-triarylimidazole derivatives (**4a–4z**) (Fig. 2). We observed that all the reactions proceeded smoothly in a very short reaction time to afford the desired 2,4,5-triarylimidazoles in good to excellent yields (88–98%).

It was observed that *para*-substituted benzaldehyde with electron withdrawing group successfully afforded the corresponding 2,4,5-triarylimidazoles in excellent yield under optimized reaction condition compared to the *para*-substituted benzaldehyde with electron-releasing group. Other aromatic/heteroaromatic aldehydes also afforded the corresponding imidazoles in good yield but they required more time for completion (15–20) min. All the synthesized compounds (**4a–4z**) were recrystallized from ethanol (10 mL), characterized by their analytical (melting point) and spectroscopic data (FT-IR and ^{1}H NMR), and compared with the literature value. The analytical results (yield and melting point) and the spectroscopic details (FT-IR and ^{1}H NMR) data of the synthesized compounds are given in the Experimental section, and the corresponding spectra are provided in ESI 1.†

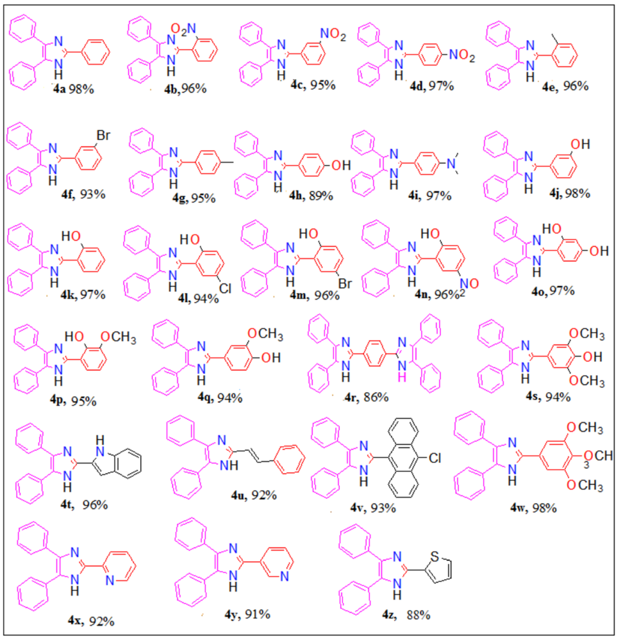


Fig. 2 Substrate scope for the synthesis of 2,4,5-triarylimidazole (**4a–z**).

Table 4 Comparison of catalytic efficacy of CuB_4O_7 with other reported catalysts

Entry	Catalyst	Solvent	Temperature	Time	Yield (%)
1	L-Proline	MeOH	60 °C	9 h	90 [lit. 70]
2	Zeolite ZSM-11	Neat	Reflux	60 min	80 [lit. 71]
3	$Zr(acac)_4$	EtOH	Reflux	2.5 h	90 [lit. 72]
4	$Yb(OTf)_3$	OHAc	70 °C	2 h	92 [lit. 73]
5	ZnO nanorod	H_2O	Reflux	1.45 h	83 [lit. 74]
6	I_2	Neat	RT	15 min	90 [lit. 75]
7	Fe_3O_4 nanoparticles	Neat	80 °C	20 min	96 [lit. 76]
8	Nano $TiCl_4 \cdot SiO_2$	Neat	110 °C	1 h	90 [lit. 77]
9	Nano MgO	Neat	100 °C	0.6 h	94 [lit. 78]
10	InF_3	Neat	60 °C	0.3 h	90 [lit. 79]
11	$CoFe_2O_4$ NPs	EtOH	40 °C	20 min	95 [lit. 80]
12	$Y(TFA)_3$	Neat	100 °C	3 h	97 [lit. 81]
13	Boric acid	H_2O	Ultrasound	30 min	98 [lit. 82]
14	ZSM-5–SO_3H	Neat	110 °C	90 min	90 [lit. 83]
15	Trichloromelamine	Neat	110 °C	1 h	92 [lit. 84]
16	TiO_2	Neat	120 °C	2.5 h	92 [lit. 85]
17	$H_2SO_4 \cdot SiO_2$	Neat	110 °C	60 min	94 [lit. 86]
18	Copper nanoparticles	Neat	RT	60 min	94 [lit. 87]
19	HMDS	Neat	120 °C	3 h	96 [lit. 88]
20	$KMnO_4/CuSO_4$	EtOH	Reflux	70 min	85 [lit. 89]
21	SiO_2–$NaHSO_4$	Neat	120 °C	30 min	95 [lit. 90]
22	Zeolite HY/silica gel	Neat	MW	6 min	89 [lit. 91]
23	**CuB_4O_7**	**Neat**	**80 °C**	**10 min**	**98 [this work]**

Comparisons of efficacy of CuB_4O_7 with those of the reported catalysts[70–91] for the synthesis of 2,4,5-trisubstituted imidazole have shown that the present system involving CuB_4O_7 has proven to be more efficient in terms of the short reaction time, milder reaction condition, and product yields (Table 4).

Since copper borate was insoluble in most of the solvents and we carried out the reaction under neat condition, therefore, we were hopeful of recycling copper borate in fair amount. Surprisingly, we found that during the addition of copper borate to the reaction mixture, the reaction mixture turned blue and after the work-up of the product, unfortunately, the desired copper borate could not be recovered. Although it efficiently afforded the desired product **4** in good to excellent yield, yet still there were some doubts due to the nonrecyclability of copper borate.

Fortunately, when 2-OH-benzaldehyde derivatives were employed as an aldehyde substrate (**2**), we found the formation of some blue colored products in the reaction mixture along with the desired product **4**. We carefully recrystallized the blue colored compound in *N*,*N*-dimethylformamide (DMF) solvent (2 mL) and after 5 days, a blue colored crystal suitable for X-ray single crystal diffraction was isolated from the reaction, where 2-OH-5-NO_2-benzaldehyde was used as an aldehyde substrate. After successful X-ray single crystal diffraction study, it was revealed that the desired 2,4,5-triarylimidazole product was formed during the reaction but the product itself acted as a bidentate chelating ligand for the copper(II) ion in the reaction mixture, thereby forming bis[2-(4,5-diphenyl-1*H*-imizol-2-yl)-4-nitro-phenolato]copper(II) dihydrate complex with molecular formula $[Cu(C_{21}H_{14}N_3O_3)_2] \cdot 2H_2O$ (**5**) (CCDC no: 1958158)[92] (Fig. 3a and b).

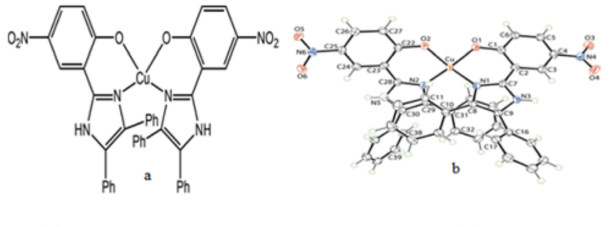
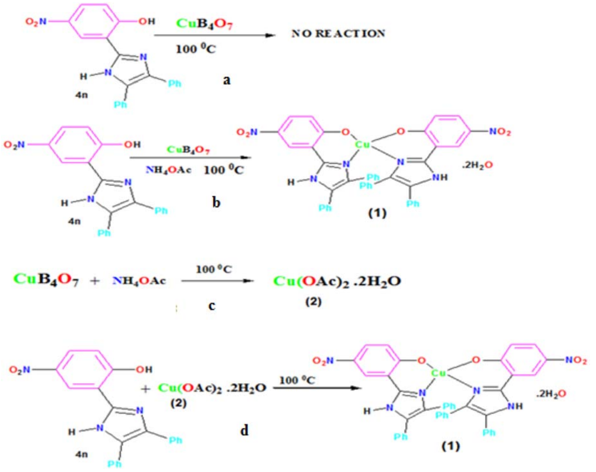


Fig. 3 (a) Molecular structure of compound **5** and (b) crystal structure of compound **5** with displacement ellipsoid drawn at 70% probability level.

Therefore, we carried out the detailed study of transformation of copper borate into other intermediate in the reaction mixture under the given conditions separately.

Firstly, we synthesized 2-(4,5-diphenyl-1*H*-imidazol-2-yl)-4-nitrophenol (**4m**) ligand and subsequently reacted with CuB_4O_7 in neat condition at 100 °C for 8–10 h; however, we failed to get the desired product (**5**) (Scheme 5a). But when we added stoichiometric amount of NH_4OAc (**3**) to the same reaction, the reaction mixture instantaneously turned blue and after reacting for 4 h, the compound was extracted with DMF (5 mL) and the filtrate was kept for 5–8 days for slow evaporation at room temperature. Surprisingly, we got the same blue colored copper(II) complex (**5**) in excellent yield (60%) (Scheme 5b).

Secondly, to understand the role of NH_4OAc, a separate reaction was set up in which 1 mmol CuB_4O_7 was directly added to 50 mL water containing 3 mmol NH_4OAc in a 100 mL round bottomed flask (Scheme 5c), and the reaction mixture was allowed to stir at room temperature for 8 h. It was seen that CuB_4O_7 slowly discharged into the NH_4OAc solution and with the passage of time, the entire CuB_4O_7 dissolved in the reaction



Scheme 5 (a) Reaction between ligand **4n** and CuB_4O_7, (b) reaction between ligand **4n** and CuB_4O_7 in the presence of NH_4OAc for the synthesis of **5**, (c) reaction between CuB_4O_7 and NH_4OAc to form compound **6**, and (d) reaction between ligand **4n** and compound **6** to form **5**.

mixture. The reaction mixture was allowed to evaporate at room temperature and finally after 14 days, a blue colored single crystal suitable for X-ray single crystal diffraction study appeared in the solution. After successful X-ray single crystal diffraction of the compound, it was found that the blue colored complex was copper(II) acetate monohydrate (**6**) (molecular formula $Cu_2C_8H_{16}O_{10}$) with monoclinic crystal system and ($C2/c$) space group having paddle wheel structure (Fig. 4a–c).

The crystallographic description, selected bond lengths bond distances, and hydrogen bonded geometry of the compound (**6**) are given in the ESI 1.†

Thus, it IS evident from the above reaction that CuB_4O_7 in the presence of NH_4OAc is converted to intermediate $Cu(OAc)_2$ monohydrate (**6**) in the reaction mixture and undergoes metallation with ligand (**4n**) to give the complex **5** under the given reaction conditions. We again set up a reaction directly with compound **4n** employing $Cu(OAc)_2$ monohydrate **6** under the same reaction conditions, and we obtained the same compound **5** (Scheme 5d).

This finding encouraged us to understand the reaction of NH_4OAc with other copper salts, and we set up a reaction between different copper(II) salts such as $CuSO_4$, $CuCl_2$, and CuO to check the conversion of these salts into $Cu(OAc)_2$ monohydrate under the same reaction. Fascinatingly, it was found that these copper salts are readily converted to $Cu(OAc)_2$ monohydrate **6**, as evident by the comparisons of the FT-IR spectra of the respective reaction with commercially available $Cu(OAc)_2$ monohydrate (Fig. 5).

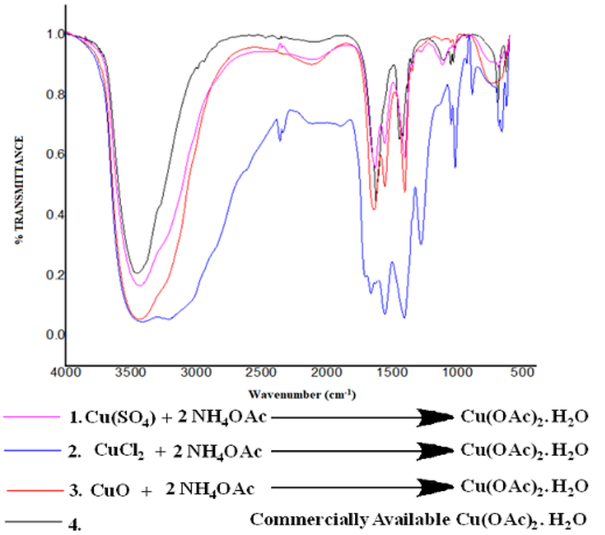


Fig. 5 (1) FT-IR spectrum of the reaction mixture of $CuSO_4$ and NH_4OAc, (2) FT-IR spectrum of the reaction mixture of $CuCl_2$ and NH_4OAc, (3) FT-IR spectrum of the reaction mixture of CuO and NH_4OAc, and (4) FT-IR spectrum of commercially available $Cu(OAc)_2 \cdot H_2O$.

Furthermore, we also investigated the catalytic activity of $Cu(OAc)_2$ monohydrate for the designed reaction under solvent-free condition, but it was observed that the desired product formed in less amount (72%), and the reaction takes a longer time to reach completion.[58] Thus, we can infer that copper borate helps to ionize NH_4OAc more efficiently, which acts as a nitrogen source for the schemed reaction to give the desired product (Table 5, entry 2).

Moreover, a plausible mechanism for the formation of 2,4,5-triarylimidazole **4** by the condensation reaction between benzil **1**, aldehyde **2** and ammonium acetate **3** catalyzed by copper borate (CuB_4O_7) is described in Scheme 4. Different pathways have been reported in the literature for the synthesis of 2,4,5-triarylimidazole using benzil as the starting material.[44,93] In our proposed mechanism, it was hypothesized that copper borate reacts with ammonium acetate to generate ammonia and copper acetate **5** *in situ* at the initial stage of the reaction.

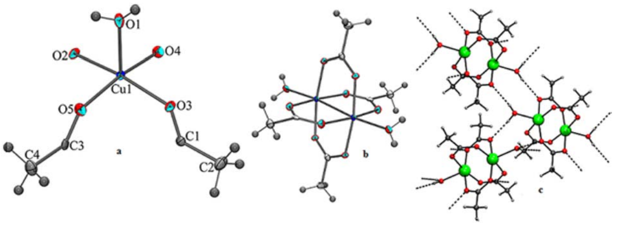
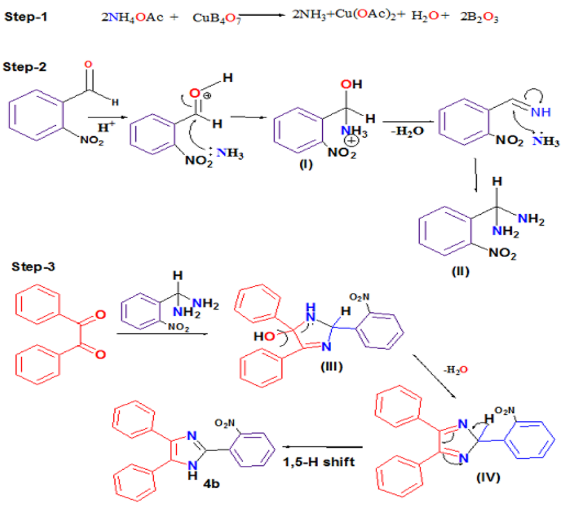


Fig. 4 (a) Asymmetric unit of compound **6**, (b) ortep diagram of **6** with 50% probability and (c) H-bonding pattern of compound **6**.

Table 5 Comparison of efficacy of CuB_4O_7 and $Cu(OAc)_2 \cdot H_2O$

Entry	Catalyst	Solvent[a]	Temperature	Time (min)	Yield[b] (%)
1	**CuB_4O_7**	**Neat**	**80 °C**	**10**	**98**
2	$Cu(OAc)_2 \cdot H_2O$	Neat	80 °C	120	72

[a] The reaction was performed with, benzil **1** (1 mmol), 2-nitrobenzaldehyde **2** (1 mmol), and NH_4OAc (**3**) in the presence of CuB_4O_7 and $Cu(OAc)_2$. H_2O under solvent-free condition. [b] Isolated yield.



Scheme 6 Proposed mechanism for the synthesis of 2,4,5-triarylimidazole using CuB_4O_7.

Furthermore, the generated ammonia reacts with 2-nitrobenzaldehyde to produce intermediate **I** and, subsequently, intermediate **I** adds a second molecule of ammonia to give intermediate (2-nitrophenyl)methanediamine (**II**). Intermediate **II** reacts with benzil to afford intermediate **III**, and the loss of water molecule from intermediate **III** affords intermediate **IV**. At the final stage of the reaction, intermediate **IV** undergoes 1,5-H shift to form the desired product **4b** (Scheme 6).

Moreover, CuB_4O_7 under the schemed protocol could not be recovered after the reaction since it gets transformed into $Cu(CH_3COO)_2 \cdot H_2O$. Thus, we repeated the same reaction with fresh CuB_4O_7 to ensure the repeatability of the desired product, and it was observed that the product can be reproduced under the given reaction protocol.

Conclusion

Herein, we have successfully developed a simple, environmentally benign, and efficient green methodology for the synthesis of 2,4,5-triarylimidazole derivatives using unconventional CuB_4O_7 utilizing one-pot three-component reaction. This environmentally green approach provides access to 2,4,5-triarylimidazole derivatives in good to excellent yield using an unconventional and inexpensive CuB_4O_7. The developed procedure was found to be operative for a wide range of aldehyde (aromatic/heteroaromatic) substrate, and we have been able to report the *in situ* formation of copper acetate in the reaction mixture from the reaction between different copper salts in the presence of ammonium acetate.

Experimental section

Materials and methods

All starting materials of high purity for the aforementioned synthesis were purchased commercially and were used as received. The FT-IR spectra of the prepared compound were recorded using a Bruker Alpha III spectrophotometer operating in the wavenumber region from 4000 to 400 cm^{-1} in dry KBr. Single crystal X-ray diffraction study was performed using an Xcalibur Sapphire 3 (manufactured by Oxford Diffraction, Poland) equipped with CCD camera. The melting point of the synthesized compounds was determined by the open capillary method. The ^{1}H-NMR spectra of the synthesized tri-aryl imidazole derivatives were recorded at room temperature on an FT-NMR (Bruker Avance-II 400 MHz) spectrometer using DMSO-d$_6$ as solvent, and chemical shifts were quoted in ppm downfield of internal standard tetramethylsilane (TMS).

General procedure for the synthesis of 2,4,5-triarylimidazole

In a typical procedure, a mixture of benzil (**1**) (1.0 mmol), substituted benzaldehyde (**2**) (1.0 mmol), ammonium acetate (**3**) (2.5 mmol), and copper borate (2 mol%) were thoroughly ground and mixed in a motor and pestle to make a homogeneous mixture. The mixture was then transferred to a test tube. The reaction was heated at 80 °C for 10 min. The progress of the reaction was monitored by TLC using hexane/ethyl acetate (80 : 20) solvent (1 mL). After the completion of the reaction, the reaction mixture was dissolved in methanol (10 mL) and filtered. The filtrate was evaporated under vacuum and subsequently dried to afford the desired product (**4a–z**). Finally, the products were recrystallized from dry ethanol (10 mL) to afford the pure product.

2,4,5-Triphenyl-1*H*-imidazole (4a). White solid, yield = 98%, melting point found (°C) = 273–275, IR (KBr, cm^{-1}) υ_{max}: 3453 (NH), 1636 (C=C), 1503 (C=N), ^{1}H NMR (400 MHz, DMSO-d$_6$): δ 12.69 (s, 1H, NH), 7.22–7.55 (m, Ar H, 13H), 8.09 (d, 2H, $J = 7.2$ Hz) ppm; ^{13}C NMR (100 MHz, DMSO-d$_6$): δ 145.4, 138.2, 136.1, 131.5, 133.4, 130.1, 129.4, 128.7, 128.2, 127.8, 126.2 ppm [lit. 94].

2-(2-Nitrophenyl)-4,5-diphenyl-1*H*-imidazole (4b). Pale yellow solid, yield = 96%, melting point (°C) = 231–233, IR (KBr, cm^{-1}) υ_{max}: 3444 (NH), 1619 (C=C), 1602 (C=N), ^{1}H NMR (400 MHz, DMSO-d$_6$): δ 12.98 (s, 1H, NH), 7.21–7.97 (m, Ar H, 12H), 8.01 (d, 2H, $J = 8.0$ Hz) ppm; ^{13}C NMR (100 MHz, DMSO d$_6$): δ 148.7, 141.4, 138.2, 135.2, 132.6, 131.2, 130.5, 130.2, 129.6, 129.4, 128.6, 128.5, 127.5, 127.2, 124.3, 123.7 ppm [lit. 95].

2-(3-Nitrophenyl)-4,5-diphenyl-1*H*-imidazole (4c). Pale yellow solid, yield = 95%, melting point (°C) = 316–318, IR (KBr, cm^{-1}) υ_{max} = 3443 (NH), 3056–3026 (aromatic C–H) 1660 (C=C), 1598 (C=N), ^{1}H NMR (400 MHz, DMSO-d$_6$): δ 12.56 (s, 1H, NH), 7.61–8.37 (m, 12H, Ar H), 8.62–8.68 (d, 2H, Ar H); ^{13}C NMR (100 MHz, DMSO d$_6$): δ 148.7, 143.6, 138.1, 135.6, 132.4, 131.8, 131.3, 131.1, 197.7, 129.4, 128.9, 128.8, 128.1, 127.8 ppm [lit. 96].

2-(4-Nitrophenyl)-4,5-diphenyl-1*H*-imidazole (4d). Yellow solid, yield: 97%, melting point (°C) = 235–237, IR (KBr, cm^{-1}) υ_{max} = 3422 (NH), 3057 (aromatic C–H), 1660 (C=C), 1600 (C=N), ^{1}H NMR (400 MHz, DMSO-d$_6$): δ 12.60 (s, 1H, NH), 7.24–8.45 (m, Ar H, 14H) ppm; ^{13}C NMR (100 MHz, DMSO d$_6$): δ 147.3, 143.9, 138.8, 136.7, 135.3, 130.5, 129.6, 129.3, 128.9, 127.5, 127.7, 126.4, 124.6 ppm [lit. 96].

2-(2-Chlorophenyl)-4,5-diphenyl-1*H*-imidazole (4e). Off white solid, yield: 96%, melting point (°C) = 194–196, IR

(KBr, cm^{-1}) υ_{max} = 3426 (NH), 3060 (aromatic C–H), 1665 (C=C), 1602 (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ 12.66 (s, 1H, NH), 7.21–7.93 (m, Ar H, 14H) ppm; ^{13}C NMR (100 MHz, DMSO d_6): δ 143.8, 137.6, 135.6, 132.3, 132.3, 131.4, 130.8, 130.5, 129.3, 128.8, 128.6, 128.4, 127.9, 127.6, 127.3 ppm [lit. 96].

2-(3-Bromophenyl)-4,5-diphenyl-1*H*-imidazole (4f). Off white solid, yield = 93%, melting point (°C) = 290–293, IR (KBr, cm^{-1}) υ_{max} = 3451 (NH), 3062, 3027 (aromatic C–H), 1694 (C=C), 1601 (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ 12.83 (s, 1H, NH), 8.30 (s, Ar H, 1H), 7.24–8.10 (m, Ar H, 13H) ppm; ^{13}C NMR (100 MHz, DMSO d_6): δ 144.4, 138.8, 135.4, 134.9, 133.5, 131.3, 130.2, 129.6, 129.4, 128.7, 127.7, 127.5, 126.3, 119.7, 110.8 ppm [lit. 97].

4-(4,5-Diphenyl-1*H*-imidazol-2-yl)benzonitrile (4g). White solid, yield: 95%, melting point (°C) = 186–188, IR (KBr, cm^{-1}) υ_{max} = 3449 (NH), 3081, 3053 (aromatic C–H), 1636 (C=C), 1610 (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ 13.03 (s, 1H, NH), 7.23–7.96 (m, Ar H, 12H), 8.26 (d, 2H, J = 8.2 Hz) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ: 144.4, 138.8, 135.4, 134.9, 133.5, 131.3, 130.2, 129.6, 129.4, 128.7, 127.7, 127.5, 126.3, 119.7, 110.8 ppm [lit. 96].

4-(4,5-Diphenyl-1*H*-imidazol-2-yl)phenol (4h). White solid, yield = 89%, melting point (°C) = 232–234, IR (KBr, cm^{-1}) υ_{max} = 3448 (NH), 3178 (aromatic C–H), 1641 (C=C), 1612 (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ 12.41 (s, 1H, NH), 9.73 (s, 1H, OH), 6.86–8.31 (m, Ar H, 14H) ppm; ^{13}C NMR (DMSO-d_6, 100 MHz) δ: 159.4, 144.9, 126.8, 126.3, 125.5, 125.3, 120.1, 113.9 ppm [lit. 96].

4-(4,5-Diphenyl-1*H*-imidazol-2-yl)-*N*,*N*-dimethylaniline (4i). Brown solid, yield = 97%, melting point (°C) = 257–260, IR (KBr, cm^{-1}) υ_{max} = 3427 (NH), 3060 (aromatic C–H), 1615 (C=C), 1552 (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ 13.03 (s, 1H, NH), 8.26 (d, 2H), 7.96 (d, 2H), 7.25–7.56 (m, Ar H, 10H) ppm; ^{13}C NMR (DMSO d_6, 100 MHz): δ 150.3, 146.6, 136.7, 135.4, 131.5, 128.8, 128.5, 128.2, 127.8, 127.2, 126.4, 118.5, 112.1, 39.6 ppm [lit. 96].

3-(4,5-Diphenyl-1*H*-imidazol-2-yl)phenol (4j). White solid, yield = 98%, melting point (°C) = 270–274, IR (KBr, cm^{-1}) υ_{max} = 3423 (NH), 3061 (aromatic C–H), 1593 (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ 13.51 (s, 1H, NH), 9.75 (s, 1H, OH), 6.87 (d, 2H, Ar–H), 7.30–7.93 (m, Ar H, 12H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6): δ 157.4, 145.2, 137.1, 135.0, 131.4, 131.3, 129.5, 128.3, 128.4, 128.1, 128.0, 127.6, 127.2, 126.4, 116.2, 115.5, 112.3 ppm [lit. 98].

2-(4,5-Diphenyl-1*H*-imidazol-2-yl)phenol (4k). White solid, yield = 97%, melting point (°C) = 205–207, IR (KBr, cm^{-1}) υ_{max} = 3443 (NH), 3060 (aromatic C–H), 1621 (C=C), 1603 (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ 12.97 (s, 1H, NH), 12.7 (s, 1H, OH), 6.95–8.13 (m, Ar H, 14H) ppm; ^{13}C NMR (100 MHz, DMSO d_6) δ: 147.8, 133.2, 130.4, 129.6, 128.3, 122, 118.6, 116.5 ppm [lit. 99].

4-Chloro-2-(4,5-diphenyl-1*H*-imidazol-2-yl)phenol (4l). White solid, yield = 94%, melting point (°C) ≥ 300, IR (KBr, cm^{-1}) υ_{max} = 3443 (NH), 3060 (aromatic C–H), 1644 (C=C), 1595 (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ 13.13 (s, 1H, NH), 10.27 (s, 1H, OH), 6.95–8.23 (m, Ar H, 13H) ppm; ^{13}C NMR (100 MHz, DMSO d_6): δ 145.1, 137.6, 135.6, 133.4, 131.6, 129.8, 129.4, 129.3, 129.2, 129.1, 128.7, 128.5, 127.7, 127.6, 127.3 ppm [lit. 99].

4-Bromo-2-(4,5-diphenyl-1*H*-imidazol-2-yl)phenol (4m). White solid, yield = 96%, melting point (°C) = 179–181, IR (KBr, cm^{-1}) υ_{max} = 3449 (NH), 3053 (aromatic C–H), 1636 (C=C), 1610 (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ 13.15 (s, 1H, NH), 13.09 (s, 1H, OH), 6.91–8.31 (m, Ar H, 13H), ^{13}C NMR (100 MHz, DMSO-d_6, 100 MHz): δ 144.7, 137.6, 135.2, 132.6, 132.4, 130.1, 129.9, 129.2, 129.1, 128.4, 127.8, 128.1, 127.8, 126.6, 121.5 ppm [lit. 99].

2-(4,5-Diphenyl-1*H*-imidazol-2-yl)-4-nitrophenol (4n). Yellow solid, yield = 96%, melting point (°C) = 262–264, IR (KBr, cm^{-1}) υ_{max} = 3443 (NH), 2958, 1633 (C=C), (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ = 13.3 (s, 1H, NH), 8.6 (s, 1H, OH), 7.20–7.50 (m, Ar H, 13H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6): δ 148.2, 147.4, 145.8, 137.8, 134.9, 131.7, 131.4, 130.5, 129.8, 128.5, 126.8, 126.6, 124.3 ppm [lit. 99].

4-(4,5-Diphenyl-1*H*-imidazol-2-yl)benzene-1,3-diol (4o). Black solid, yield = 97%, melting point (°C) = 270–273, IR KBr, (cm^{-1}) υ_{max} = 3443 (NH), 3063 (aromatic C–H), 1619 (C=C), 1606 (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ = 12.943 (s, 1H, NH), 9.73 (s, 1H, OH), 6.18–8.11 (m, Ar H, 13H), 4.049 (s, 1H, OH) ppm.

2-Methoxy-6-(4,5-diphenyl-1*H*-imidazol-2-yl)phenol (4p). White solid, yield = 95%, melting point (°C) = 168–172, IR (KBr, cm^{-1}) υ_{max} = 3464 (NH), 3050, 1632 (C=C), 1604 (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ 12.97 (s, 1H, NH), 6.86–7.67 (m, Ar H, 13H), 4.28 (s, 1H, OH), 3.87 (s, 3H, OCH_3) ppm; ^{13}C NMR (100 MHz, DMSO-d_6): δ 155.6, 147.3, 146.1, 145.1, 129.8, 127.4, 127.3, 126.6, 126.3, 117.1, 112.1, 110.9, 54.3 ppm [lit. 100].

4-(4,5-Diphenyl-1*H*-imidazol-2-yl)-2-methoxyphenol (4q). White solid, yield = 94%, melting point (°C) = 258–262, IR (KBr, cm^{-1}) υ_{max} = 3447 (NH), 2970 (aromatic C–H), 1647 (C=C), 1606 (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ 12.41 (s, 1H, NH), 6.83–7.67 (m, Ar H, 13H), 4.03 (s, 1H, OH), 3.90 (s, 3H, OCH_3) ppm; ^{13}C NMR (100 MHz, DMSO d_6): δ 146.8, 146.3, 132.3, 127.5, 127.3, 126.2, 121.1, 118.1, 114.6, 108.5, 55.1 ppm [lit. 96].

2-(4-(4,5-Diphenyl-1*H*-imidazol-2-yl)phenyl)-5,6-diphenyl-1*H*-benzo[*d*]imidazole (4r). Yellow solid, yield = 86%, melting point (°C) = 210–212, IR (KBr, cm^{-1}) υ_{max} = 3443, 3424 (NH), 3048 (aromatic C–H), 1654 (C=C), 1605, 1531 (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ 12.72 (s, 1H, NH), 8.21 (d, 2H), 7.25–7.55 (m, Ar H, 14H) ppm; ^{13}C NMR (100 MHz, DMSO d_6): δ 148.2, 137.7, 133.3, 130.4, 128.9, 128.6, 127.1, 126.9, 124.2 ppm [lit. 101].

4-(4,5-Diphenyl-1*H*-imidazol-2-yl)-2,6-dimethoxyphenol (4s). White solid, yield = 94%, melting point (°C) = 190–192, IR (KBr, cm^{-1}) υ_{max} = 3511 (NH), 3027 (aromatic C–H), 1615 (C=C), 1543 (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ 13.97 (s, 1H, NH), 9.15 (s, 1H, OH), 7.19–8.18 (m, Ar H, 12H), 3.36 (s, OCH_3, 6H) ppm [lit. 102].

2-(4,5-Diphenyl-1*H*-imidazol-2-yl)-1*H*-indole (4t). Yellow solid, yield = 96%, melting point (°C) = 306–308, IR (KBr, cm^{-1}) υ_{max} = 3413 (NH), 3056 (aromatic C–H), 1622 (C=C), 1598 (C=N), ^1H NMR (400 MHz, DMSO-d_6): δ 12.31 (s, 1H, NH), 11.40 (s, 1H, NH), 7.13–8.50 (m, Ar H, 15H), ^{13}C NMR (100 MHz, DMSO d_6): δ 143.5, 135.8, 135.4, 135.1, 131.4, 128.6, 127.8, 127.3, 126.4, 125.9, 125.5, 124.8, 123.5, 121.5, 121.4, 119.1, 111.4, 106.6 ppm [lit. 96].

4,5-Diphenyl-2-styryl-1*H*-imidazole (4u). White solid, yield = 92%, melting point (°C) = 140–143, IR (KBr, cm^{-1}) υ_{max} = 3449 (NH), 3060 (aromatic C–H), 1646 (C═C), 1600 (C═N), ^{1}H NMR (400 MHz, DMSO-d_6): δ 12.6 (s, 1H, NH), 7.58–7.92 (m, Ar H, 15H), 6.52 (d, 2H, CH═CH), ^{13}C NMR (100 MHz, DMSO d_6): δ 146.2, 138.1, 134.3, 130.1, 128.6, 128.6, 128.2, 128.0, 127.7, 125.8, 123.5, 110.8 ppm [lit. 103].

2-(10-Chloroanthracen-9-yl)-4,5-diphenyl-1*H*-imidazole (4v). Yellow solid, yield = 93%, melting point (°C) > 300, IR (KBr, cm^{-1}) υ_{max} = 3465 (NH), 3428 (Ar C–H stretch), 3048, 1654 (C═C), 1605 (C═N), ^{1}H NMR (400 MHz, DMSO-d_6): δ 13.07 (s, 1H, NH), 8.56 (d, 2H Ar–H *J* = 8.0 Hz), 7.29–8.51 (m, Ar H, 16H) ppm.

4,5-Diphenyl-2-(3,4,5-trimethoxyphenyl)-1*H*-imidazole (4w). White solid, yield = 98%, melting point (°C) = 162–166, IR (KBr, cm^{-1}) υ_{max} = 3354 (NH), 2930 (aromatic C–H), 1625 (C═C), 1590 (C═N), ^{1}H NMR (400 MHz, DMSO-d_6): δ 12.3 (s, 1H, NH), 7.20–7.62 (m, Ar H, 12H), 3.89 (s, 9H, OCH_3) ppm.

2-(4,5-Diphenyl-1*H*-imidazol-2-yl)pyridine. Light green solid, yield = 92%, melting point (°C) = 175–176, IR (KBr, cm^{-1}) υ_{max} = 3443 (NH), 1633 (C═C), 1486 (C═N), ^{1}H NMR (400 MHz, DMSO-d_6): δ 9.46 (s, 1H, NH), 7.42–7.69 (m, Ar H, 5H), 8.11–8.13 (m, 4H, *J* = 7.2 Hz) ppm; ^{13}C NMR (100 MHz, DMSO d_6): δ 150.1, 149.6, 139.2, 137.6, 135.8, 131.5, 130.1, 129.3, 129.1, 128.5, 128.1, 127.8, 127.4, 123.6, 120.5 [lit. 96].

3-(4,5-Diphenyl-1*H*-imidazol-2-yl)pyridine. Yellow solid, yield = 91%, melting point = 220–223 °C, IR (KBr, cm^{-1}) υ_{max} = 3444 (NH), 1637 (C═C), 1503 (C═N), ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.93 (s, 1H, NH), 7.25–7.94 (m, Ar H, 5H), 8.08–8.51 (m, 4H) ppm [lit. 104].

4,5-Diphenyl-2-(thiophen-2-yl)-1*H*-imidazole. Yellow solid, yield = 88%, melting point found (°C) = 170–173 °C, IR (KBr, cm^{-1}) υ_{max} = 3483 (NH), 1637 (C═C), 1501 (C═N), ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.81 (s, 1H, NH), 7.31–7.68 (m, Ar H, 5H), 8.10–8.13 (m, 4H, *J* = 7.2 Hz) ppm; ^{13}C NMR (100 MHz, DMSO d_6): δ 142.4, 137.6, 135.4, 134.6, 131.3, 130.1, 128.6, 128.3, 128.1, 127.7, 127.3, 126.9, 124.7 ppm [lit. 96].

Author contributions

Sailesh Chettri and Dhiraj Brahman: conceptualization, experimental and data curation; Biswajit Sinha, Sumiran Tamang, Dhiraj Brahman and Sailesh Chettri: formal analysis, writing original draft and software; Biswajit Sinha, Kiran Pradhan and Dhiraj Brahman: supervision, review, editing and visualization.

Conflicts of interest

There are no conflicts of interest to declare.

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