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Original article

An expedient one-pot synthesis of highly substituted imidazoles using supported ionic liquid-like phase (SILLP) as a green and efficient catalyst and evaluation of their anti-microbial activity

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ABSTRACT

An efficient method for the synthesis of imidazole derivatives by a three-component condensation of benzil or 9,10-phenanthrenequinone, aldehydes and ammonium acetate using supported ionic liquid-like phase (SILLP) catalyst under ultrasonic irradiation or classical heating conditions is reported. The present methodology offers several advantages, such as excellent yields, simple procedures, short reaction times, simple work-up and mild conditions. The catalyst is easily separated from the products by filtration and also exhibits remarkable reusable activity. These highly substituted imidazoles were also evaluated for their anti-microbial activity.

1. Introduction

Multi-substituted imidazoles, an important class of pharmaceutical compounds [1–3], exhibit a wide spectrum of biological activities such as nitric-oxide synthase inhibition [4], anti-inflammatory [5], anti-parasitic [6], antifungal [7], antidepressant [8], antitubercular [9], anticancer [10] and antiviral activities [11] as well as antileishmanial activity against *Leshmaniadonovani* [12]. Some of these compounds could also be used as organic optical materials in many fields, for example as signaling, fluorescent biosensory/chemosensory materials, molecular switches and organic light emitting diodes (OLEDs) [12–15]. They can also be useful in asymmetric organic synthesis [16] and polymer and material science [17]. Therefore, preparation of substituted imidazoles has attracted considerable attention in recent years and numerous methods for their synthesis have been reported [17–23]. Some of these methods are associated with one or more disadvantages such as using expensive reagents, long reaction time, tedious work-up procedures and generation of large amount of toxic waste. However, there are few reports on the synthesis of phenanthroimidazole compounds due to their lower solubility in organic solvents and the steric hindrance in these molecules [24–26]. On the other hand one important aspect of clean technology is the use of environmentally friendly catalysts, typically a solid catalyst that can be easily recovered when the reaction is complete.

Due to the resistance of some microorganisms to imidazole action because of outer membrane modifications [27], development of new and different anti-microbial agents has become a prime objective of medicinal and synthetic chemists. Therefore much of the current research effort is oriented toward the design of new and readily available drugs [28].

2. Experimental

Melting points were measured on an Electrothermal 9100 apparatus and are uncorrected. ^{1}H NMR spectra were obtained on a Bruker AVANCE III 400 MHz and ^{13}C NMR spectra were collected on an AVANCE III 100 MHz advanced spectrometer. FT-IR spectra were recorded on a Shimadzu FT-IR 8400S spectrometer. Chemical shifts on ^{1}H NMR and ^{13}C NMR were expressed in ppm downfield from tetramethylsilane. Sonication was performed in an Elmasonic S 40H ultrasonic cleaning unit. Elemental analyses were conducted on a Carlo-Erba EA1110CNNO-S analyser and agreed with the calculated values. All chemicals were purchased from Merck and used without further purification.

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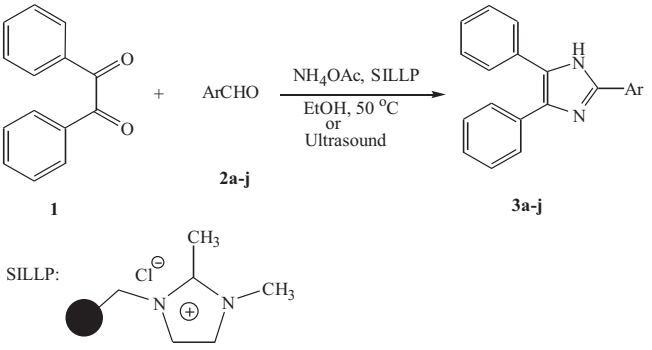
A mixture of benzil or 9,10-phenanthrenequinone (1 mmol), corresponding aldehyde (1 mmol), ammonium acetate (4 mmol) and a catalytic amount of supported ionic liquid-like phase (SILLP) (0.1 g) in ethanol (3 mL) was stirred in an oil bath at 50 °C or under ultrasonic irradiations (40 kHz, 50 °C) for a specified period of time. The progress of the reaction was monitored by TLC analysis (petroleum ether/ethyl acetate, 2/1). After the completion of the reaction, the crude product from the reaction mixture was dissolved in ethanol and the catalyst was separated by filtration. The filtrate was evaporated under reduced pressure to remove ethanol. The solid was then recrystallized from methanol to obtain the pure product. The data of some new compounds are listed below:

*4,5-Diphenyl-2-(1,3-diphenyl-1H-pyrazol-4-yl)-1H-imidazole (**3g**):* White solid, mp 239–240 °C; ^{1}H NMR (DMSO-d_6): δ 7.23–7.58 (m, 14H), 7.96–8.11 (m, 6H), 9.01 (s, 1H), 12.52 (s, 1H). ^{13}C NMR (DMSO-d_6): δ 113.4, 119.7, 126.9, 127.1, 127.2, 127.4, 128.6, 128.7, 129, 129.2, 130.2, 133.1, 135.7, 136.8, 139.7, 140, 150.4. FT-IR (KBr, cm^{-1}): ν 3450, 3010, 2950, 1665, 1590, 1520, 1500, 1440, 1365, 1220, 1120, 1062, 960, 940, 910, 862, 800, 760, 750, 725, 690, 680. Anal. Calcd. for $C_{30}H_{22}N_4$ (438.52): C 82.17, H 5.06, N 12.78; found: C 82.07, H 5.01, N 12.61.

*2-(1-(4-Bromophenyl)-3-phenyl-1H-pyrazol-4-yl)-4,5-diphenyl-1H-imidazole (**3h**):* White solid, mp 259–261 °C; ^{1}H NMR (DMSO-d_6): δ 7.24–7.60 (m, 13H), 7.69 (d, 2H), 8.05 (d, 2H), 8.15 (d, 2H), 9.03 (s, 1H), 12.54 (s, 1H). ^{13}C NMR (DMSO-d_6): δ 113.4, 118.9, 122.1, 128.2, 128.6, 128.8, 129.2, 130.1, 130.3, 130.8, 131.5, 132.3, 135.6, 136.9, 139.5, 139.8, 149.2. FT-IR (KBr, cm^{-1}): ν 3400, 3010, 2950, 2068, 1662, 1586, 1500, 1465, 1440, 1362, 1300, 1220, 1065, 1005, 960, 940, 820, 760, 745, 735, 720, 693; Anal. Calcd. for $C_{30}H_{21}BrN_4$ (517.42): C 69.64, H 4.09, N 10.83; found: C 69.80, H 4.14, N 11.05.

*2-(1-(4-Nitrophenyl)-3-phenyl-1H-pyrazol-4-yl)-4,5-diphenyl-1H-imidazole (**3i**):* Yellow solid, mp 248–250 °C; ^{1}H NMR (DMSO-d_6): δ 7.32–7.65 (m, 13H), 7.98 (d, 2H), 8.36 (d, 2H), 8.53 (d, 2H), 9.10 (s, 1H), 12.62 (s, 1H). ^{13}C NMR (DMSO-d_6): δ 113.1, 119.1, 123.8, 126.8, 127.5, 128.6, 128.8, 129.3, 129.8, 130.4, 130.5, 131, 135, 139.4, 145.1, 145.9, 147.4. FT-IR (KBr, cm^{-1}): ν 3450, 3010, 2990, 2070, 1710, 1700, 1625, 1590, 1518, 1500, 1485, 1338, 1380, 1180, 1160, 1100, 1060, 1020, 960, 920, 850, 835, 760, 690. Anal. Calcd. for $C_{30}H_{21}N_5O_2$ (483.52): C 74.52, H 4.38, N 14.48; found: C 74.65, H 4.25, N 14.19.

*2-(4-Chlorophenyl)-1H-phenanthro[9,10-d]imidazole (**6b**):* Milky solid, mp > 300 °C; ^{1}H NMR (DMSO-d_6): δ 7.76–7.64 (m, 6H), 8.33 (d, 2H), 8.53 (d, 1H), 8.58 (d, 1H), 8.85 (d, 1H), 8.88 (d, 1H), 13.55 (s, 1H). ^{13}C NMR (DMSO-d_6): δ 122.3, 122.4, 122.8, 124.3, 124.6, 125.8, 126, 127.3, 127.7, 128.1, 128.3, 129.5, 129.7, 134.2, 148.4. FT-IR (KBr, cm^{-1}): ν 3450, 3050, 2950, 2360, 1610, 1508, 1468, 1450, 1420, 1370, 1230, 1100, 1090, 960, 825, 745, 720, 710, 680. Anal. Calcd. for $C_{21}H_{13}ClN_2$ (328.79): C 76.71, H 3.98, N 8.52; found: C 76.45, H 4.22, N 8.33.



Scheme 1. Synthesis of tri-substituted imidazoles (**3a–j**).

Table 2
Comparison of efficiency of the catalysts in one-pot synthesis of **3b** in ethanol at 50 °C under conventional heating.

Entry	Catalyst	Time (min)	Yield (%)[a]
1	SILLP	30	95
2	Fe^{3+}-montmorillonite	80	89
3	Celloluse-sulfuric acid	120	68
4	L-proline	540	85

[a] Isolated yields.

*2-(4-Fluorophenyl)-1H-phenanthro[9,10-d]imidazole (**6c**):* Milky solid, mp > 300 °C; ^{1}H NMR (DMSO-d_6): δ 7.46 (t, 2H), 7.63 (t, 2H), 7.74 (s, br. 2H), 8.37 (dd, 2H), 8.57 (s, br., 2H), 8.84 (d, 2H), 13.49 (s, 1H). ^{13}C NMR (DMSO-d_6): δ 116.3, 116.6, 122.4, 122.9, 124.5, 125.7, 127.5, 127.6, 128.1, 128.8, 128.9, 148.7, 162, 164.4. FT-IR (KBr, cm^{-1}): ν 3455, 3050, 2900, 2070, 1600, 1520, 1485, 1450, 1420, 1378, 1347, 1225, 1132, 1100, 1038, 830, 750, 690, 610, 505. Anal. Calcd. for $C_{21}H_{13}FN_2$ (312.34): C 80.75, H 4.20, N 8.97; found: C 80.60, H 4.39, N 8.75.

*2-(1,3-Diphenyl-1H-pyrazol-4-yl)-1H-phenanthro[9,10-d]imidazole (**6d**):* Pale yellow solid, mp 228–230 °C; ^{1}H NMR (DMSO-d_6): δ 7.38–7.75 (m, 10H), 8.03 (m, 4H), 8.36 (d, 1H), 8.50 (d, 1H), 8.88 (m, 2H), 9.20 (s, 1H), 13.42 (s, 1H). ^{13}C NMR (DMSO-d_6): δ 113.4, 119.3, 122.6, 125.7, 126, 127.1, 127.2, 127.4, 127.5, 128.7, 130.1, 130.2, 130.4, 131.7, 132.3, 141.3, 143.5, 149.8. FT-IR (KBr, cm^{-1}): ν 3455, 2900, 2850, 2065, 1658, 1620, 1600, 1540, 1460, 1378, 1270, 1180, 1080, 1060, 960, 820, 745, 720, 690. Anal. Calcd. for $C_{30}H_{20}N_4$ (436.51): C 82.55, H 4.62, N 12.84; found: C 82.60, H 4.38, N 12.57.

*2-(1-(4-Bromophenyl)-3-phenyl-1H-pyrazol-4-yl)-1H-phenanthro[9,10-d]imidazole (**6e**):* Pale yellow solid, mp 122–124 °C; ^{1}H NMR (DMSO-d_6): δ 7.46 (t, 1H), 7.61–7.76 (m, 6H), 7.74 (m, 2H),

Table 1
One-pot synthesis of highly substituted benzimidazoles (**3a–j**) in the presence of supported ionic liquid-like phases as a solid green catalyst at 50 °C, under ultrasonic irradiation and conventional condition.

Entry	Ar	Conventional		Ultrasound		MP (°C)	
		Time (min)	Yield (%)[a]	Time (min)	Yield (%)[a]	Found	Reported[c]
3a	Ph	30	97	5	95	270–272	272–273 [14]
3b	4-ClC_6H_4	30 (60)[b]	95 (75)[b]	5 (5)[b]	94 (70)[b]	259–262	261–262 [14]
3c	2-ClC_6H_4	35	92	6	90	195–196	190–191 [18]
3d	4-FC_6H_4	35	96	6	92	240–242	239–241 [19]
3e	4-OHCC_6H_4	40	90	6	87	238–240	242–244 [20]
3f	4-$MeO_2CC_6H_4$	40	90	6	89	234–235	235–236 [21]
3g	1,3-Diphenyl-1*H*-pyrazol-4-yl	30	89	6	88	239–240	–
3h	1-(4-Bromophenyl)-3-phenyl-1*H*-pyrazol-4-yl	30	86	6	85	259–261	–
3i	1-(4-Nitrophenyl)-3-phenyl-1*H*-pyrazol-4-yl	35	83	6	85	248–250	–

[a] Isolated yields.
[b] Reaction at room temperature.
[c] Identified by comparison of their melting points and spectral data with those reported in the literature.

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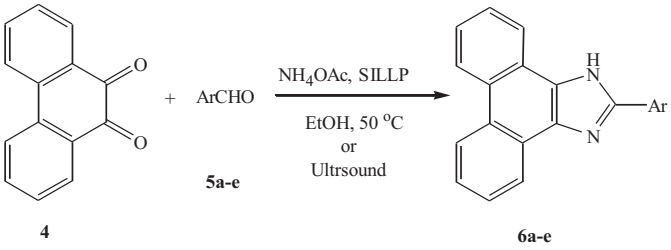
Table 3
One-pot synthesis of highly substituted phenanthroimidazoles (**6a–e**) in the presence of SILLP at 50 °C, under ultrasonic irradiation and conventional heating.

Entry	Ar	Conventional		Ultrasound		MP (°C)	
		Time (min)	Yield (%)[a]	Time (min)	Yield (%)[a]	Found	Reported
6a	Ph	20	95	3	94	>300	322–324 [24]
6b	4-ClC_6H_4	20	93	3	93	>300	–
6c	4-FC_6H_4	20	95	3	94	>300	–
6d	4-(1,3-Diphenyl-1*H*-pyrazole)	20	92	4	89	228–230	–
6e	1-(4-Bromophenyl)-3-phenyl-1*H*-pyrazol-4-yl	20	90	4	91	122–124	–

[a] Isolated yields.



Scheme 2. Synthesis of tri-substituted phenantroimidazoles (**6a–e**).

8.04 (d, 2H), 8.09 (d, 2H), 8.39 (d, 1H), 8.52 (d, 1H), 8.88 (d, 1H), 8.92 (d, 1H), 9.24 (s, 1H), 13.43 (s, 1H). ^{13}C NMR (DMSO): δ 113.6, 119.1, 122.2, 122.4, 124.7, 125.8, 127.5, 127.6, 127.7, 127.9, 128.1, 130.3, 130.7, 130.9, 131.7, 132.1, 139.5, 143.4, 149.6. FT-IR (KBr, cm^{-1}): ν 3450, 2900, 2850, 2360, 1735, 1580, 1500, 1460, 1400, 1245, 1220, 1160, 1140, 1080, 960, 820, 750, 720, 680. Anal. Calcd. for $C_{30}H_{19}BrN_4$ (515.4): C 69.91, H 3.72, N 10.87; found: C 69.75, H 3.50, N 11.04.

3. Results and discussion

In the course of our search for the development of efficient and environmentally friendly protocols for the synthesis of biologically important heterocyclic products [29], we studied the synthesis of highly substituted imidazole derivatives as potential drug candidates, over a supported ionic liquid-like phase (SILLP) catalyst (Scheme 1). Initially the SILLP was prepared by using Merrifield resin (1% cross linked, 200–400 mesh, 1–1.3 mmol/g) and the procedure employed by Luis *et al.* [30]. In order to optimize the effect of the amount of catalyst on the efficiency of the reaction, the condensation of benzil (1 mmol), 4-chlorobenzaldehyde (1 mmol) and ammonium acetate (4 mmol) in the presence of SILLP was selected as a model reaction (Scheme 1). This study gave the optimized amount of the catalyst (0.1 g per mmol of substrate). The effect of different solvents was also examined for the model reaction in the presence of SILLP and we obtained the maximum yield of the product in shortest reaction time, when ethanol was used as solvent. Therefore all the reactions described in this report were carried out under these optimized conditions. The results of this study using various aryl- and heteroaryl aldehydes are given in Table 1.

Table 2 compares efficiency of SILLP with other catalysts in the synthesis of **3b**. It is clear from the results that SILLP is more efficient, and less time-consuming for the synthesis of the desired product (**3b**).

In order to determine the scope of this protocol, the reaction of 9,10-phenanthrenequinone was also carried out in the presence of SILLP with various aromatic and heteroaromatic aldehydes under the optimized conditions (Scheme 2). The results are summarized in Table 3. It can be easily seen that in all cases, regardless of the nature of the substituents, the reactions gave the products in very high yields.

Due to environmental concerns, ultrasonic reactions have attracted more and more attention as clean, green and benign routes for the preparation of organic compounds of synthetic and biological value [31].

To improve the efficiency of the present method for the synthesis of highly substituted imidazole derivatives, we also carried out the reaction under ultrasonic irradiation (40 Hz, EtOH, 50 °C). The results are presented in Tables 1 and 3. Using ultrasound gave comparable yields of products but with shorter reaction time (3–6 min), compared to conventional heating.

In this study the catalyst was separated simply by filtration from the reaction mixture, washed by ethanol and used in the next

Table 4
Antimicrobial activity of compounds **3a–i** and **6a–e**.

Compound	Conc. of compound (μg/well)	Antimicrobial activity (zone of inhibition in mm)			
		Escherichia coli	*Micrococcus luteum*	*Bacillus subtillis*	*Pseudomonas aeruginosa*
3a	6.25	9	10	9	11
3b	6.25	11	13	12	13
3c	6.25	11	12	12	13
3d	6.25	9	9	11	13
3e	6.25	10	10	12	12
3f	6.25	12	12	13	13
3g	6.25	17 (19)[a] (18)[b]	19 (18)[a] (20)[b]	18 (19)[a] (18)[b]	18 (17)[a] (19)[b]
3h	6.25	18 (18)[a] (17)[b]	19 (19)[a] (20)[b]	20 (18)[a] (19)[b]	18 (18)[a] (19)[b]
3i	6.25	19 (20)[a] (19)[b]	20 (19)[a] (21)[b]	20 (21)[a] (19)[b]	19 (18)[a] (20)[b]
6a	6.25	7	9	9	8
6b	6.25	11	11	10	9
6c	6.25	11	10	10	9
6d	6.25	17 (18)[a] (19)[b]	18(19)[a] (19)[b]	19 (18)[a] (19)[b]	18(20)[a] (19)[b]
6e	6.25	19 (19)[a] (18)[b]	18 (17)[a] (20)[b]	20 (21)[a] (19)[b]	18 (18)[a] (19)[b]
Erythromycin	15	16	10	12	10
Tetracycline	30	12	16	14	18

[a] Data of duplicated experiments.
[b] Data of triplicated experiments.

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run. The high catalytic activity was maintained even after fourth reuse of the catalyst.

The antibacterial activity of synthesized compounds **3a–i** and **6a–e** was examined against *Escherichia coli* (EC), *Micrococcus luteus* (ML), *Bacillus subtillis* (BS) and *Pseudomonas aeruginosa* (PS). For comparison, two routinely used antibiotics, tetracycline and erythromycin, were also included (Table 4). Although nearly all the compounds exhibited antimicrobial activity, the most active ones were compounds **3g**, **3h**, **3i**, **6d** and **6e**. Compared to the control antibiotics used, these compounds showed much improved activity.

4. Conclusion

In summary the work reported here offers an efficient one-pot multicomponent route for the synthesis of trisubstituted imidazole derivatives in the presence of supported ionic liquid-like phase (SILLP) as a green catalyst. The simple procedures combined with easy recovery and reuse of the catalyst make this method an economical, environmentally benign and user-friendly process for the synthesis of highly substituted imidazoles. Furthermore, some of the synthesized products (**3g**, **3h**, **3i**, **6d** and **6e**) exhibited highly potent antimicrobial activity, which needs further investigation.

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