Tetrahedron Letters 51 (2010) 5252–5257

Contents lists available at ScienceDirect

Tetrahedron Letters

journal homepage: www.elsevier.com/locate/tetlet

DABCO as a mild and efficient catalytic system for the synthesis of highly substituted imidazoles via multi-component condensation strategy

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ARTICLE INFO

Article history:
Received 31 May 2010
Revised 19 July 2010
Accepted 22 July 2010
Available online 29 July 2010

Keywords:
Aldehydes
Ammonium acetate
1,2-Dicarbonyl compounds
DABCO
Multi-component
Condensation
Imidazoles
Aromatic amines

ABSTRACT

A simple and efficient protocol for the synthesis of highly substituted imidazoles is developed through the condensation of 1,2-dicarbonyl compound, aldehyde, and ammonium acetate or amine via multi-component condensation strategy. The present method gives good to excellent yields of substituted imidazoles.

Heterocyclic chemistry is one of the most important topics in the synthetic organic chemistry arena that covers a wide variety of potent molecules. Imidazoles and benzimidazoles are structural scaffolds present as substructures in many natural products which possess wide spectrum of biological activities covering anti-inflamatory,[1] anti-allergic,[2] analgesic,[3] and glucagon receptor antagonism.[4] They are also the key intermediates in the synthesis of many therapeutic agents. Omeprazole,[5] Pimobendan,[6] Losartan, Olmesartan, Eprosartan, and Trifenagrel[7] (Fig. 1) are some of the leading drugs in the market with diverse functionalization around the imidazole motif. Apart from being biologically active, this structural core in the recent years has been in other advanced areas of research such as fluorescence labeling agents,[8–10] biological imaging,[11] and chromophores for non-linear optic systems.[12] In view of the numerous biological, pharmacological, and material properties associated with this five-membered heterocyclic moiety, the development of new synthetic protocols under varied mild reaction conditions is always a matter of interest.

In the last decade numerous methods have been developed for the synthesis of highly substituted imidazoles by using various catalytic systems including silica gel or Zeolite HY,[13] silica gel/$NaHSO_4$,[14] molecular iodine,[15] $K_5CoW_{12}O_{40}$-$3H_2O$,[16] heteropolyacids,[17] $HClO_4$–SiO_2,[18] L-proline,[19] $FeCl_3{\cdot}6H_2O$,[20] $BF_3{\cdot}SiO_2$[21], and silica-supported Wells–Dawson acid.[22] They can also be obtained by use of microwave irradiation,[23] and refluxing in acetic acid[24] silica sulfuric acid,[25] $NiCl_2{\cdot}6H_2O/Al_2O_3$,[26] $ZrCl_4$,[27] ionic liquids,[28] CAN,[29] and $InCl_3{\cdot}3H_2O$.[30] However, many of the methods reported above suffer from one or more disadvantages such as the use of expensive moisture-sensitive metallic reagents, longer reaction times, tedious separation procedures, and large amount of catalyst loadings which in turn results in the generation of huge amount of metal wastes into the environment.

Multi-component reaction (MCRs) is convergent, in analogy to the convergent synthesis and in contrast to a divergent multi-step synthesis. These reactions are classified into various ways based on the number of components involved in the reaction or their intrinsic variability. Now-a-days organic chemical syntheses involving multi-component condensation strategy attained greater value, as the target molecules are often obtained in a single step rather

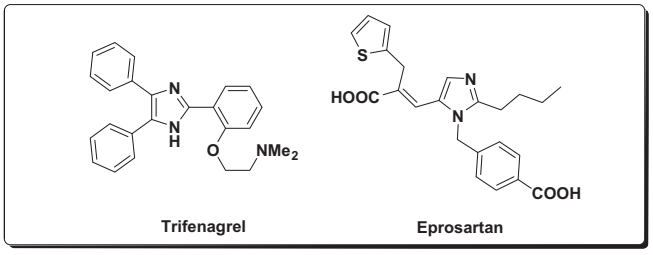

Figure 1. Potent multi-substituted imidazole derivatives.

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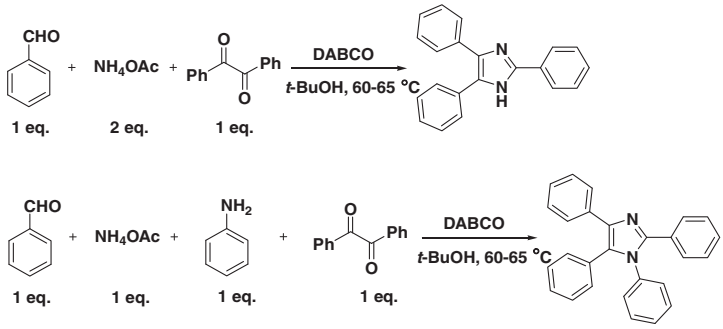
doi:10.1016/j.tetlet.2010.07.128



Scheme 1. DABCO-catalyzed synthesis of multi-substituted imidazole derivatives.

than multiple steps which minimize the tedious work-up procedures and environmental hazardous wastes. In a multi-component reaction the starting materials with desired functionalities reacted together in a specified path resulting in a complex adduct, incorporating all the reactants, making it an ideal synthetic setting.[31] In continuation of our efforts toward the development of novel methodologies under green chemical approaches herein we report a mild, efficient, and facile one-pot synthesis of multi-substituted imidazole derivatives for the first time by the multi-component reaction of 1,2-diketone, amine source, and aldehyde using DABCO as a base (Scheme 1).

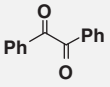
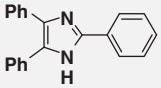
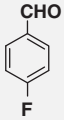
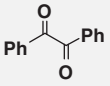
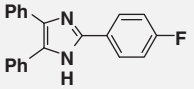
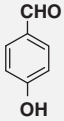
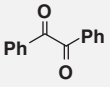
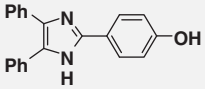
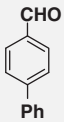
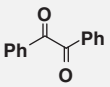
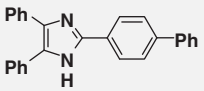
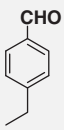
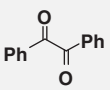
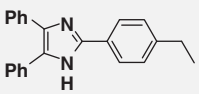
In our initial experiments toward the development of this methodology, we have reacted benzaldehyde (1.0 mmol) with ammonium acetate (2.0 mmol) and 1,2-diketone derivative (1.0 mmol) in ethanol resulting in the corresponding substituted imidazole in lower yields at room temperature. It was observed that the same reaction proceeded efficiently when we use DABCO as a catalyst in ethanol at room temperature yielding the corresponding substituted imidazole in 85% yield after 24 h. When we attempted the same reaction in *t*-butanol at 60–65 °C the reaction proceeded to completion within 12 h and yielded the corresponding imidazole in 92% yield.[32] Encouraged by this result we have explored the efficiency of various bases on the reaction. Among the bases tested for the condensation of benzaldehyde, ammonium acetate, and benzil to yield 2,4,5-triphenyl-1*H*-imidazole are triethyl amine (75%), piperidine (78%), DBU (81%), and DABCO (92%). However DABCO gave excellent yields.

In order to determine the most appropriate choice of solvent system for this DABCO-catalyzed synthesis of substituted imidazoles, we have screened the solvents such as methanol, ethanol, isopropanol, and *tert*-butanol. Of the solvents tested in the screening we got the maximum yield of the product in shorter reaction times, when we use *tert*-butanol as solvent (Table 2).

In order to determine the scope of this reaction, we have synthesized differently substituted 2,4,5-imidazoles and 1,2,4,5-imidazoles by varying differently substituted aldehydes including both electron-donating and electron-withdrawing groups. It is observed that the reaction gave good yields of products with faster reaction rate when the aldehyde bearing electron-withdrawing group is used compared to the aldehydes with electron-donating groups. The corresponding results are tabulated in Tables 1 and 3.

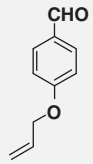
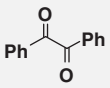
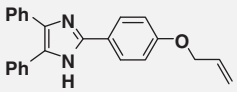
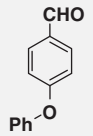
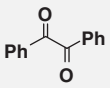
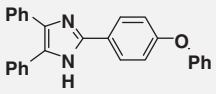
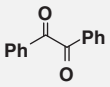
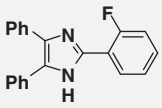
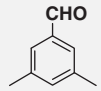
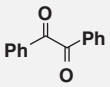
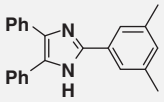
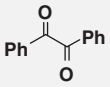
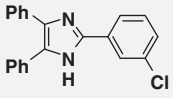
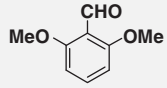
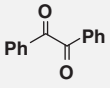
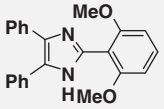
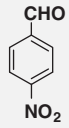
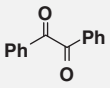
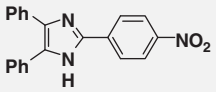
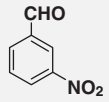
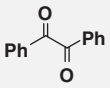
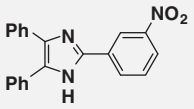
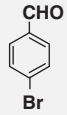
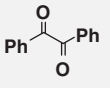
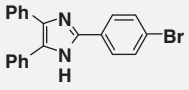
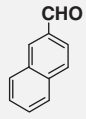
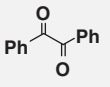
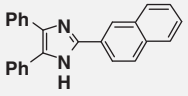
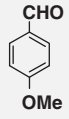
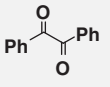
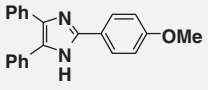
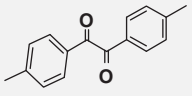
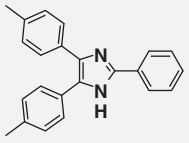
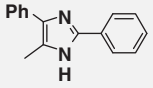
The plausible mechanism for the synthesis of substituted imidazoles in the presence of DABCO involves the initial reaction of DABCO with aldehyde leading to the formation of intermediate (A),

Table 1
Synthesis of 2,4,5-substituted imidazoles by using DABCO[a]

Entry	Aldehyde	Amine source	1,2-diketone	Product	Reported yield [ref]	Yield[b] (%)
1		NH_4OAc			90[19]	92
2		NH_4OAc			–	81
3		NH_4OAc			89[19]	88
4		NH_4OAc			–	86
5		NH_4OAc			–	89

(*continued on next page*)

Table 1 (*continued*)

Entry	Aldehyde	Amine source	1,2-diketone	Product	Reported yield [ref]	Yield[b] (%)
6		NH_4OAc			–	85
7		NH_4OAc			–	88
8		NH_4OAc			–	83
9		NH_4OAc			–	84
10		NH_4OAc			78[25b]	86
11		NH_4OAc			–	83
12		NH_4OAc			87[19]	89
13		NH_4OAc			86[19]	86
14		NH_4OAc			87[25b]	82
15		NH_4OAc			–	83
16		NH_4OAc			82[19]	84
17		NH_4OAc			91[28]	87
18		NH_4OAc			–	75
19		NH_4OAc			93[28]	72

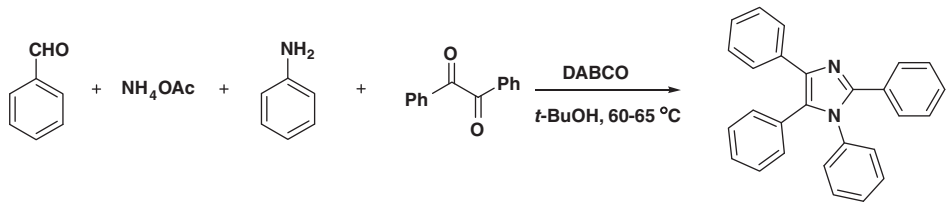
[a] Reaction conditions: aldehyde (1.0 mmol), ammonium acetate (2.0 mmol), 1,2-diketone (1.0 mmol), DABCO (0.7 mol %), *t*-BuOH (10 mL), 60–65 °C, 12–15 h.
[b] Isolated yield.

Table 2
Screening of solvents for the condensation of benzaldehyde, ammonium acetate, benzil using DABCO

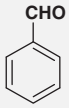
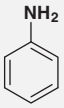
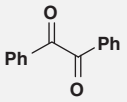
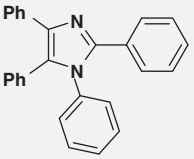
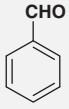
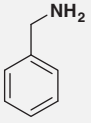
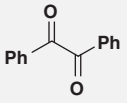
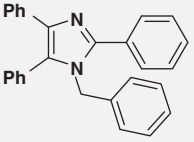
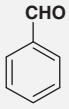
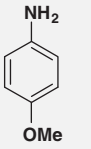
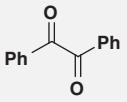
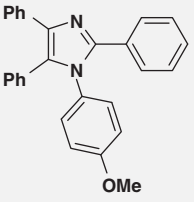
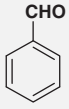
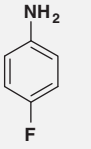
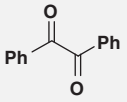
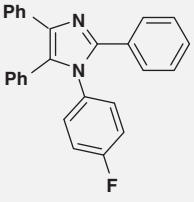
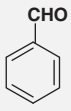
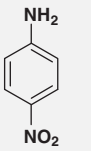
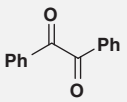
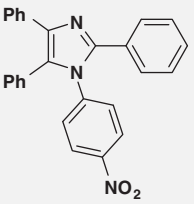
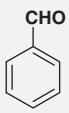
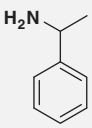
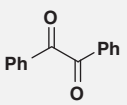
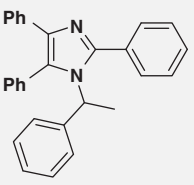
Entry	Solvent	Time (h)	Yield (%)
1	Methanol	12	67
2	Ethanol	12	80
3	*iso*-Propanol	12	78
4	*tert*-Butanol	12	92

which attacked by the amine source gives another intermediate (B) which in turn reacts with another molecule of DABCO, followed by a mole of amine source generating intermediate (C), with subsequent elimination of DABCO. This intermediate (C) on cyclocondensation with 1,2-diketone and 1,5-proton migration leads to the desired product (Scheme 2). All the products were characterized by ^{1}H, ^{13}C NMR, IR, and mass spectra and compared with authentic samples.[33]

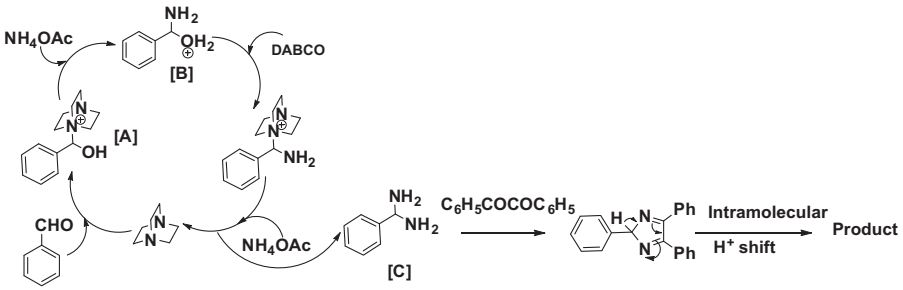
Table 3
Synthesis of 1,2,4,5-substituted imidazoles by using DABCO[a]



1 2 3 4 5

Entry	Aldehyde (1)	Amine (2)	Amine(3)	1,2-diketone (4)	Product (5)	Reported yield [ref]	Yield[b] (%)
1		NH_4OAc				83[30]	82
2		NH_4OAc				79[30]	74
3		NH_4OAc				–	77
4		NH_4OAc				–	73
5		NH_4OAc				–	72
6		NH_4OAc				92[23b]	70

[a] Reaction conditions: aldehyde (1.0 mmol), ammonium acetate (1.0 mmol), amine (1.0 mmol), 1,2-diketone (1.0 mmol), DABCO (0.7 mol %), *t*-BuOH(10 mL), 60–65 °C, 12–15 h.
[b] Isolated yield.



Scheme 2. Plausible mechanistic pathway for the formation of substituted imidazole scaffold.

In conclusion, we have developed a one-pot multi-component reaction for the synthesis of 2,4,5-trisubstituted and 1,2,4,5-tetra-substituted imidazoles catalyzed by DABCO in excellent yields. This method involves mild reaction conditions, easy work-up, and cleaner reaction profiles.

Acknowledgment

S.N.M. and B.M. are grateful to the Council of Scientific and Industrial Research (CSIR), New Delhi, India for providing fellowships.

Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.tetlet.2010.07.128.

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32. (a) *General procedure for the synthesis of 2,4,5-substituted imidazole derivatives*: to a stirred solution of *t*-butanol (10 mL), aldehyde (1.0 mmol) and DABCO (0.7 mol %) were added and stirred for 10 min. To this ammonium acetate (2.0 mmol) followed by 1,2-diketone (1.0 mmol) were added, after which the reaction mixture was heated at 60–65 °C until completion of the reaction as indicated by TLC. The reaction mixture was cooled to room temperature and the solvent was removed by rotary evaporator. The crude residue was extracted with ethyl acetate (3 × 10 mL). The organic layers were washed with water, saturated brine solution, and dried over anhydrous Na_2SO_4. The combined organic layers were evaporated under reduced pressure and the resulting crude product was purified by column chromatography by using ethyl acetate and hexane (7:3) as eluent to give the corresponding substituted imidazole derivative in (72–92%) yield. The identity and purity of the product were confirmed by ^1H and ^{13}C NMR spectroscopic analysis; (b) *General procedure for the synthesis of 1,2,4,5-substituted imidazole derivatives*: to a stirred solution of *t*-butanol (10 mL), aldehyde (1.0 mmol) and DABCO (0.7 mol %) were added and stirred for 10 min. To this ammonium acetate (1.0 mmol) and amine (1.0 mmol) followed by 1,2-diketone (1.0 mmol) were added, after which the reaction mixture was heated at 60–65 °C until completion of the reaction as indicated by TLC. The reaction mixture was cooled to room temperature and the solvent was removed by rotary evaporator. The crude residue was extracted with ethyl acetate (3 × 10 mL). The combined organic layers were washed with water, saturated brine solution, and dried over anhydrous Na_2SO_4. The organic solvent was evaporated under reduced pressure and the resulting crude product was purified by column chromatography by using ethyl acetate and hexane (7:3) as eluent to give the corresponding substituted imidazole derivative in (70–82%) yield. The identity and purity of the product was confirmed by ^1H and ^{13}C NMR spectroscopic analyses.
33. *Data of representative examples*:
2,4,5-Triphenyl-1H-imidazole (Table 1, *entry 1*): ^1H NMR (DMSO-d_6, 300 MHz): δ 8.05 (d, J = 7.5 Hz, 1H), 7.92 (d, J = 7.5 Hz, 1H), 7.52–7.24 (m, 13H), 12.59 (br s, 1H); IR (KBr): ν_{max} 1584, 1629, 3440 cm^{-1}; HRMS m/z calcd for $C_{21}H_{16}N_2$ ([M+H]$^+$): 297.1391, found 297.1399.
2-(4-(Allyloxy)phenyl)-4,5-diphenyl-1H-imidazole (Table 1, *entry* 6): ^1H NMR (DMSO-d_6, 300 MHz): δ 4.61 (d, J = 5.0 Hz, 2H), 5.29 (dd, J_1 = 10.3 Hz, J_2 = 1.1 Hz, 1H), 5.45 (dd, J_1 = 17.1 Hz, J_2 = 1.5 Hz, 1H), 6.00–6.13 (m, 1H), 7.07 (d, J = 8.8 Hz, 2H), 7.34–7.53 (m, 10H), 8.03(d, J = 8.8 Hz, 2H), 12.52 (br s, 1H);

^{13}C NMR (300 MHz, DMSO-d_6): δ 177.8, 158.3, 145.5, 133.5, 128.3, 127.6, 126.6, 123.2, 117.4, 114.7, 68.1; HRMS m/z calcd for $C_{24}H_{20}N_2O$ ([M+H]$^+$): 353.1653, found 353.1654.

*2-(4-Phenoxyphenyl)-4,5-diphenyl-1H-imidazole (*Table 1*, entry 7)*: ^{1}H NMR (DMSO-d_6, 300 MHz): δ 7.05–7.91 (m, 19H), 12.75 (br s, 1H); ^{13}C NMR (300 MHz, DMSO-d_6): δ 178.5, 157.3, 145.2, 130.9, 130.5, 128.8, 123.9, 120.9, 118.8, 115.9; HRMS m/z calcd for $C_{27}H_{20}N_2O$ ([M+H]$^+$): 389.1653, found 389.1643.

*2-(Naphthalen-2-yl)-4,5-diphenyl-1H-imidazole (*Table 1*, entry 15)*: δ 7.32–7.63 (m, 10H), 7.92–8.02 (m, 5H), 8.25 (d, J = 8.3 Hz, 1H), 8.62 (s, 1H), 12.60 (br s, 1H); ^{13}C NMR (300 MHz, DMSO-d_6): δ 177.9, 132.9, 132.7, 129.3, 128.5, 128.3, 128.2, 127.8, 126.8, 126.4, 123.7, 123.5; HRMS m/z calcd for $C_{25}H_{18}N_2$ ([M+H]$^+$): 347.1548, found 347.1538.

*4,5-Di(furan-3-yl)-2-phenyl-1H-imidazole (*Table 1*, entry 19)*: ^{1}H NMR ($CDCl_3$, 300 MHz): δ 6.31–6.33 (m, 2H), 6.79 (d, J = 3.3 Hz, 2H), 7.13–7.51 (m, 5H), 7.62–7.78 (m, 2H), 11.62 (br s, 1H); ^{13}C NMR (300 MHz, $CDCl_3$): δ 176.9, 149.3, 146.4, 141.3, 128.9, 128.4, 125.7, 107.7; ESI-MS (m/z): 277 (M$^+$+1).

*1-(4-Methoxyphenyl)-2,4,5-triphenyl-1H-imidazole (*Table 3*, entry 3)*: ^{1}H NMR ($CDCl_3$, 300 MHz): δ 3.76 (s, 3H), 6.72 (d, J = 8.8 Hz, 2H), 6.92 (d, J = 8.8 Hz, 2H), 7.09–7.55 (m, 13H), 8.04 (d, J = 8.8 Hz, 2H); HRMS m/z calcd for $C_{28}H_{22}N_2O$ ([M+H]$^+$): 403.1810, found 403.1795.

*1-(4-Fluorophenyl)-2,4,5-triphenyl-1H-imidazole (*Table 3*, entry 4)*: ^{1}H NMR (DMSO-d_6, 300 MHz): δ 7.25–7.37 (m, 4H), 7.42–7.46 (m, 2H), 7.50–7.63 (m, 9H), 7.72–7.81 (m, 2H), 7.89 (d, J = 6.7 Hz, 2H); HRMS m/z calcd for $C_{27}H_{19}FN_2$ ([M+H]$^+$): 391.1610, found 391.1598.

*2,4,5-Triphenyl-1-(1-phenylethyl)-1H-imidazole (*Table 3*, entry 6)*: ^{1}H NMR (DMSO-d_6, 300 MHz): δ 1.61 (d, J = 7.2 Hz, 3H), 5.55 (q, J_1 = 14.2 Hz, J_2 = 7.1 Hz, 1H), 6.89–7.57 (m, 18H), 8.09 (d, J = 7.2 Hz, 2H); HRMS m/z calcd for $C_{29}H_{24}N_2$ ([M+H]$^+$): 401.2017, found 401.2012.