



Simple and efficient method for the synthesis of highly substituted imidazoles using zeolite-supported reagents

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ABSTRACT

Cu(II) nitrate impregnated zeolite has been used as an efficient supported reagent for an improved and rapid one-pot synthesis of 2,4,5-trisubstituted and 1,2,4,5-tetrasubstituted imidazoles in excellent yields. Condensation in the presence of supported reagents with operational simplicity, inexpensive reagents, high yield of products, and the use of non-toxic reagents makes this synthetic protocol, an attractive one.

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The imidazole ring system is one of the most important substructure found in a large number of natural products and pharmacologically active compounds^{1–5} and the members of this class of compounds are known to possess NO synthase inhibition and antifungal, antimycotic, antibiotic, antiulcerative, antibacterial, antitumor, and CB1 receptor antagonistic activities.^{6,7} Various substituted imidazoles act as inhibitors of p38 MAP kinase^{8a} and B-Raf kinase,^{8b} glucagon receptors,⁹ plant growth regulators,¹⁰ therapeutic agents,¹¹ and pesticides.¹² Accordingly, a number of synthetic methods have been reported for the construction of this important structure.

Recently, multi-component reactions (MCRs) have attracted considerable attention since they are performed without the need to isolate any intermediate and save both energy and raw materials and also reduce time.¹³ In 1882, Radziszewski and Japp reported the first synthesis of the highly substituted imidazole from a 1,2-dicarbonyl compound, different aldehydes, and ammonia.^{14,15} Also a number of methods have been developed for the synthesis of 1,2,4,5-tetrasubstituted imidazoles and 2,4,5-trisubstituted imidazoles. The syntheses of 1,2,4,5-tetrasubstituted imidazoles are carried out by four-component condensation of a 1,2-diketone/ α -hydroxyketone with an aldehyde, primary amine, and ammonium acetate using microwaves,^{16a} molecular iodine,^{16b} HClO₄-SiO₂,^{16c} heteropolyacid,^{16d,e} silica gel/NaHSO₄,^{16f} L-proline,¹⁷ FeCl₃·6H₂O,¹⁸ BF₃·SiO₂,¹⁹ and silica-supported Wells-Dawson acid.²⁰ In addition, they can also be accessed by the condensation

of a 1,2-diketone with an aryl nitrile and primary amine under microwave irradiation,^{21a} by hetero-Cope rearrangement,^{21b} and by N-alkylation of trisubstituted imidazoles.^{21c} On the other hand, 2,4,5-trisubstituted imidazoles are generally synthesized by three-component cyclo-condensation of a 1,2-diketone/ α -hydroxyketone with an aldehyde and ammonium acetate, which comprise the use of microwaves,^{22a–d} refluxing in acetic acid,^{22e–g} silica sulfuric acid,^{22h} NiCl₂·6H₂O/Al₂O₃,²²ⁱ ZrCl₄,^{22j} ionic liquids,^{22k} and CAN.^{22l}

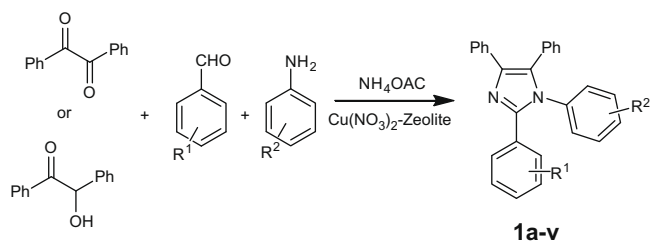
These methods are suitable for certain synthetic conditions, however, many of these procedures are associated with one or more disadvantages such as expensive reagents, longer reaction times, tedious work-up procedure, low selectivity, and large amounts of catalysts which would eventually result in the generation of large amounts of toxic waste.

Many organic reactions have been devised in which reagents are supported on various inorganic solid supports. These solid-supported reagents have several advantages over various conventional reagents. Laszlo and co-workers have successfully investigated reagents like iron(III) nitrate supported on K-10 montmorillonite and copper(II) nitrate supported on K-10. The variety and versatility of these supported reagent-based organic synthesis reported in recent years are the formation of new carbon–carbon bonds in cycloaddition reaction,²³ porphyrin synthesis by condensation of pyrrole with aldehydes,²⁴ oxidation of alcohols to aldehydes or ketones,²⁵ benzoin to benzils,²⁶ hydrolytic cleavage of thioacetals,²⁷ preparation of azides from hydrazines,²⁸ and nitration of phenols.²⁹

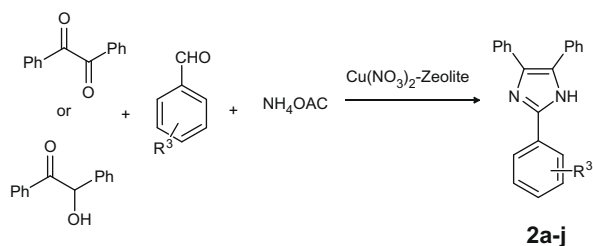
In this Letter, we have presented a novel, mild, and efficient method for the synthesis of tri and tetrasubstituted imidazoles

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Scheme 1.



Scheme 2.

Table 1

Synthesis of 1-(4-chlorophenyl)-2,4,5-triphenylimidazole **1a** using Cu(NO₃)₂-zeolite in different solvents

Entry	Solvent	Temperature °C	Time (h)	Yield ^a (%)
1	Methanol	65	3	56
2	Ethanol	70	3	53
3	Acetonitrile	70	3	41
4	Chloroform	50	6	22
5	Dichloromethane	45	6	12
6	Solvent-free	80	0.5	96

^a Yields refer to isolated products.

using supported reagents (Schemes 1 and 2). Out of range of zeolite supported metal nitrates, Cu(NO₃)₂ has attracted much attention because of its suitable acidity, eco-friendliness, easy availability, and low cost thereby acting as a promising table top reagent.

The typical procedure for 1,2,4,5-tetrasubstituted imidazoles involves impregnating the mixture of zeolite-supported metal nitrate (supported reagent) and ammonium acetate (ammonia source) with a dichloromethane solution of benzil, benzaldehyde, and 4-chloroaniline and was used as a model reaction to optimize the reaction conditions.^{30,31} Among the tested solvents such as methanol, ethanol, acetonitrile, chloroform, dichloromethane, and solvent-free system, the formation of product **1a** was more facile and proceeded to give highest yield, only under solvent-free reaction conditions (Table 1).

To evaluate and optimize the catalytic system, four-component condensation to give **1a** was examined with different solid acids and supported Lewis acid catalysts (Table 2). Interestingly, it was found that Cu(NO₃)₂ supported on zeolite-HY with low loading [one gram of zeolite-HY-supported copper(II) nitrate reagent contains about 0.241 g of Cu(NO₃)₂ (1 mmol)] was proved to be an efficient catalyst and gave exclusively 1-(4-chlorophenyl)-2,4,5-triphenylimidazole **1a** in 96% yield in 30 min under solvent-free conditions (Table 2).

From these experiments it was clearly demonstrated that the zeolite-supported cupric nitrate was indeed an effective catalyst and was convincingly superior to the reported procedures (Table 2, entries 16–18) with respect to reaction time, amount of catalyst and yields; and under solvent-free conditions. In order to

Table 2

Synthesis of 1,2,4,5-tetraphenylimidazole (**1a**) using different supported reagents under classical heating condition^a

Entry	Catalyst	Temp (°C)/time (min)	Yield ^b (%)
1	Cu(NO ₃) ₂ /zeolite-HY	80/30	96
2	Fe(NO ₃) ₃ /zeolite-HY	60/30	91
3	Bi(NO ₃) ₃ /zeolite-HY	120/30	88
4	Co(NO ₃) ₂ /zeolite-HY	120/60	45
5	Mn(NO ₃) ₂ /zeolite-HY	120/60	19
6	Ni(NO ₃) ₂ /zeolite-HY	120/60	18
7	Cu(NO ₃) ₂ /MCM-41	80/30	89
8	Fe(NO ₃) ₃ /MCM-41	60/30	77
9	Bi(NO ₃) ₃ /MCM-41	120/60	56
10	Co(NO ₃) ₂ /MCM-41	120/90	11
11	Mn(NO ₃) ₂ /MCM-41	120/90	—
12	Ni(NO ₃) ₂ /MCM-41	120/90	—
13	ZnCl ₂ /MCM-41	80/90	56
14	Zeolite-HY	120/90	31
15	Cu(NO ₃) ₂	80/60	Charred
16	SiO ₂ /NaHSO ₄ (400 mg)	140/120	92 ^{16f}
17	SiO ₂ /HClO ₄ (1 mol %)	140/6	90 ^{16c}
18	BF ₃ /SiO ₂ (21 mol %)	140/120	92 ¹⁹

^a The reaction condition: benzaldehyde (1 mmol), benzil (1 mmol), 4-chloroaniline (1 mmol), ammonium acetate (1 mmol), and catalyst (0.03 g) (metal salt loading = 1 mmol g⁻¹ of solid support).

^b Yields refer to isolated products.

Table 3

Cu(NO₃)₂-zeolite catalyzed synthesis of 1,2,4,5-tetrasubstituted imidazoles

Entry	Product	-R ₁	-R ₂	Reaction time		Yield ^a	
				Benzil	Benzoin	Benzil	Benzoin
1	1a	-H	4-Cl	0.5	1.0	96	92
2	1b	2-Cl	4-OCH ₃	0.5	1.0	92	80
3	1c	2-OH	4-Cl	0.5	1.0	76	72
4	1d	2-NO ₂	2-Cl	0.5	1.0	86	73
5	1e	4-NO ₂	2-NO ₂	0.5	1.0	93	82
6	1f	-H	-H	0.5	1.0	96	94
7	1g	4-OCH ₃	-H	0.5	1.0	95	92
8	1h	4-Cl	4-Cl	0.5	1.0	86	79
9	1i	2-Cl	4-CH ₃	0.5	1.0	94	90
10	1j	2-OH	4-CH ₃	0.5	1.0	86	75
11	1k	4-NO ₂	-H	0.5	1.0	97	82
12	1l	4-NO ₂	4-CH ₃	0.5	1.0	93	87
13	1m	4-Cl	-H	0.5	1.0	86	72
14	1n	4-OCH ₃	2-Cl	0.5	1.0	86	82
15	1o	4-Cl	2-NO ₂	0.5	1.0	95	79
16	1p	2-Cl	4-NO ₂	0.5	1.0	86	80
17	1q	2-OH	-H	0.5	1.0	84	77
18	1r	3-NO ₂	4-CH ₃	0.5	1.0	79	80
19	1s	4-Cl	4-CH ₃	0.5	1.0	86	82
20	1t	Furfural	2-NO ₂	0.5	1.0	63	52
21	1u	Furfural	4-NO ₂	0.5	1.0	67	62
22	1v	Furfural	-H	0.5	1.0	77	72

^a Yields refer to pure isolated solid products, characterized by mp, spectral (IR, ¹H, ¹³C NMR, and mass) data.

evaluate the generality of the process, several diversified examples illustrating the present method for the synthesis of 1,2,4,5-tetrasubstituted imidazoles **1a–v** were studied (Table 3).

The cyclo-condensation of benzil/benzoin with various aromatic aldehydes bearing electron-withdrawing groups (such as nitro and halide) or electron-releasing groups (such as methyl, hydroxyl, mono, di, or tri methoxy groups), substituted anilines, and ammonium acetate was carried out in the presence of Cu(NO₃)₂-zeolite as a catalyst. The yields obtained were good to excellent without the formation of any side products such as the formation of 2,4,5-trisubstituted imidazoles, oxidized products of anilines, and aldehydes, which are normally observed under the influence of strong acids. The results obtained in the current method are illustrated in Table 3. All the products obtained were fully

Table 4
Cu(NO₃)₂-zeolite catalyzed synthesis of 2,4,5-trisubstituted imidazoles

Entry	Product	-R ₃	Reaction time		Yield ^a	
			Benzil	Benzoin	Benzil	Benzoin
1	2a	4-Cl	0.5	1.0	98	91
2	2b	2-Cl	0.5	1.0	92	80
3	2c	2-OH	0.5	1.0	86	72
4	2d	2-NO ₂	0.5	1.0	87	75
5	2e	4-NO ₂	0.5	1.0	93	82
6	2f	-H	0.5	1.0	96	94
7	2g	4-OCH ₃	0.5	1.0	85	81
8	2h	Furfural	0.5	1.0	77	72
9	2i	2-Br 6-OH	0.5	1.0	71	62
10	2j	2-OH 4-OMe	0.5	1.0	81	72

^a Yields refer to pure isolated solid products, characterized by mp, spectral (IR, ¹H, ¹³C NMR, and mass) data.

characterized by spectroscopic methods such as IR, ¹H NMR, ¹³C NMR, and mass spectroscopy and also by making a comparison with the reported spectral data. The simplicity, together with the use of inexpensive, non-toxic, and environmentally benign nature of Cu(NO₃)₂-zeolite catalyst under solid state reaction condition is another remarkable feature of the procedure. From these results, it is clear that ring substituent like methoxy was not affected and the carbonyl derivative was not oxidized further.

In order to explore the applicability of this method, the same reaction conditions were applied for the synthesis of 2,4,5-trisubstituted imidazoles **2a–j** via a similar one-pot, three-component condensation of 1,2-diketone (1 mmol), aldehyde (1 mmol), and ammonium acetate (2 mmol) as depicted in Scheme 2.^{30,31}

We studied the synthesis of 2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazole (**2a**) using benzil, ammonium acetate, and 4-chloro benzaldehyde under different temperatures and with various mol % of Cu(NO₃)₂ in zeolite (Scheme 2). The compound **2a** was isolated with 98% yield using optimized reaction conditions (maximum % conversion at 80 °C and 1 mmol Cu(NO₃)₂ in zeolite-HY). Fascinated by the higher yields under mild reaction conditions, the same procedure has been extended for the synthesis of a range of 2-aryl-4,5-diphenyl-1H-imidazoles and the results are summarized in Table 4.

Generally, the synthetic procedure employed was similar to that of tetrasubstituted imidazoles. Adding ammonium acetate (as ammonia source) and a CH₂Cl₂ solution of benzoin/benzil and aldehyde, to the catalyst followed by evaporation of the solvent, and heating the resultant solid residue over an oil bath under optimized reaction condition lead to the efficient formation of the desired products. The yields obtained were in the range of 64–96%. There were no remarkable differences in yields and reaction time between arylaldehydes with electron-donating groups and those with electron-withdrawing groups. In the case of benzoin, comparable yields are possible as that of benzil only with longer reaction times.

The structures of compounds **2a–j** were deduced from their ¹H and ¹³C NMR, and IR spectral data and also confirmed from their melting points. The three-component condensation was also performed in the absence of Cu(NO₃)₂ on silica gel under classical heating; however, the yield of **2a** was low (~50%) illustrating the superiority of the supported reagent.

To check the reusability of the catalyst, the condensation of benzaldehyde, benzil, and NH₄OAc to provide the trisubstituted imidazoles under the conditions described with zeolite-HY-Cu(NO₃)₂ as a catalyst was run for five consecutive cycles, furnishing the corresponding imidazole, with 96%, 94%, 93%, 92%, and 92% isolated yields which proved the efficiency of the catalyst for multiple usage. Similar studies in the synthesis of tetrasubstituted imidazoles also

led to the conclusion that the catalyst is reusable without considerable loss in activity.

In summary, this Letter describes a convenient and efficient procedure for the synthesis of tetra and trisubstituted imidazoles through the one-pot cyclo-condensation reaction using zeolite-HY-Cu(NO₃)₂ as a solid supported reagent. Present methodology offers very attractive features such as reduced reaction times, higher yields, and economic viability of the catalyst, when compared with conventional methods as well as with other catalysts, which will have wide scope in organic synthesis. The simple procedures combined with easy recovery and reuse of this catalyst make this method economic, environmentally benign synthesis of tetra and trisubstituted imidazoles of biological and medicinal importance. The catalyst can be prepared easily with readily available inexpensive reagents, which is heterogeneous and non-hazardous.

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30. To 0.241 g of $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ (1 mmol) dissolved in 15 ml of acetone, 1 g of activated zeolite-HY (obtained by activating the $\text{NH}_4\text{-Y}$ form of zeolite in a muffle furnace at about 500 °C for 12 h) was added at once with stirring over a magnetic stirrer for 2 h. Then the solvent was removed in a rotary evaporator. The blue powder formed was dried further at 130 °C under reduced pressure. The reagent was stored in a vessel sealed with para film.
31. A solution of benzil or benzoin (1 mmol), aldehyde (1 mmol), amine (1 mmol), and ammonium acetate (1 mmol) in dichloromethane (10 mL) was added to supported reagent (300 mg) [one gram of zeolite-HY-supported copper(II) nitrate reagent contains about 0.241 g of $\text{Cu}(\text{NO}_3)_2$ (1 mmol)] in a reaction tube. The solvent was allowed to evaporate and the dry residue was heated at 80 °C for specified time in an oil bath. The progress of reaction was monitored by TLC. After 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 and the resulting product was washed with water. The solid product was then crystallized from hot ethanol. After extraction of the product, the supported reagent was washed thrice with ethanol and dried under vacuum before reuse.
- 1,2,4,5-Tetraphenyl-1H-imidazole (1f)**:¹⁹ mp, 220 °C; IR (KBr): ν_{max} 1595, 1572 cm^{-1} ; $^1\text{H NMR}$ (DMSO- d_6) δ : 7.24–7.47 (m, 20H); $^{13}\text{C NMR}$ (DMSO- d_6) δ : 124.1 (2C, CH_{arom}), 124.5 (2C, CH_{arom}), 125.1 (2C, CH_{arom}), 126.9 (CH_{arom}), 128.3 (CH_{arom}), 129.1 (CH_{arom}), 130.3 (CH_{arom}), 130.6 (2C, CH_{arom}), 132.9 (2C, CH_{arom}), 134.3 (2C, CH_{arom}), 136.8 (C_q), 139.0 (C_q), 139.2 (C_q), 139.5 (C_q), 139.8 (C_q), 140.1 (C_q), 144.6 (C=N).
- 2-(4-Methoxyphenyl)-1,4,5-triphenyl-1H-imidazole (1g)**:^{16b} mp, 183–184 °C; IR (KBr): ν_{max} 1617, 1576 cm^{-1} ; $^1\text{H NMR}$ (DMSO- d_6) δ : 3.74 (s, 3H), 6.80–6.84 (d, 2H), 7.20–7.38 (m, 15H), 7.49–7.51 (d, 2H); $^{13}\text{C NMR}$ (DMSO- d_6) δ : 54.7 (OCH_3), 115.2 (2C, CH_{arom}), 124.2 (2C, CH_{arom}), 126.1 (CH_{arom}), 127.7 (CH_{arom}), 128.1 (2C, CH_{arom}), 128.2 (2C, CH_{arom}), 128.4 (2C, CH_{arom}), 128.7 (2C, CH_{arom}), 130.1 (C_q), 132.3 (C_q), 132.7 (C_q), 134.9 (C_q), 136.7 (C_q), 137.1 (C_q), 146.5 (C=N), 159.4 (C_q).
- 1,2-Bis(4-chlorophenyl)-4,5-diphenyl-1H-imidazole (1h)**:^{16e} mp, 189–190 °C; IR (KBr): ν_{max} 1599, 1497, 1412 cm^{-1} ; $^1\text{H NMR}$ (DMSO- d_6) δ : 6.92–7.63 (m, 18H, Ar-H). $^{13}\text{C NMR}$ (DMSO- d_6) δ : 122.84 (2C, CH_{arom}), 126.98 (2C, CH_{arom}), 127.90 (CH_{arom}), 128.15 (2C, CH_{arom}), 129.21 (2C, CH_{arom}), 129.82 (2C, CH_{arom}), 129.82 (2C, CH_{arom}), 130.01 (C_q), 130.12 (C_q), 130.39 (C_q), 130.47 (C_q), 131.55 (C_q), 132.23 (C_q), 132.70 (C_q), 138.01 (C_q), 144.08 (C=N).
- 2-(2-Hydroxyphenyl)-4,5-diphenyl-1-p-tolyl-1H-imidazole (1j)**:^{16e} mp, 227–229 °C; IR (KBr): ν_{max} 1600, 1539, 1482, 1411 cm^{-1} ; $^1\text{H NMR}$ (DMSO- d_6) δ : 2.38 (s, 3H, CH_3), 6.41–7.42 (m, 18H, Ar-H), 13.21 (s, 1H, OH). $^{13}\text{C NMR}$ (DMSO- d_6) δ : 20.45 (CH_3), 113.72 (CH_{arom}), 116.81 (C_q), 121.97 (CH_{arom}), 125.85 (2C, CH_{arom}), 126.46 (CH_{arom}), 126.75 (2C, CH_{arom}), 128.24 (2C, CH_{arom}), 128.36 (2C, CH_{arom}), 128.57 (C_q), 129.45 (C_q), 129.73 (C_q), 129.97 (C_q), 130.71 (C_q), 131.14 (C_q), 134.06 (C_q), 134.17 (C_q), 144.45 (C=N), 156.39 (C_q).
- 2-(4-Nitrophenyl)-1,4,5-triphenyl-1H-imidazole (1k)**:¹⁷ mp, 190–192 °C; IR (KBr, cm^{-1}): ν_{max} 3057, 1590, 1513, 1342; $^1\text{H NMR}$ (DMSO- d_6) δ : 8.12 (d, 2H), 7.63–7.08 (m, 17H, Ar-H); $^{13}\text{C NMR}$ (DMSO- d_6) δ : 123.4 (2C, CH_{arom}), 124.0 (2C, CH_{arom}), 124.2 (2C, CH_{arom}), 127.4 (2C, CH_{arom}), 127.8 (CH_{arom}), 128.2 (CH_{arom}), 128.4 (2C, CH_{arom}), 128.6 (2C, CH_{arom}), 129.4 (C_q), 131.6 (C_q), 132.4 (C_q), 136.5 (C_q), 137.2 (C_q), 137.9 (C_q), 144.3 (C=N), 147.4 (C_q).
- 2-(4-Nitrophenyl)-4,5-diphenyl-1-p-tolyl-1H-imidazole (1l)**:^{16c} mp, 217–219 °C; IR (KBr): ν_{max} 1591, 1507, 1337 cm^{-1} ; $^1\text{H NMR}$ (DMSO- d_6) δ : 2.14 (s, 3H, CH_3), 7.17–7.49 (m, 18H, Ar-H). $^{13}\text{C NMR}$ (DMSO- d_6) δ : 21.31 (CH_3), 122.9 (2C, CH_{arom}), 124.8 (2C, CH_{arom}), 127.2 (2C, CH_{arom}), 128.7 (2C, CH_{arom}), 128.9 (CH_{arom}), 129.24 (2C, CH_{arom}), 129.39 (2C, CH_{arom}), 131.53 (C_q), 133.29 (C_q), 134.05 (C_q), 134.39 (C_q), 136.83 (C_q), 137.22 (C_q), 144.26 (C=N), 147.43 (C_q), 148.03 (C_q).
- 2-(4-Chlorophenyl)-1,4,5-triphenyl-1H-imidazole (1m)**:¹⁹ mp, 152–154 °C; IR (KBr): ν_{max} 1600, 1580; $^1\text{H NMR}$ (DMSO- d_6) δ : 7.31–7.65 (m, 15H), 7.69–7.70 (d, 2H), 7.94–7.97 (d, 2H); $^{13}\text{C NMR}$ (DMSO- d_6) δ : 124.2 (2C, CH_{arom}), 124.7 (2C, CH_{arom}), 125.2 (2C, CH_{arom}), 126.8 (CH_{arom}), 128.3 (CH_{arom}), 130.3 (CH_{arom}), 130.6 (2C, CH_{arom}), 132.9 (2C, CH_{arom}), 134.3 (2C, CH_{arom}), 136.8 (C_q), 138.1 (C_q), 139.0 (C_q), 139.2 (C_q), 139.5 (C_q), 139.8 (C_q), 140.1 (C_q), 144.4 (C=N).
- 2-(1,4,5-Triphenyl-1H-imidazol-2-yl)phenol (1q)**:^{16b} mp, 254–256 °C; IR (KBr): ν_{max} 1605, 1579 cm^{-1} ; $^1\text{H NMR}$ (DMSO- d_6) δ : 6.60–6.67 (d, 2H), 6.94–6.96 (d, 2H), 7.19–7.43 (m, 15H); $^{13}\text{C NMR}$ (DMSO- d_6) δ : 115.8 (CH_{arom}), 122.1 (C_q), 122.7 (CH_{arom}), 125.2 (2C, CH_{arom}), 126.5 (2C, CH_{arom}), 127.9 (CH_{arom}), 128.3 (CH_{arom}), 129.2 (2C, CH_{arom}), 130.8 (2C, CH_{arom}), 131.9 (CH_{arom}), 135.1 (CH_{arom}), 136.5 (C_q), 137.6 (C_q), 139.7 (C_q), 140.1 (C_q), 140.4 (C_q), 144.3 (C=N), 156.7 (C_q).
- 2-(3-Nitrophenyl)-4,5-diphenyl-1-p-tolyl-1H-imidazole (1r)**:^{16c} mp, 150–151 °C; IR (KBr): ν_{max} 1598, 1498, 1413 cm^{-1} ; $^1\text{H NMR}$ (DMSO- d_6) δ : 2.27 (s, 3H, CH_3), 7.17–8.85 (m, 18H, Ar-H). $^{13}\text{C NMR}$ (DMSO- d_6) δ : 21.31 (CH_3), 122.87 (CH_{arom}), 123.28 (2C, CH_{arom}), 126.80 (CH_{arom}), 127.25 (2C, CH_{arom}), 128.71 (CH_{arom}), 128.82 (2C, CH_{arom}), 129.32 (2C, CH_{arom}), 129.01 (CH_{arom}), 129.12 (CH_{arom}), 130.39 (C_q), 130.47 (C_q), 131.55 (C_q), 132.23 (C_q), 132.70 (C_q), 134.01 (C_q), 134.30 (C_q), 144.15 (C=N), 149.02 (C_q).
- A solution of benzil or benzoin (1 mmol), aldehyde (1 mmol), ammonium acetate (2 mmol), and 10 mL of dichloromethane was added to 300 mg of the zeolite-HY-Cu(NO_3)₂ mixture in a reaction tube. The solvent was allowed to evaporate and the dry residue was heated at 80 °C for specified time. The progress of reaction was monitored by TLC. After 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 and the resulting product was washed with water. The solid product was then crystallized from hot ethanol.
- 2-(4,5-Diphenyl-1H-imidazol-2-yl)phenol (2c)**:¹⁷ mp, 205 °C; IR (KBr): ν_{max} 1214, 1638, 2466, 2988, 3430, 3594; $^1\text{H NMR}$ (DMSO- d_6) δ : 6.87–6.95 (d, 2H), 6.97–7.01 (d, 2H), 7.17–7.23 (m, 10H), 12.74 (br s, 1H); $^{13}\text{C NMR}$ (DMSO- d_6) δ : 114.8 (CH_{arom}), 116.4 (C_q), 118.1 (CH_{arom}), 126.8 (2C, CH_{arom}), 127.2 (CH_{arom}), 127.8 (2C, CH_{arom}), 129.1 (CH_{arom}), 130.3 (CH_{arom}), 135.2 (C_q), 136.1 (C_q), 136.4 (C_q), 138.1 (C_q), 147.7 (C=N), 156.6 (C_q).
- 2-(4-Nitro-phenyl)-4,5-diphenyl-1H-imidazole (2e)**:¹⁷ mp 196 °C; IR (KBr): ν_{max} 845, 1448, 1528, 1542, 1604, 3058; $^1\text{H NMR}$ (DMSO- d_6) δ : 7.25–7.57 (m, 10H), 7.68 (d, 2H), 8.52 (d, 2H), 12.52 (br s, 1H); $^{13}\text{C NMR}$ (DMSO- d_6) δ : 124.2 (2C, CH_{arom}), 125.2 (2C, CH_{arom}), 126.3 (2C, CH_{arom}), 127.6 (CH_{arom}), 132.8 (2C, CH_{arom}), 133.1 (C_q), 135.2 (C_q), 138.3 (C_q), 144.7 (C_q), 164.8 (C=N).
- 2,4,5-Triphenyl-1H-imidazole (2f)**:¹⁷ mp, 272 °C; IR (KBr): ν_{max} 1216, 1640, 2474, 2993, 3438; $^1\text{H NMR}$ (DMSO- d_6) δ : 7.40–8.14 (m, 15H), 12.60 (br s, 1H); $^{13}\text{C NMR}$ (DMSO- d_6) δ : 127.1 (2C, CH_{arom}), 127.2 (CH_{arom}), 128.5 (2C, CH_{arom}), 129.1 (C_q), 131.5 (CH_{arom}), 137.2 (C_q), 138.2 (C_q), 168.7 (C=N).
- 2-(4-Methoxy-phenyl)-4,5-diphenyl-1H-imidazole (2g)**:¹⁷ mp, 224 °C; IR (KBr): ν_{max} 1226, 1640, 2462, 2892, 3424; $^1\text{H NMR}$ (DMSO- d_6) δ : 3.86 (s, 3H), 6.92–6.98 (d, 2H), 7.24–7.60 (m, 10H), 8.02–8.05 (d, 2H), 12.52 (br s, 1H); $^{13}\text{C NMR}$ (DMSO- d_6) δ : 54.6 (CH_3), 113.2 (CH_{arom}), 114.9 (2C, CH_{arom}), 126.7 (2C, CH_{arom}), 126.9 (CH_{arom}), 127.5 (2C, CH_{arom}), 127.9 (C_q), 129.6 (2C, CH_{arom}), 132.8 (C_q), 133.2 (C_q), 154.7 (C_q), 158.1 (C=N).