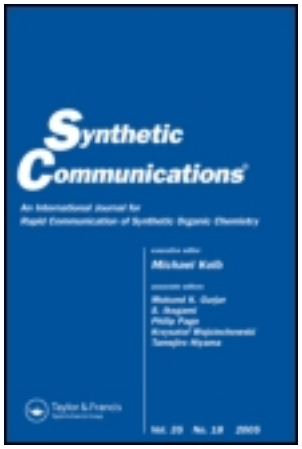

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# Sulfated Tin Oxide: A Reusable and Highly Efficient Heterogeneous Catalyst for the Synthesis of 2,4,5-Triaryl-1H-imidazole Derivatives

Satish A. Dake [a], Mahesh B. Khedkar [a], Ghanshyam S. Irmale [a], Suhas J. Ukalgaonkar [a], Vinod V. Thorat [b], Suhas A. Shintre [b] & Rajendra P. Pawar [b]

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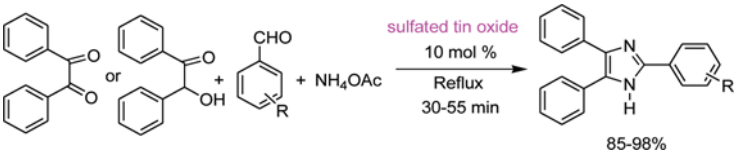
# SULFATED TIN OXIDE: A REUSABLE AND HIGHLY EFFICIENT HETEROGENEOUS CATALYST FOR THE SYNTHESIS OF 2,4,5-TRIARYL-1*H*-IMIDAZOLE DERIVATIVES

**Satish A. Dake,[1] Mahesh B. Khedkar,[1] Ghanshyam S. Irmale,[1] Suhas J. Ukalgaonkar,[1] Vinod V. Thorat,[2] Suhas A. Shintre,[2] and Rajendra P. Pawar[2]**

[1]*Organic Synthesis Laboratory, Department of Chemistry, Dnyanopasak College, Parbhani, India*
[2]*Department of Chemistry, Deogiri College, Aurgangabad, India*

## GRAPHICAL ABSTRACT




**Abstract** *One-pot, three-component condensation of benzil/benzoin, substituted aromatic aldehydes, and ammonium acetate in an ethanol–water (1:1,* v/v*) solvent system using sulfated tin oxide catalyst under reflux condition afforded corresponding 2,4,5-triaryl-1*H*-imidazoles in excellent yield. The remarkable advantages offered by this method include green and reusable catalyst, mild reaction conditions, fast reaction rate, and excellent yield of products. This novel methodology maintains atom economy and an environmentally friendly approach.*

**Keywords** Benzil/benzoin; ethanol; sulfated tin oxide; 2,4,5-triaryl-1*H*-imidazole; water

## INTRODUCTION

2,4,5-Triaryl-1*H*-imidazole compounds have gained remarkable importance because of their widespread biological activities. The heterocyclic imidazole ring is one of the most important structures found in a large number of natural products and pharmacologically active compounds such as antiulcerative agent cimetidine, the proton pump inhibitor omeprazole, and the benzodiazepine antagonist flumazenil. Trifenagrel, a 2,4,5-triaryl-1*H*-imidazole, reduces the platelet aggregation in several animal species and humans.

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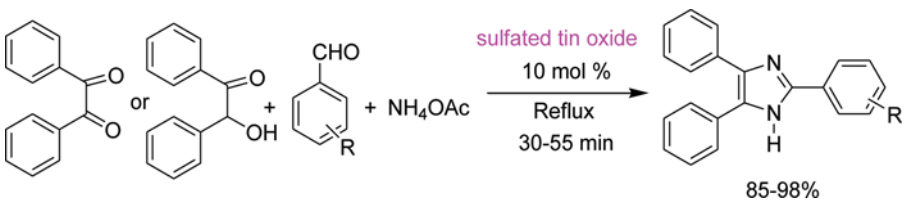
These compounds also play an important role in biochemical processes.[1] Imidazole derivatives show estrogen receptor and cytotoxic inhibitory activities of cyclooxygenase,[2] antifungal,[3] antihelmitic,[4] and pesticidal and herbicidal properties. Substituted imidazole possesses potentially novel therapeutic activities[5] and absolute controlled processes.[6] In addition, they also show analgesic,[7] fungicidal,[8] anti-inflammatory,[9] and antithrombotic activities.[10] Despite their importance from pharmacological and synthetic points of view, few methods for the preparation of trisubstituted imidazoles have been reported. These include the condensation of 1,2-diketone, aldehydes, and ammonium acetate in the presence of $H_3PO_4$,[11] $H_2SO_4$,[12] AcOH,[13] silica sulfuric acid,[14] *para*-toluenesulfonic acid (p-TSA),[15] boric acid,[16] and room-temperature ionic liquid,[17] as well as palladium-catalyzed multicomponent coupling,[18] palladium-catalyzed cyclization,[19] organocatalysis,[20] and solid-support synthesis.[21] Recently, substituted imidazoles have been synthesized using catalyst *L*-proline[22] and $InCl_3 \cdot H_2O$.[23]

Sulfated tin oxide was found to be efficient and suitable in aldol condensation reactions, particularly for benzoylation.[24] In continuation of our work by grinding with $I_2$[25a] and microwave irradiation using solvent polyethylene glycol,[25b] herein we report a new methodology for the synthesis of 2,4,5-triaryl-1*H*-imidazole using sulfated tin oxide (STO) catalyst and inexpensive starting materials benzil/benzoin, aromatic aldehydes, and ammonium acetate in a ethanol–water (1:1, *v*/*v*) system (Scheme 1).

## RESULTS AND DISCUSSION

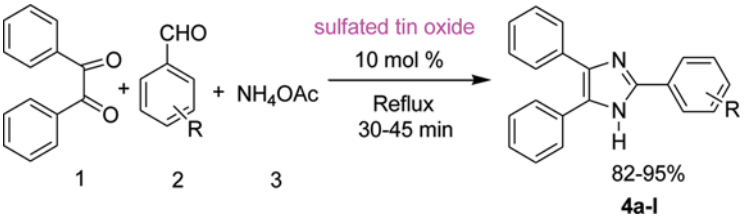
In this protocol, benzil/benzoin and ammonium acetate were refluxed with substituted aromatic aldehydes in the presence of sulfated tin oxide catalyst for an appropriate time (30–55 min) to afford substituted 2,4,5-triaryl-1*H*-imidazoles (Tables 1 and 2). More important, it can be recovered and reused several times without any change in its efficiency. The reaction was carried out using 1:1.2:4 molar ratios of reactants in ethanol–water (1:1, *v*/*v*) medium. In a similar fashion, a variety of substituted 2,4,5-triaryl-1*H*-imidazoles were also synthesized using different aldehydes. The time required for the condensation was considerably reduced, and yields of the products were excellent as compared to the reported methods. This evidence indicates that the catalyst efficiently makes the condensation reaction much faster with good yield of products.

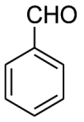
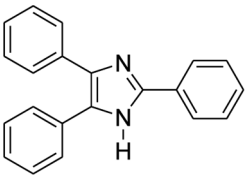
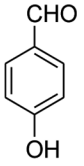
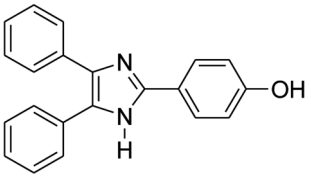
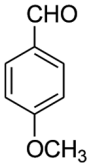
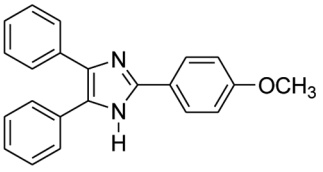
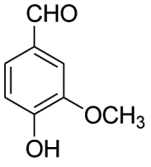
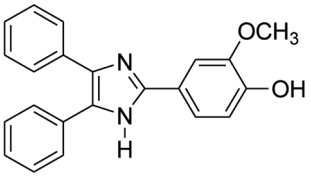
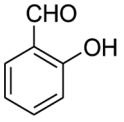
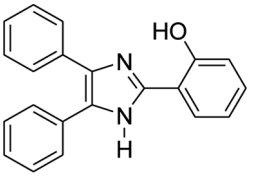
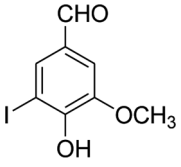
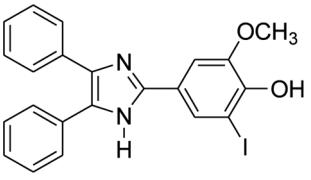
To determine the role of the catalyst, the same reaction was carried out in the absence of catalyst. The desired product was not obtained in 4–5 h. It indicates that the catalyst exhibits high catalytic activity in this transformation. The reaction




**Scheme 1.** Synthesis of 2,4,5-triaryl-1*H*-imidazole derivatives. (Figure is provided in color online.)

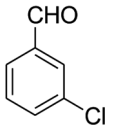
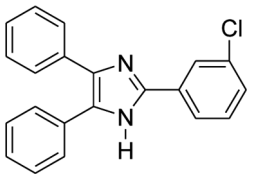
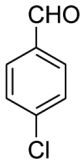
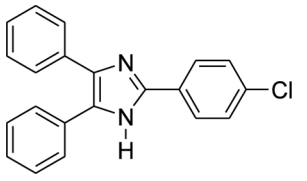
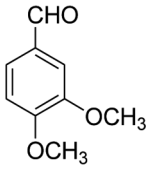
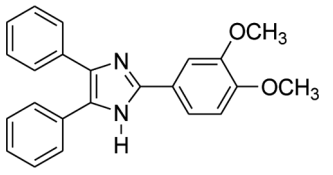
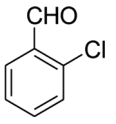
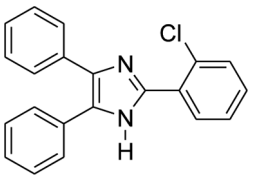
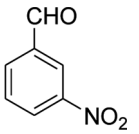
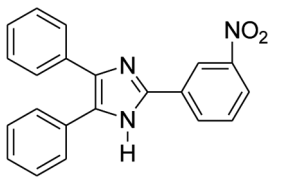
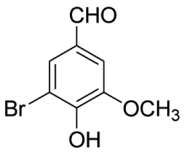
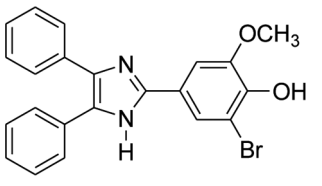
**Table 1.** One-pot synthesis of 2,4,5-triaryl-1*H*-imidazoles using benzil, aromatic aldehydes, and ammonium acetate in ethanol–water (1:1, *v*/*v*) system using sulfated tin oxide (10 mol%)




| Entries | Reactants (2) | Products (4) | Solvent system (1:1, *v*/*v*) | Reaction time (min) | Yields (%)[a] | MP (°C) [lit.] |
|---|---|---|---|---|---|---|
| a |  |  | Ethanol–water (1:1, *v*/*v*) | 30 | 92 | 276 °C [276–278][16] |
| b |  |  | Ethanol–water (1:1, *v*/*v*) | 35 | 95 | 269 °C [268–270][16] |
| c |  |  | Ethanol–water (1:1, *v*/*v*) | 30 | 89 | 229 °C [230–233][15] |
| d |  |  | Ethanol–water (1:1, *v*/*v*) | 40 | 92 | 260 °C |
| e |  |  | Ethanol–water (1:1, *v*/*v*) | 45 | 89 | 201 °C [200–203][15] |
| f |  |  | Ethanol–water (1:1, *v*/*v*) | 30 | 90 | 220 °C |

(*Continued*)

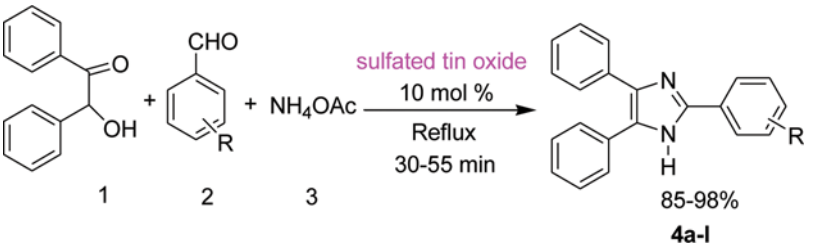
**Table 1.** Continued

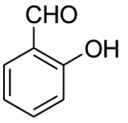
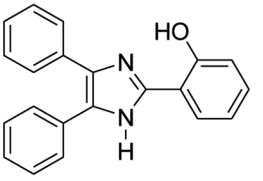
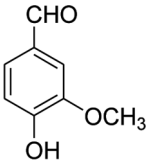
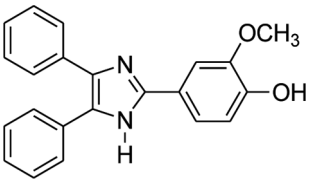
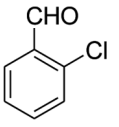
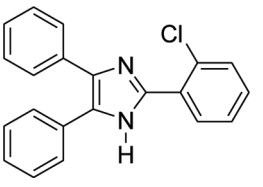
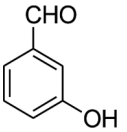
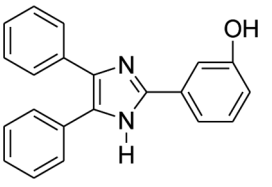
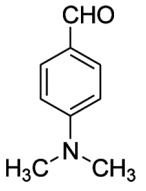
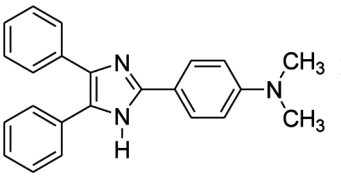
| Entries | Reactants (2) | Products (4) | Solvent system (1:1, *v*/*v*) | Reaction time (min) | Yields (%)[a] | MP (°C) [lit.] |
|---|---|---|---|---|---|---|
| g |  |  | Ethanol–water (1:1, *v*/*v*) | 30 | 91 | 261 °C |
| h |  |  | Ethanol–water (1:1, *v*/*v*) | 30 | 94 | 263 °C [260–262][16] |
| i |  |  | Ethanol–water (1:1, *v*/*v*) | 35 | 86 | 220 °C [220–221][16] |
| j |  |  | Ethanol–water (1:1, *v*/*v*) | 30 | 89 | 195 °C [195–196][16] |
| k |  |  | Ethanol–water (1:1, *v*/*v*) | 40 | 84 | 230 °C |
| l |  |  | Ethanol–water (1:1, *v*/*v*) | 35 | 82 | 262 °C |

[a]Yield of isolated products.

proceeds in solvent water, which is nontoxic, imflammable, inexpensive, and abundantly available. However, the use of only water in this reaction gave moderate yield of products (73%). Therefore, we studied the effects of different organic solvents on the synthesis of 2,4,5-triaryl-1*H*-imidazole derivatives, and the results are summarized

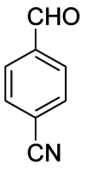
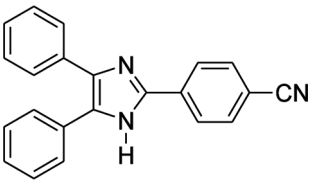
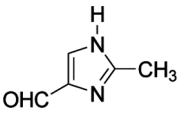
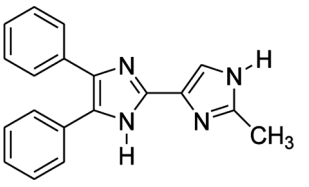
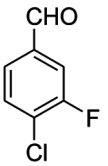
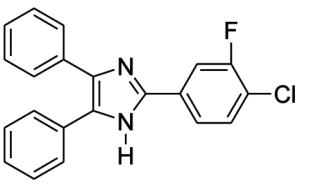
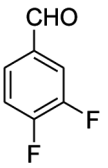
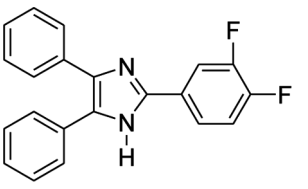
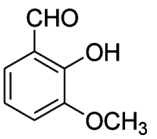
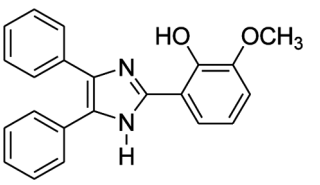
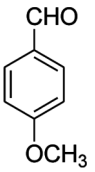
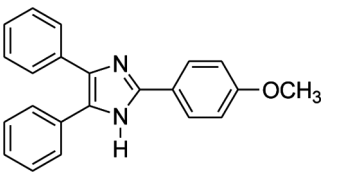
**Table 2.** One-pot synthesis of 2,4,5-triaryl-1*H*-imidazoles using benzoin, aromatic aldehydes, and ammonium acetate in ethanol–water (1:1, *v*/*v*) system using sulfated tin oxide (10 mol%)




| Entries | Reactants (2) | Products (4) | Solvent system (1:1, *v*/*v*) | Reaction time (min) | Yields (%)[a] | MP (°C) [lit.] |
|---|---|---|---|---|---|---|
| a |  |  | Ethanol–water (1:1, *v*/*v*) | 35 | 85 | 201 °C [200–203][15] |
| b |  |  | Ethanol–water (1:1, *v*/*v*) | 30 | 87 | 261 °C |
| c |  |  | Ethanol–water (1:1, *v*/*v*) | 30 | 90 | 195 °C [195–196][16] |
| d |  |  | Ethanol–water (1:1, *v*/*v*) | 35 | 98 | 203 °C |
| e |  |  | Ethanol–water (1:1, *v*/*v*) | 40 | 91 | 255 °C [256–257][15] |

(*Continued*)

**Table 2.** Continued

| Entries | Reactants (2) | Products (4) | Solvent system (1:1, *v*/*v*) | Reaction time (min) | Yields (%)[a] | MP (°C) [lit.] |
|---|---|---|---|---|---|---|
| f |  |  | Ethanol–water (1:1, *v*/*v*) | 45 | 96 | 185 °C |
| g |  |  | Ethanol–water (1:1, *v*/*v*) | 55 | 85 | 157 °C |
| i |  |  | Ethanol–water (1:1, *v*/*v*) | 40 | 86 | 215 °C |
| j |  |  | Ethanol–water (1:1, *v*/*v*) | 35 | 93 | 207 °C |
| k |  |  | Ethanol–water (1:1, *v*/*v*) | 50 | 89 | 170 °C |
| l |  |  | Ethanol–water (1:1, *v*/*v*) | 30 | 92 | 231 °C [230–233][15] |

[a]Yield of isolated products.

in Table 3. Among them, ethanol–water (1:1, *v*/*v*) was most efficient with respect to reaction time and yield of products. Both electron-donating and electron-withdrawing aromatic aldehydes offered 2,4,5-triaryl-1*H*-imidazole derivatives in good to excellent yields.

**Table 3.** Effect of various solvents for synthesis of 2,4,5-triaryl-1*H*-imidazole[a]

| Entries | Solvents | Temperature (°C) | Yields[b] (%) | Time |
|---|---|---|---|---|
| 1 | Dichloromethane | 40 | 75 | 12 h |
| 2 | Toluene | 75 | 80 | 4.0 h |
| 3 | DMF | 75 | 78 | 4.5 h |
| 4 | MeOH | 65 | 80 | 3.0 h |
| 5 | EtOH | 75 | 95 | 2.0 h |
| 6 | $CH_3CN$ | 75 | 97 | 2.0 h |
| 7 | Ethanol–water (1:1, v/v) | 80 | 80–98 | 30–55 min |

[a]Reaction conditions: 4-hydroxy benzalaldehyde (12 mmol), benzil (10 mmol), ammonium acetate (40 mmol), and sulfated tin oxide (10 mol%).
[b]Yield of isolated products.



The catalyst was separated and reused after washing with n-hexane, drying at 60 °C, and activating at 500 °C for 1 h before each catalytic run. The reusability of catalyst was investigated in the reaction of 4-hydroxybenzaldehyde with benzoin and ammonium acetate in the presence of sulfated tin oxide (10 mol%) as catalyst, which showed that the catalyst can be used three times without any loss in activity (Table 4).

## EXPERIMENTAL

### Synthesis of Sulfated Tin Oxide as Catalyst

Sulfated tin oxide was synthesized using following literature procedure.[26]

To a solution of 100 g stannous chloride ($SnCl_2$ $nH_2O$) in 300 ml water, 25% ammonia solution was added dropwise with constant stirring untill the pH of the solution was adjusted to 8. The precipitated product was collected by filtration and poured on 500 ml cold ammonium acetate solution (0.5 to 4%). The precipitated solid was filtered and dried at 100 °C for 24 h. Tin oxide gel obtained was taken in glass suction funnel, 30 ml of concentrated sulfuric acid was added slowly to it, and the mixture was allowed to stand for 1 h. The solid obtained was filtered and dried at 100 °C for 2 h. Further, the solid was heated in air at 500 °C for 3 h and stored in a sealed sample bottle until use.

### General Experimental Procedure for 2,4,5-Triaryl-1*H*-imidazoles

Aromatic aldehyde (12 mmol) was added to a mixture of benzil or benzoin (10 mmol), ammonium acetate (40 mmol), and sulfated tin oxide (10 mol %) in

**Table 4.** Recovery of sulfated tin oxide in the synthesis of 2,4,5-triaryl-*1*H-imidazole from Table 1

| | | | Yields % | | |
|---|---|---|---|---|---|
| Entry | Product | Time (min) | *Run 1* | *Run 2* | *Run 3* |
| 1 | **4b** | 35 | 95 | 92 | 89 |
| 2 | **4c** | 40 | 97 | 94 | 91 |

ethanol–water (20 ml, 1:1, *v*/*v*). The reaction mixture was heated at 80 °C under refluxed condition for appropriate times (Tables 1 and 2). After completion of reaction as confirmed by thin-layer chromatography (TLC) using eluent petroleum ether–ethyl acetate (7:3), the reaction mixture was cooled to room temperature and poured onto ice water (50 ml) to get precipitate the solid. The catalyst, sulfated tin oxide, was recovered as a residue, washed with ethanol, dried, and reused. The product was collected by filtration, washed with water, and dried to give the corresponding 2,4,5-triaryl-1*H*-imidazole derivatives.

**Synthesis of 4-(4,5-diphenyl-1H-imidazol-2-yl)phenol (4b).** A mixture of 4-hydroxybenzalaldehyde (1.46 g, 12 mmol), benzil (2.10 g, 10 mmol), ammonium acetate (3.08 g, 40 mmol), sulfated tin oxide (10 mol%), and 2 ml ethanol–water system (1:1, *v*/*v*) were taken in a round-bottom flask. The reaction mixture was heated at 80 °C under refluxed condition for an appropriate time (Table 1). After completion of the reaction as confirmed on thin-layer chromatography (TLC) using eluent (7:3 ml, petroleum ether–ethyl acetate), the reaction mixture was cooled at room temperature and poured onto ice water (50 ml) to precipitate the solid. The catalyst, sulfated tin oxide, was recovered as a residue, washed with ethanol, dried, and reused. The product was collected by filtration, washed with water, and dried to give the corresponding 4-(4,5-diphenyl-1*H*-imidazol-2-yl)phenol.



## Spectral Data of Selected Compounds

FT-IR (KBr), $^{1}$H NMR, and $^{13}$C NMR spectra were recorded at room temperature on a Varian Inova spectrometer in $CDCl_3$/dimethylsulfoixde (DMSO-$d_6$) using tetramethylsilane (TMS) as internal standard.

**2-Methoxy-4-(4,5-diphenyl-1H-imidazol-2-yl)phenol (Table 1, 4d).** IR (KBr, $cm^{-1}$): 3475 (N-H), 3360 (Ar-OH), 2923 (C-H), 1646 (C=N), 1620 (C=C), 1355 (C-N), 1237 (C-O); $^{1}$H NMR ($CDCl_3$/DMSO-$d_6$, 400 MHz, δ ppm): 3.67 (s, 3H, -$OCH_3$), 5.01 (s, 1H, -OH), 6.61 (dd, 1H, Ar-H), 6.85 (s, 1H, Ar-H), 7.06 (dd, 1H, Ar-H), 7.05–7.25 (m, 10H, Ar-H), 12.24 (brs, 1H, -NH); $^{13}$C NMR (100 MHz, $CDCl_3$/DMSO-$d_6$): 55.7, 111.5, 115.9, 121.4, 123.9, 125.5, 127.5, 128.9, 129.1, 129.7, 132.7, 146.1, 152.1; LCMS: 344.15 [$M^+$], 345.29 [M + 1].

**2-Iodo-6-methoxy-4-(4,5-diphenyl-1H-imidazol-2-yl)phenol (Table 1, 4f).** IR (KBr, $cm^{-1}$): 3478 (N-H), 3345 (br, O-H), 2965 (C-H), 1590 (C=C), 1655 (C=N), 1250(C-O), 545 (C-I); $^{1}$H NMR ($CDCl_3$/DMSO-$d_6$, 400 MHz, δ ppm): 3.67 (s, 3H, -$OCH_3$), 5.01 (s, 1H, -OH), 6.75 (s, 1H, Ar-H), 7.35 (s, 1H, Ar-H), 7.05–7.15 (m, 10H, Ar-H), 12.27 (brs, s, 1H, -NH); $^{13}$C NMR (100 MHz, $CDCl_3$/DMSO-$d_6$): 56.2. 88.0, 110.9, 126.0, 127.2, 128.9, 129.1, 129.7, 130.2, 132.8, 148.0, 151.8, 154.1; LCMS: 368.03 [$M^+$], 369.05 [M + 1].

**2-(3-Chlorophenyl)-4,5-diphenyl-1H-imidazole (Table 1, 4g).** IR (KBr, $cm^{-1}$): 3465 (N-H), 2950 (C-H), 1669 (C=N), 1345 (C-N), 1590 (C=C),1085 (C-Cl); $^{1}$H NMR ($CDCl_3$/DMSO-$d_6$, 400 MHz, δ ppm): 7.19 (dd, 1H, Ar-H), 7.30 (dd, 1H, Ar-H), 7.40 (dd, 1H, Ar-H), 7.43 (s, 1H, Ar-H), 7.05–7.19 (m, 10H, Ar-H), 12.25 (brs, s, 1H, -NH); $^{13}$C NMR (100 MHz, $CDCl_3$/DMSO-$d_6$): 125.9,

127.2, 132.8, 128.9, 129.0, 129.1, 129.7, 131.0, 132.7, 132.8, 133.0, 148.0; LCMS: 330.81, [$M^+$], 331.21, isotopic peak-333.16, 334.21.

**2-(4-Chlorophenyl)-4,5-diphenyl-1H-imidazole (Table 1, 4h).** IR (KBr, $cm^{-1}$): 3465 (N-H), 2950 (C-H), 1670 (C=N), 1345 (C-N), 1593 (C=C), 1082 (C-Cl); $^1$H NMR ($CDCl_3$/DMSO-$d_6$, 400 MHz, δ ppm): 7.25 (dd, 2H, Ar-H), 7.07–7.25 (m, 10H, Ar-H), 7.60 (dd, 2H, Ar-H), 12.24 (brs, s, 1H, -NH); $^{13}$C NMR (100 MHz, $CDCl_3$/DMSO-$d_6$): 127.2, 128.9, 129.1, 129.3, 129.7, 130.1, 132.8, 135.0, 148.1; LCMS: 330.81 [$M^+$], 331.12 [M + 1], [M + 1], isotopic peaks 333.14, 334.18.

**2-(3-Nitrophenyl)-4,5-diphenyl-1H-imidazole (Table 1, 4k).** IR (KBr, $cm^{-1}$): 3424 (N-H), 2955 (C-H), 1659 (C=N), 1385 (C-N), 1535 (C=C), 1520 (Ar-N-O); $^1$H NMR ($CDCl_3$/DMSO-$d_6$, 400 MHz, δ ppm): 7.62 (dd, 1H, Ar-H), 7.75 (dd, 1H, Ar-H), 7.02–7.20 (m, 10H, Ar-H), 8.05 (dd, 1H, Ar-H), 8.45 (s, 1H, Ar-H), 12.30 (brs, s, 1H, -NH).

**2-Bromo-6-methoxy-4-(4,5-diphenyl-1H-imidazol-2-yl)phenol (Table 1, 4l).** IR (KBr, $cm^{-1}$): 3479 (N-H), 3357 (br, Ar-OH), 2972 (C-H), 1556 (C=C), 1637 (C=N), 1265 (C-O), 1078 (C-Br); $^1$H NMR ($CDCl_3$/DMSO-$d_6$, 400 MHz, δ ppm): 3.63 (s, 3H, -$OCH_3$), 5.07 (s, 1H, -OH), 6.80 (s, 1H, Ar-H), 7.00 (s, 1H, Ar-H), 7.30–7.55 (m, 10H, Ar-H), 12.35 (brs, s, 1H, -NH).

**3-(4,5-Diphenyl-1H-imidazol-2-yl)phenol (Table 2, 4d).** IR (KBr, $cm^{-1}$): 3492 (N-H), 3378 (Ar-OH), 2945 (C-H), 1633 (C=N), 1554 (C=C), 1380 (C-N), 1242 (C-O); $^1$H NMR ($CDCl_3$/DMSO-$d_6$, 400 MHz, δ ppm): 6.57 (dd, 1H, Ar-H), 6.75 (s, 1H, Ar-H), 7.00 (dd, 1H, Ar-H), 7.10 (dd, 1H, Ar-H), 7.05–7.17 (m, 10H, Ar-H), 12.21 (brs, s, 1H, -NH); $^{13}$C NMR (100 MHz, $CDCl_3$/DMSO-$d_6$): 113.1, 116.2, 119.4, 127.2, 128.9, 129.1, 129.7, 130.3, 131.8, 132.8, 148.1, 158.5.

**4-(4,5-Diphenyl-1H-imidazol-2-yl)benzonitrile (Table 2, 4f).** IR (KBr, $cm^{-1}$): 3407 (N-H), 2921 (C-H), 2240 (Ar-CN), 1649 (C=N), 1555 (C=C), 1675 (C = N), 1240 (C-N); $^1$H NMR ($CDCl_3$/DMSO-$d_6$, 400 MHz, δ ppm): 7.01–7.31 (m, 10H, Ar-H), 7.47 (dd, 2H, Ar-H), 7.95 (dd, 2H, Ar-H), 12.10 (brs, s, 1H, -NH); $^{13}$C NMR (100 MHz, $CDCl_3$/DMSO-$d_6$): 113.1, 116.0, 127.2, 127.6, 128.4, 128.9, 129.1, 129.7, 131.0, 132.8, 133.5, 137.0, 148.1; LCMS: 321.13 [$M^+$], 322.13 [M + 1].

**2-(2-Methyl-1H-imidazol-4-yl)-4,5-diphenyl-1H-imidazole (Table 2, 4g).** IR (KBr, $cm^{-1}$): 3436 (N-H), 2922 (C-H), 1659 (C=N), 1540 (C=C), 1384 (C-N); $^1$H NMR ($CDCl_3$/DMSO-$d_6$, 400 MHz, δ ppm): 2.40 (s, 3H, -$CH_3$), 7.09–7.95 (m, 10H, Ar-H), 6.91 (s, 1H), 12.05 (brs, s, 1H, -NH), 13.57 (s, 1H, -NH); $^{13}$C NMR (100 MHz, $CDCl_3$/DMSO-$d_6$): 17.9, 120.0, 121.1, 127.2, 128.9, 129.1, 129.7, 132.8, 144.0, 165.5; LCMS: 300.36 [$M^+$], 301.36 [M + 1].

**2-(4-Chloro-3-fluorophenyl)-4,5-diphenyl-1H-imidazole (Table 2, 4i).** IR (KBr, $cm^{-1}$): 3445 (N-H), 2920 (C-H), 1659 (C=N), 1639 (C-N), 1540 (C=C), 1170 (C-F), 1075 (C-Cl); $^1$H NMR ($CDCl_3$/DMSO-$d_6$, 400 MHz, δ ppm): 6.08 (dd, 1H, Ar-H), 7.20 (s, 1H, Ar-H), 7.50 (dd, 1H, Ar-H), 7.09–7.73 (m, 10H, Ar-H), 12.47 (s, 1H, -NH); $^{13}$C NMR (100 MHz, $CDCl_3$/DMSO-$d_6$): 116.9, 121.2,

125.0, 127.2, 128.9, 129.1, 129.7, 130.7, 132.0, 132.8, 148.1, 164.0; LCMS: 348.80 [$M^+$], 349.80 [M + 1].

**2-(3,4-Difluorophenyl)-4,5-diphenyl-1H-imidazole (Table 2, 4j).** IR (KBr, $cm^{-1}$): 3465 (N-H), 2985 (C-H), 1615 (C=N), 1575 (C=C), 1175 (C-F); $^1$H NMR ($CDCl_3$/DMSO-$d_6$, 400 MHz, δ ppm): 7.01 (dd, 1H, Ar-H), 7.12 (s, 1H, Ar-H), 7.47 (dd, 1H, Ar-H), 7.09–7.72 (m, 10H, Ar-H), 12.45 (s, 1H, -NH).

**2-(4,5-Diphenyl-1H-imidazol-2-yl)-6-meyhoxy phenol (Table 2, 4k).** IR (KBr, $cm^{-1}$): 3412 (N-H), 3350 (-OH), 2925 (C-H), 1654 (C=C), 1565 (C=N), 1335 (C-N); $^1$H NMR ($CDCl_3$/DMSO-$d_6$, 400 MHz, δ ppm): 3.68 (s, 3H, -$OCH_3$), 5.01 (s, 1H, -OH), 6.59 (dd, 1H, Ar-H), 6.77 (dd, 1H, Ar-H), 6.80 (dd, 1H, Ar-H), 7.05–7.25 (m, 10H, Ar-H), 12.21 (brs, 1H, -NH); $^{13}$C NMR (100 MHz, $CDCl_3$/DMSO-$d_6$): 54.7, 110.9, 112.1, 115.6, 117.1 121.1, 126.2, 127.3, 127.4, 129.8, 145.1, 146.1, 147.3; LCMS: 342.14 [$M^+$], 343.14 [M + 1].

## CONCLUSION

In summary, we developed a simple, efficient, clean methodology and mild protocol for the synthesis of 2,4,5-triary-1*H*-imidazole derivatives by condensation of benzoin/benzil, substituted aromatic aldehydes, and ammonium acetate in the presence of sulfated tin oxide (STO) solid heterogeneous catalyst. In this procedure, ethanol–water (1:1, *v*/*v*) is used as solvent and therefore is relatively ecofriendly. The notable merits of this method are short reaction times, simple workup procedure, excellent yield of products, nontoxicity, cost-efficiency, and reusability of the catalyst.

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