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Zinc (II) [*tetra*(4-methylphenyl)] Porphyrin: a Novel and Reusable Catalyst for Efficient Synthesis of 2,4,5-trisubstituted Imidazoles Under Ultrasound Irradiation

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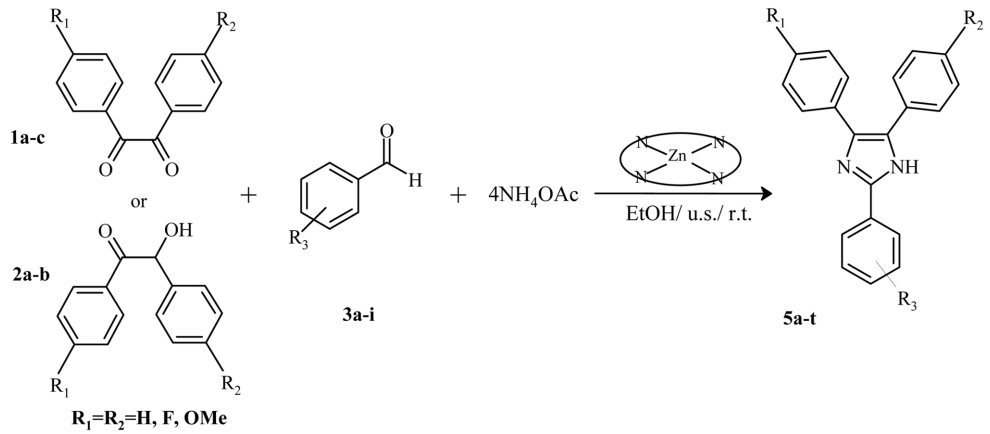
ABSTRACT: An efficient three-component one-step synthesis of 2,4,5-trisubstituted imidazoles by condensation reaction of 1,2-diketones or α-hydroxyketones with aromatic aldehydes and ammonium acetate using Zinc (II) [tetra (4-methylphenyl)] porphyrin as a novel and reusable catalyst under ultrasound irradiation at ambient temperature is described. In this method, α-hydroxyketones as well as 1,2-diketones were converted to their corresponding 2,4,5-trisubstituted imidazoles in excellent yields.

Key words: Three-component condensation, 2,4,5-Trisubstituted imidazoles, Zinc (II) [tetra (4-methylphenyl)] porphyrin, Ultrasound irradiation

INTRODUCTION

Multicomponent reactions (MCRs) have drawn great interest enjoying an outstanding status in modern organic synthesis and medicinal chemistry because they are one-pot processes bringing together three or more components and show high atom economy and high selectivity.[1-4] MCRs have great contribution in convergent synthesis of complex and important organic molecules from simple and readily available starting materials, and have emerged as powerful tools for drug discovery.[5-7] Ultrasound has increasingly been used in organic synthesis. A large number of ultrasonic reactions can be carried out in higher yield, shorter reaction time or milder conditions.[8-11] The phenomenon responsible for the beneficial effects of ultrasound on chemical reactions is cavitation. During the rarefaction cycle of the wave, the molecules of the liquid are separated, generating bubbles that subsequently collapse in the compression cycle.[12] These rapid and violent implosions generate short-lived regions with temperatures of roughly 5000 ºC, pressures of about 1000 atm and heating and cooling rates above 10 billion ºC per second.[13] Such localized hot spots can be thought as microreactors in which the energy of sound is transformed into a useful chemical form.

The imidazole ring system is one of the most important substructures found in a large number of natural products and pharmacologically active compounds.[14] Multisubstituted imidazoles have received much attention because of their wide range of pharmaceutical and biological properties such as antitumor,[15] anti-inflammatory,[16,17] anti-allergic,[18] analgesic[19] and antibacterial effects.[20] Various substituted imidazoles act as inhibitors of p38 MAP kinase[21] and B-Raf kinase[22] and glucagon receptors.[23] Accordingly, a number of synthetic methods have been reported for the construction of this important structure. Generally 2,4,5-trisubstituted imidazoles are synthesized by three-component cyclocondensation of 1,2-diketone, α-hydroxyketone or α-ketomonoxime with an aldehyde and ammonium acetate. A variety of catalysts including protonic and Lewis acid catalysts have been introduced for synthesis of 2,4,5-trisubstituted imidazoles such as H_2SO_4,[24] H_3PO_4,[25] $NiCl_2{\cdot}6H_2O$,[26] $Yb(OTf)_3$,[27] $InCl_3{\cdot}3H_2O$,[28] I_2,[29] polymer-supported $ZnCl_2$,[30] oxalic acid,[31] boric acid,[32] *p*-toluenesulfonic acid,[33] L-proline,[34] L-proline triflate,[35] $Cu(NO_3)_2$-zeolite,[36] and ionic liquids.[37-39] Despite their potential utility, most of these methods have a number of drawbacks including poor yields, high temperature, use of toxic catalyst, strongly acidic conditions and the requirement for harsh reaction conditions. Therefore, the development of facile and environmentally benign method for the synthesis of biologically active 2,4,5-trisubstituted imidazoles is the necessary part of organic synthesis. Electron-deficient metalloporphyrins have been used as mild and non-toxic Lewis acid catalysts.[40-44] In continuation of our interest on the synthesis of 2,4,5-trisubsti-



***Scheme* 1.** One-pot synthesis of 2,4,5-trisubstituted imidazoles catalyzed by [ZnT(4-CH_3)PP] under ultrasound irradiation at ambient temperature.

tuted imidazoles,[45,46] we decided to explore ability of metalloporphyrins in the synthesis of 2,4,5-trisubstituted imidazoles for the first time. Therefore, Zinc (II) [tetra (4-methylphenyl)] porphyrin was used as a mild Lewis acid catalyst for the three-component one-step synthesis of 2,4,5-trisubstituted imidazoles by condensation reaction of 1,2-diketones or α-hydroxyketones with aromatic aldehydes and ammonium acetate under ultrasound irradiation at ambient temperature (*Scheme* 1).

EXPERIMENTAL

Chemical reagents in high purity were purchased from the Merck Chemical Company. All materials were of commercial reagent grade. Zinc (II) [tetra (4-methylphenyl)] porphyrin was prepared and metallated according to the literature.[47,48] Melting points were determined in open capillaries using an Electrothermal Mk3 apparatus and are uncorrected. FT-IR spectra were obtained with potassium bromide pellets in the range 400-4000 cm^{-1} with a Perkin-Elmer 550 spectrometer. ^{1}H NMR and ^{13}C NMR spectra were recorded with a Bruker DRX-400 spectrometer at 400 and 100 MHz respectively. NMR spectra were obtained in DMSO-d_6 solutions and are reported as parts per million (ppm) downfield from tetramethylsilane as internal standard. The abbreviations used are: singlet (s), doublet (d), triplet (t) and multiplet (m). Elemental analysis (C, H, N) was performed with a Carlo Erba Model EA 1108 analyzer or a Perkin-Elmer 240c analyzer, and results agreed favorably with calculated values. The UV-vis measurements were obtained with a GBC cintra 6 UV-vis spectrophotometer. Ultrasonication was performed in a EUROSONIC® 4D ultrasound cleaner with a frequency of 50 kHz and an output power of 350 W. The reaction ask was located in the maximum energy area in the cleaner, where the surface of reactants (reaction vessel) is slightly lower than the level of the water and the temperature of the water bath was controlled at 25 °C.

General procedure for the synthesis of 2,4,5-trisubstituted imidazoles catalyzed by [ZnT(4-CH_3)PP] under ultrasound irradiation

A mixture of benzoin or benzil derivatives (1 mmol), aldehyde (1 mmol), ammonium acetate (0.31 g, 4 mmol) and [ZnT(4-CH_3)PP] (3 μmol %) in 10 ml ethanol was taken in a 50 ml conical flask and the reaction mixture was irradiated in the water bath of the ultrasonic cleaner at 25 °C for a period as indicated in *Table* 2 and *Table* 3. The progress of reaction was followed by TLC. After completion of reaction, the solvent was evaporated, then the solid residue was purified by column chromatography (eluent petroleum ether: ethyl acetate=9:2) on silica gel to give analytically pure products in good to excellent yield and were fully characterized by spectroscopic methods such as IR, ^{1}H NMR, ^{13}C NMR and element analyses (C, H, N) and have been identified by the comparison of the reported spectral data. The spectral data for new compounds are presented below.

Spectroscopic data for new 2,4,5-trisubstituted imidazoles

3-[4,5-bis(4-methoxyphenyl)-1H-imidazol-2-yl]phenol (**5n**). Cream solid. IR (KBr) (υ_{max}/cm^{-1}): 3415 (N-H), 3338 (O-H), 1613 (C=C), 1500 (C=N), 1247 (C-O); ^{1}H NMR (400 MHz, DMSO-d_6): δ_H 12.42 (s, 1H, NH), 9.52 (s, 1H, OH), 7.38-7.49 (m, 6H, Ar-H), 7.23 (t, 1H, *J*=8.0

Hz, Ar-H), 7.00 (d, 2H, *J*=8.4 Hz, Ar-H), 6.86 (d, 2H, *J*= 8.4 Hz, Ar-H), 6.74 (dd, 1H, *J*=8.0, 2.8 Hz, Ar-H), 3.78 (s, 3H, OMe), 3.73 (s, 3H, OMe) ppm; ^{13}C NMR (100 MHz, DMSO-d_6): δ_C 159.2, 158.4, 158.0, 145.4, 136.7, 132.2, 130.2, 130.1, 128.6, 128.4, 127.5, 124.0, 116.4, 115.6, 114.5, 114.1, 112.5, 55.6, 55.5 ppm; Anal. Calcd. for $C_{23}H_{20}N_2O_3$: C, 74.18; H, 5.41; N, 7.52 %. Found: C, 74.16; H, 5.39; N, 7.49%.

2-(3-nitrophenyl)-4,5-bis(4-methoxyphenyl)-1H-imidazole (**5o**). Yellow solid. IR (KBr) (υ_{max}/cm^{-1}): 3428 (N-H), 1615 (C=C), 1523 (C=N), 1460 (N=O), 1348 (N-O), 1249 (C-O); ^{1}H NMR (400 MHz, DMSO-d_6): δ_H 12.92 (s, 1H, NH), 8.92 (s, 1H, Ar-H), 8.50 (d, 1H, *J*=8.2 Hz, Ar-H), 8.20 (d, 1H, *J*=8.2 Hz, Ar-H), 7.75 (t, 1H, *J*=8.2 Hz, Ar-H), 7.47 (d, 2H, *J*=8.4 Hz, Ar-H), 7.43 (d, 2H, *J*=8.4 Hz, Ar-H), 7.00 (d, 2H, *J*=8.4 Hz, Ar-H), 6.90 (d, 2H, *J*= 8.4 Hz, Ar-H), 3.80 (s, 3H, OMe), 3.75 (s, 3H, OMe) ppm; ^{13}C NMR (100 MHz, DMSO-d_6): δ_C 159.4, 158.6, 148.8, 143.1, 137.5, 132.5, 131.4, 130.8, 130.2, 128.8, 128.7, 127.9, 123.5, 122.7, 119.7, 114.6, 114.1, 55.7, 55.5 ppm; Anal. Calcd. for $C_{23}H_{19}N_3O_4$: C, 68.82; H, 4.77; N, 10.47%. Found: C, 68.79; H, 4.75; N, 10.44%.

2-(3-methoxyphenyl)-4,5-bis(4-methoxyphenyl)-1H-imidazole (**5p**). White solid. IR (KBr) (υ_{max}/cm^{-1}): 3430 (N-H), 1608 (C=C), 1519 (C=N), 1246 (C-O); ^{1}H NMR (400 MHz, DMSO-d_6): δ_H 12.50 (s, 1H, NH), 7.64 (d, 1H, *J*=8.0 Hz, Ar-H), 7.62 (s, 1H, Ar-H), 7.36-7.46 (m, 5H, Ar-H), 7.00 (d, 2H, *J*=8.4 Hz, Ar-H), 6.91 (dd, 1H, *J*=8.4, 2.2 Hz, Ar-H), 6.87 (d, 2H, *J*=8.4 Hz, Ar-H), 3.82 (s, 3H, OMe), 3.79 (s, 3H, OMe), 3.74 (s, 3H, OMe) ppm; ^{13}C NMR (100 MHz, DMSO-d_6): δ_C 160.0, 158.9, 158.2, 145.1, 136.2, 132.3, 130.2, 130.2, 128.6, 128.2, 127.2, 124.1, 117.0, 115.5, 114.4, 114.3, 110.5, 55.6, 55.5, 55.5 ppm; Anal. Calcd. for $C_{24}H_{22}N_2O_3$: C, 74.59; H, 5.74; N, 7.25%. Found: C, 74.58; H, 5.75; N, 7.24%.

5-[4,5-bis(4-methoxyphenyl)-1H-imidazol-2-yl]-2-methoxyphenol (**5q**). Ash-gray solid. IR (KBr) (υ_{max}/cm^{-1}): 3424 (N-H), 3320 (O-H), 1615 (C=C), 1504 (C=N), 1249 (C-O); ^{1}H NMR (400 MHz, DMSO-d_6): δ_H 12.26 (s, 1H, NH), 9.11 (s, 1H, OH), 7.52 (s, 1H, Ar-H), 7.37-7.46 (m, 6H, Ar-H), 6.98 (d, 2H, *J*= 8.4 Hz, Ar-H), 6.86 (d, 2H, *J*= 8.4 Hz, Ar-H), 3.80 (s, 3H, OMe), 3.78 (s, 3H, OMe), 3.73 (s, 3H, OMe) ppm; ^{13}C NMR (100 MHz, DMSO-d_6): δ_C 159.1, 158.7, 148.4, 146.9, 145.6, 129.8, 129.2, 128.4, 126.3, 126.0, 123.5, 124.1, 116.8, 114.3, 114.0, 113.2, 112.5, 56.1, 55.5, 55.1 ppm; Anal. Calcd. for $C_{24}H_{22}N_2O_4$: C, 71.63; H, 5.51; N, 6.96%. Found: C, 71.61; H, 5.49; N, 6.95%.

2-(3,5-dimethoxyphenyl)-4,5-bis(4-methoxyphenyl)-1H-imidazole (**5r**). White solid. IR (KBr) (υ_{max}/cm^{-1}): 3420 (N-H), 1604 (C=C), 1519 (C=N), 1248 (C-O); ^{1}H NMR (400 MHz, DMSO-d_6): δ_H 12.50 (s, 1H, NH), 7.45 (d, 2H, *J*=7.6 Hz, Ar-H), 7.40 (d, 2H, *J*=8.0 Hz, Ar-H), 7.26 (s, 2H, Ar-H), 7.0 (d, 2H, *J*=8.0 Hz, Ar-H), 6.87 (d, 2H, *J*= 8.0 Hz, Ar-H), 6.48 (s, 1H, Ar-H), 3.80 (s, 3H, OMe), 3.79 (s, 3H, OMe), 3.73 (s, 6H, OMe), ppm; ^{13}C NMR (100 MHz, DMSO-d_6): δ_C 161.2, 158.9, 158.1, 145.1, 137.0, 134.3, 132.8, 130.1, 128.8, 128.6, 124.0, 114.3, 114.1, 103.4, 100.8, 55.8, 55.5 ppm; Anal. Calcd. for $C_{25}H_{24}N_2O_4$: C, 72.10; H, 5.81; N, 6.73%. Found: C, 72.09; H, 5.79; N, 6.72%.

2-(4-methoxyphenyl)-4,5-bis(4-fluorophenyl)-1H-imidazole (**5s**). White solid. IR (KBr) (υ_{max}/cm^{-1}): 3427 (N-H), 1613 (C=C), 1504 (C=N), 1248 (C-O); ^{1}H NMR (400 MHz, DMSO-d_6): δ_H 12.53 (s, 1H, NH), 8.0 (d, 2H, *J*=8.4 Hz, Ar-H), 7.48-7.53 (m, 4H, Ar-H), 7.28 (t, 2H, *J*=8.8 Hz, Ar-H), 7.14 (t, 2H, *J*=8.8 Hz, Ar-H), 7.03 (d, 2H, *J*= 8.4 Hz, Ar-H), 3.80 (s, 3H, OMe) ppm; ^{13}C NMR (100 MHz, DMSO-d_6): δ_C 163.0 (C-F, d, *J*=243.0 Hz), 160.6 (C-F, d, *J*=241.0 Hz), 159.0, 146.2, 146.2, 133.5 (d, *J*= 10.0 Hz), 130.2 (d, *J*=9.0 Hz), 129.5, 129.3, 127.5, 123.5, 117.2 (d, *J*=22.0 Hz), 115.8 (d, *J*=21.0 Hz), 114.5, 112.3, 55.6 ppm; Anal. Calcd. for $C_{22}H_{16}F_2N_2O$: C, 72.92; H, 4.45; N, 7.73%. Found: C, 72.89; H, 4.43; N, 7.71%.

3-[4,5-bis(4-fluorophenyl)-1H-imidazol-2-yl]phenol (**5t**). Golden crystalline solid. IR (KBr) (υ_{max}/cm^{-1}): 3437 (N-H), 3300 (O-H), 1605 (C=C), 1517 (C=N); ^{1}H NMR (400 MHz, DMSO-d_6): δ_H 12.62 (s, 1H, NH), 9.54 (s, 1H, OH), 7.45-7.58 (m, 6H, Ar-H), 7.30 (t, 2H, *J*=7.6 Hz, Ar-H), 7.24 (d, 1H, *J*=8.0 Hz, Ar-H), 7.15 (t, 2H, *J*=8.3 Hz, Ar-H), 6.78 (d, 1H, *J*=8.0 Hz, Ar-H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6): δ_C 162.2 (C-F, d, *J*=243.0 Hz), 161.6 (C-F, d, *J*=243.0 Hz), 160.1, 145.9, 136.7, 132.0, 131.9, 131.1 (d, *J*=7.0 Hz), 130.3, 129.4 (d, *J*=7.0 Hz), 127.8, 127.6, 118.1, 116.1 (d, *J*=21.0 Hz), 115.6 (d, *J*=22.0 Hz), 114.7, 110.7 ppm; Anal. Calcd. for $C_{21}H_{14}F_2N_2O$: C, 72.41; H, 4.05; N, 8.04%. Found: C, 72.38; H, 4.04; N, 8.02%.

RESULTS AND DISCUSSION

To achieve suitable conditions for the synthesis of 2,4,5-trisubstituted imidazoles, various reaction conditions have been investigated in the reaction of 4-methoxybenzaldehyde **3b**, benzil **1a**, and ammonium acetate as a model reaction. We examined the effect of different solvents such as water, EtOH, THF, DMF, CH_3CN, and $CHCl_3$ on a model reaction under ultrasound irradiation at 25 °C (*Table* 1, entry 1-6). The reaction using EtOH as solvent gave the best result and **5b** was obtained in 80% yield

***Table* 1.** Optimization of the reaction conditions for synthesis of 2,4,5-trisubstituted imidazoles[a]

Entry	Solvent	Method	Catalyst (μmol %)	Time (min)	Yield[b] (%)
1	Water	Ultrasound	2.0	70	20
2	EtOH	Ultrasound	2.0	70	80
3	THF	Ultrasound	2.0	70	42
4	DMF	Ultrasound	2.0	70	35
5	CH_3CN	Ultrasound	2.0	70	40
6	CH_3Cl	Ultrasound	2.0	70	53
7	EtOH	High speed stirring	2.0	120	35
8	EtOH	Ultrasound	None	70	42
9	EtOH	Ultrasound	1.0	70	70
10	EtOH	Ultrasound	1.5	70	77
11	EtOH	Ultrasound	2.5	70	88
12	EtOH	Ultrasound	3.0	70	96
13	EtOH	Ultrasound	4.0	70	96

[a]Benzil (1 mmol), 4-methoxybenzaldehyde (1 mmol), NH_4OAc (4 mmol), [b]Isolated yield based on aldehyde.

within 70 min (*Table* 1, entry 2). To demonstrate the effect of ultrasound, the synthesis of **5b** as a model was investigated under high speed stirring conditions. The reaction failed to give low yield under high speed stirring conditions (*Table* 1, entry 7). It was apparent that the ultrasound irradiation accelerates this transformation under ambient conditions. To delineate the role of catalyst, the reaction was investigated with and without catalyst. As shown, in the absence of catalyst the yield of the product was found to be low (*Table* 1, entry 8). On the other hand, ultrasound irradiation and the catalyst have a great influence on the model reaction. As indicated (*Table* 1, entry 12) the best result have been obtained with amount of 3 μmol% [ZnT(4-CH_3)PP] in EtOH under ultrasound irradiation at ambient temperature and the yield of reaction with increasing the amount of [ZnT(4-CH_3)PP] is not increased (*Table* 1,

***Table* 2.** One-pot synthesis of of 2,4,5-trisubstituted imidazoles catalyzed by [ZnT(4-CH_3)PP] under ultrasound irradiation at ambient temperature[a]

Entry	Benzil	R_1, R_2	Aldehyde	R_3	Time (min)	Product	Yield[b] (%)	M.p. C (lit)
1	1a	H	3a	H	70	5a	94	270-272 (272-273)[29]
2	1a	H	3b	*p*-OMe	70	5b	96	228-231 (230-232)[17]
3	1a	H	3c	*p*-Me	70	5c	95	230-233 (232-235)[17]
4	1a	H	3d	*m*-Br	70	5d	93	301-303 (303-304)[17]
5	1a	H	3e	*m*-OH	70	5e	92	259 (260-261)[37]
6	1a	H	3f	*m*-NO_2	70	5f	90	269-271 (265-267)[29]
7	1a	H	3g	*m*-OMe	70	5g	92	259-262 (259-261)[46]
8	1a	H	3h	*m*-OH, *p*-OMe	70	5h	97	214-216 (215-216)[46]
9	1a	H	3i	*m*-OMe, *m*-OMe	70	5i	93	256-257 (254-256)[46]
10	1b	OMe	3a	H	80	5j	90	201.5-203 (202-204)[17]
11	1b	OMe	3b	*p*-OMe	80	5k	93	183-185 (184-186)[17]
12	1b	OMe	3c	*p*-Me	80	5l	93	186-188 (185-187)[17]
13	1b	OMe	3d	*m*-Br	80	5m	90	248-251 (250-252)[17]
14	1b	OMe	3e	*m*-OH	80	5n	91	230-232
15	1b	OMe	3f	*m*-NO_2	80	5o	88	240-242
16	1b	OMe	3g	*m*-OMe	80	5p	91	234-236
17	1b	OMe	3h	*m*-OH, *p*-OMe	80	5q	95	132-134
18	1b	OMe	3i	*m*-OMe, *m*-OMe	80	5r	92	195-197
19	1c	F	3b	*p*-OMe	50	5s	98	248-250
20	1c	F	3e	*m*-OH	50	5t	95	271-273

[a]Benzil (1 mmol), Aldehyde (1 mmol), NH_4OAc (4 mmol), [ZnT(4-CH_3)PP] (3 μmol %), [b]Isolated yield based on aldehyde.

entry 13).

In order to evaluate the generality of the process, several diversified examples illustrating the present method for the synthesis of 2,4,5-trisubstituted imidazoles **5a-t** were studied. The reaction of benzil with various aromatic aldehydes bearing electron-withdrawing groups or electron-releasing groups and ammonium acetate was carried out in the presence of [ZnT(4-CH_3)PP] as catalyst under ultrasound irradiation at ambient temperature (*Table* 2). As shown the process tolerates both electron withdrawing and electron donating substituents on the aldehydes. The aryl group substituted with different positions of the aromatic ring has not shown much effect on the formation of the final product. We also concentrated our study on different benzils (4,4'-difluorobenzil, 4,4'-dimethoxybenzil) (*Table* 2). The results illustrate the high ability of this method for the synthesis of 2,4,5-triarylimidazoles with different groups.

The reusability of the [ZnT(4-CH_3)PP] catalyst was studied using multiple synthesis of 2,4,5-trisubstituted imidazoles. The results showed that after reusing the catalyst for several times, no change was observed in its catalytic activity. The nature of the recovered catalyst was monitored by its UV-Vis spectrum *Fig*. 1, in which no change was observed in its spectrum and since the catalyst retained its activity in the condensation reaction.

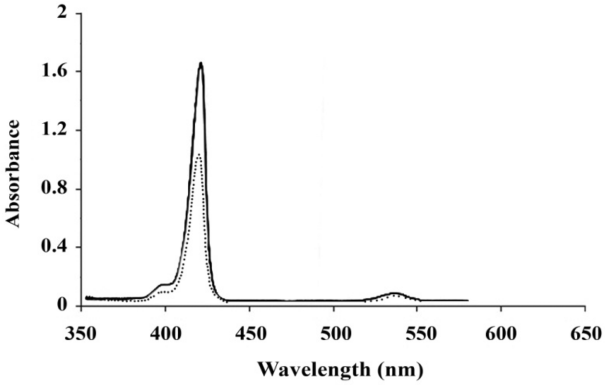
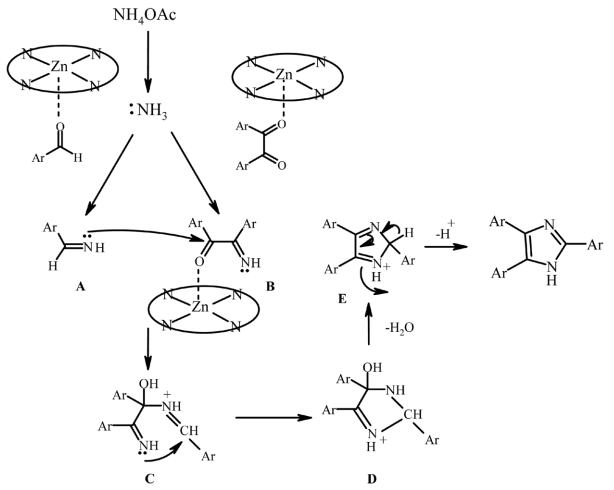


***Fig.* 1.** UV-Vis spectrum of [ZnT(4-CH_3)PP]: before reaction (solid line) and after reaction (dashed line).



***Scheme* 2.** Plausible mechanism.

We have not established an exact mechanism for the formation of 2,4,5-trisubstituted imidazoles, however, a reasonable possibility is shown in *Scheme* 2. A plausible mechanism for these reactions is that aldehyde and 1,2-diketone are first activated by [ZnT(4-CH_3)PP] to afford A and B respectively. Then, imine intermediate (A), condenses further with the carbonyl carbon of 1,2 diketone imine (B) and formation of carbocation (C) followed by attack imine nitrogen to positive center and dehydration to afford the iso-imidazole (E), which rearranges via [1,5] sigmatropic shift to the required imidazole (*Scheme* 2).

The high catalytic activity of [ZnT(4-CH_3)PP] catalyst in the synthesis of trisubstituted imidazoles by condensation reaction of benzil derivatives, aromatic aldehyde and ammonium acetate under ultrasound irradiation, prompted

***Table* 3.** Synthesis of 2,4,5-trisubstituted imidazoles using benzoin derivatives[a]

Entry	Benzoin	R_1, R_2	Aldehyde	R_3	Product	Time (min)	Yield[b] (%)
1	2a	H	3a	H	5a	90	88
2	2a	H	3b	*p*-OMe	5b	90	90
3	2a	H	3d	*m*-Br	5d	90	86
4	2a	H	3h	*m*-OH, *p*-OMe	5h	90	90
5	2a	H	3i	*m*-OMe, *m*-OMe	5i	90	87
6	2b	OMe	3a	H	5j	100	86
7	2b	OMe	3b	*p*-OMe	5k	100	87
8	2b	OMe	3d	*m*-Br	5m	100	85
9	2b	OMe	3h	*m*-OH, *p*-OMe	5q	100	88
10	2b	OMe	3i	*m*-OMe, *m*-OMe	5r	100	87

[a]Benzoin (1 mmol), Aldehyde (1 mmol), NH_4OAc (4 mmol), [ZnT(4-CH_3)PP] (3 μmol %), [b] Isolated yield based on aldehyde.

us to explore its catalytic activity in the condensation of benzoin or *p*-anisoin, aldehyde and ammonium acetate. The synthesis of different trisubstituted imidazoles with benzoin derivatives was carried out under the same reaction conditions which applied for benzil derivatives and the corresponding trisubstituted imidazoles were obtained in high yields (*Table* 3).

CONCLUSION

In summary an efficient and convenient procedure for the three-component one-step synthesis of 2,4,5-trisubstituted imidazoles has been developed by condensation reaction of 1,2-diketones or α-hydroxyketones, aromatic aldehydes, and ammonium acetate using [ZnT(4-CH_3)PP] as a novel and reusable catalyst under ultrasound irradiation at ambient temperature. Benzoin as well as benzil efficiently converted to their corresponding 2,4,5-trisubstituted imidazoles. The approach has several advantages, for example high yields, easy work up, reusability of catalyst, and applicability for both 1,2-diketones and α-hydroxyketones.

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