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*Research Article*

# $K_7Na_3P_2W_{18}Cu_4O_{68}$: A Mild, Efficient, and Reusable Catalyst for the One-Pot Synthesis of 1,2,4,5-Tetra Substituted Imidazoles

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An efficient method for the synthesis of 1,2,4,5-tetra substituted imidazoles using $K_7Na_3P_2W_{18}Cu_4O_{68}$ as catalyst is reported. This four-component condensation of benzil, aldehydes, amines, and ammonium acetate proceeds under solvent-free conditions. The catalyst is handling and recoverable.

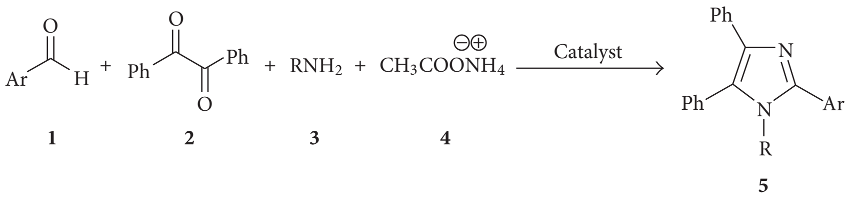
## 1. Introduction

Multicomponent reactions (MCRs) are interesting and important methods for the modern organic synthesis and medicinal chemistry because they are one-pot processes that three or more component are reacted to form one product with high atom economy and high selectivity; therefore, there are powerful tools for drug discovery [1, 2]. Compounds with the imidazole ring system have many pharmacological properties. Among them tetra substituted imidazoles are important fragments in many biological systems [3, 4]. There are many methods for the synthesis of highly substituted imidazoles synthesis [5–13]. Highly substituted imidazole rings generally cannot be synthesized under natural conditions [14, 15].

Recently, four-component condensation of benzil, benzaldehyde derivatives, primary amines, and ammonium acetate has been reported for this system using silicagel or zeolite HY [16], silicagel, $NaHSO_4$ [17], molecular iodine [18], $K_5CoW_{12}O_{40}$,$3H_2O$ [19], Keggin, Preyssler, and Wells-Dawson-type heteropolyacids [20–22], MCM-41 [23], $HClO_4$-$SiO_2$ [24], $BF_3$, $SiO_2$ [25], poly(AMPS-co-AA) [26], L-proline [27] high surface area [28], and $InCl_3$, $3H_2O$ [29], DABCO [30], $Al_2O_3$ [31], MPS [32], zeolite $Cu(NO_3)_2$ [33], SBPPSA [34], N-methyl-2-pyrrolidonium hydrogen sulfate [35], PEG-400 [36], $NaH_2PO_4$ [37], TFA [38], carbon based/solid acid [39], [Bmim]Br [40], $Fe^{+3}$-K10 [41], $CU(OAc)_2$ [42], CAN [43], $FeCl_3$ [44], $NiCl_2 \cdot 6H_2O$ [45], Bronsted acidic ionic liquids [46], $ZrCl_4$ [47], and solid acid zirconium nanocatalyst [48]. These methods have their own merits and drawbacks. Some of them are hazardous, toxic, and expensive reagents, giving side reactions, and have moderate yields. Therefore, introduction of new and environmentally benign methods for the synthesis of this heterocyclic system is in much demand. For these reasons, use of heteropolyacids (HPAs) as heterogenous or homogeneous acid and oxidation catalysts is worthwhile being investigated. Because of the advantages such as the easy workup, good recyclability and green processes, being nonvolatile, nontoxic, nonexplosive and stable towards humidity, thermally and air stable [49], they are good candidate for our purpose. Armed with these experiences [49] herein, we wish the four-component condensation of benzil, benzaldehyde derivatives, primary amines, and ammonium acetate to be catalyzed by $K_7Na_3P_2W_{18}Cu_4O_{68}$ in solvent-free conditions. The structure of catalyst has $[M_4(H_2O)_2(PW_9O_{34})_2]^{10-}$ anion, M4 cluster of four metal-oxygen octahedral (M = Cu + 2) is sandwiched by two trivacant lacunary Keggin units B-$\{PW_9O_{34}\}$ see Scheme 1 [50].

## 2. Experimental

All reported yields are isolated yields. All melting points were measured on an electrothermal melting point apparatus.



Scheme 1

Table 1: Solvent effect for the synthesis of **(5-a)**.

| Entry | Solvent | Time (min) | Temperature (°C) | Catalyst (mol %) | Yield (%) |
|---|---|---|---|---|---|
| 1 | $CH_3CN$ | 180 | Reflux | (0.2) | 30 |
| 2 | AcOH | 180 | Reflux | (0.2) | 30 |
| 3 | EtOH | 180 | Reflux | (0.2) | 40 |
| 4 | — | 90 | 140 | (0.2) | 92 |

Table 2: Effect of catalyst amount for the synthesis of **(5-a)**.

| Entry | Catalyst (mol%) | Time (min) | Yield (%) |
|---|---|---|---|
| 1 | 0.05 | 100 | 70 |
| 2 | 0.1 | 90 | 80 |
| 3 | 0.2 | 90 | 92 |
| 4 | 0.5 | 120 | 80 |
| 5 | — | 180 | — |

IR spectra were run on the furrier transform (FT) IR Bruker tensor 27 spectrometer and expressed in $cm^{-1}$ (KBr). The reaction was monitored by TLC. All reagents were purchased from Aldrich and Merck in high quality without purification. All products were known and characterized by comparison of their physical and spectral data with those already reported [18–26].

*2.1. Preparation of the Catalyst.* The catalyst synthesized according to the literature [51]. $CuCl_2$, $2H_2O$ (3.6 mmol, 0.62 g) dissolved in 12 mL water. $\Delta$-$Na_8HPW_9O_{34}$, $19H_2O$ ($\Delta$-$PW_9$) [52] (1.8 mmol, 5 g) added to this blue solution with stirring at room temperature until $\Delta$-$PW_9$ dissolved. KCl (8.8 mmol, 0.66 g) added to light green solution and a pale green solid precipitated. This mixture stirred for 10 min and warmed under hot (60°C) water or on a steam bath for periods (1–5 min) until the balk of precipitated solid had redissolved the slightly cloudy solution was centrifuged for 5 min to remove the fine blue suspension. The clear light green super-supernatant removed and crystallized at room temperature. Pale green crystalline cubes formed and continued to crystallized for 6–12 h. After that the product was filtered and dried.

*2.2. Synthesis of Tetrasubstituted Imidazoles: General Procedure.* Benzil (2 mmol), aldehyde (2 mmol), ammonium acetate, and (0.25 g) primary amine (2 mmol) were dissolved in 3 mL dichloromethane and were added to a mixture of ammonium acetate (0.25 g) and catalyst $K_7\,Na_3P_2W_{18}Cu_4O_{68}$ (0.2 mol%). The solvent was evaporated and the dry residue was heated on oil bath at 140°C for 90–120 min. The progress of reactions were monitored by TLC. The reaction mixture was cooled to room temperature and 30 mL of acetone was added. This mixture was filtered to separate the catalyst and the solvent was evaporated to dryness to afford the crude product which was purified by recrystallization from acetone-water (10 : 1 V/V).

*2.3. Selected Spectroscopic Data*

*1-Benzyl-2,4,5-triphenyl-1-H-imidazole* **(5-a)**. Mp 162–164°C; IR (KBr): 3055, 2925, 1600, 1497, 1444 $cm^{-1}$; $^1$HNMR ($CDCl_3$, 300 MHz): $\delta = 5.16$ (S, 2H, $CH_2$), 6.80–7.7 (m, 20H, Ph) ppm.

*2-(2-Hydroxyphenyl)-4,5-diphenyl-1-p-tolyl-1H-imidazole (5-j).* Mp 222–224°C; IR (KBr): 1601, 1535, 1479, 1401; $^1$HNMR ($CDCl_3$, 300 MHz): $\delta = 2.38$ (S, 3H, $CH_3$), 6.41–7.43 (m, 18H, ArH), 13.2 (S, 1H, OH) ppm.

*2-(2-Methoxyphenyl)-1,4,5-triphenyl-1-H-imidazole (5-k).* Mp 149–152°C; IR (KBr): 1600, 1580, 1467, 1493, 1397; $^1$HNMR ($CDCl_3$, 300 MHz): $\delta = 3.55$ (S, 3H, $CH_3$), 6.65–7.88 (m, 19H, ArH) ppm.

*1-Benzyl-2-(2-methoxyphenyl)-4,5-diphenyl-1-H-imidazole (5-l).* Mp 175–178°C; IR (KBr): 1602, 1579, 1525, 1496, 1396; $^1$HNMR ($CDCl_3$, 300 MHz): $\delta = 3.92$ (S, 3H, $CH_3$), 5.20 (S, 2H,$CH_2$), 6.65–7.65 (m, 19H, ArH) ppm.

## 3. Result and Discussion

To optimize the reaction conditions, synthesis of 1-(benzyl)-2,4,5-triphenyl-1-H-imidazole (**5-a**) was used as a model reaction. Condensation of benzil (2 mmol), benzylamine (2 mmol), benzaldehyde (2 mmol), ammonium acetate (o.25 g), and different amounts of catalyst $K_7Na_3P_2W_{18}Cu_4O_{68}$ occurred in various solvents but gave best result at 140°C in solvent-free conditions to give the desirable product (Tables 1 and 2). To establish the

Table 3: Synthesis of 1,2,4,5-tetra substituted imidazoles under different conditions.

| Product | Ar | R | Time (h) | Yield (%) | Mp (fou.) | Mp (rep.) |
|---|---|---|---|---|---|---|
| 5-a | Ph | $CH_2Ph$ | 1.5 | 92 | 162–164 | 163–165 [20] |
| 5-b | Ph | Ph | 1.5 | 87 | 212 | 216–218 [23] |
| 5-c | $3\text{-}NO_2\text{-}C_6H_4$ | Ph | 2 | 82 | 245–247 | 244–246 [18] |
| 5-d | $3\text{-}NO_2\text{-}C_6H_4$ | $4\text{-}CH_3\text{-}Ph$ | 2 | 80 | 148–150 | 149–151 [19] |
| 5-e | $4\text{-}NO_2\text{-}C_6H_4$ | Ph | 2 | 80 | 195–197 | 192–195 [21] |
| 5-f | $4\text{-}NO_2\text{-}C_6H_4$ | $4\text{-}CH_3\text{-}Ph$ | 2 | 80 | 220–222 | 218–220 [22] |
| 5-g | $4\text{-}OH\text{-}C_6H_4$ | Ph | 1.5 | 90 | 285-286 | 281–283 [26] |
| 5-h | $4\text{-}OH\text{-}C_6H_4$ | $CH_2Ph$ | 1.5 | 92 | 132–134 | 134-135 [24] |
| 5-i | $2\text{-}OH\text{-}C_6H_4$ | $CH_2Ph$ | 1.5 | 95 | 148-149 | 147-148 [25] |
| 5-j | $2\text{-}OH\text{-}C_6H_4$ | $4\text{-}CH_3\text{-}Ph$ | 1.5 | 90 | 222–224 | 226–228 [19] |
| 5-k | $2\text{-}OCH_3\text{-}C_6H_4$ | Ph | 1.5 | 90 | 149–152 | — |
| 5-l | $2\text{-}OCH_3\text{-}C_6H_4$ | $CH_2Ph$ | 1.5 | 92 | 175–178 | — |

Table 4: Comparison of different catalysts in the synthesis 1,2,4,5-tetra substituted imidazoles.

| No. | Catalyst | Conditions | Time (h) | Yield (%) [Ref.] |
|---|---|---|---|---|
| 1 | $K_5COW_{12}O_{40}$, $3H_2O$ | 140°C | 2–2.5 | 80–93 [19] |
| 2 | Silica gel/$NaHSO_4$ | 140°C | 2 | 85–92 [17] |
| 3 | High surface $SiO_2$ | $CH_2Cl_2$/sun light | 2–2.5 | 84–93 [28] |
| 4 | $InCl_3\text{-}3H_2O$ | MeOH/R.T | 8–9.5 | 69–82 [29] |
| 5 | L-Proline | Reflux in MeOH | 8.5–10 | 78–88 [27] |
| 6 | DABCO | t-ButOH/65°C | 12–15 | 70–82 [30] |
| 7 | HPA/silica | 140°C | 2 | 85 [22] |
| 8 | MPS | $H_2O$-MeOH/R.T | 4–6 | 84–90 [32] |
| 9 | PEG-400 | Reflux | 1.5–2 | 89–96 [36] |
| 10 | Carbon-based/solid acid | 130°C | 2 | 93 [39] |
| 11 | $K_7Na_3P_2W_{18}Cu_4O_{68}$ | 140°C | 1.5–2 | Present work |

Table 5: Effect of reused catalyst for the synthesis of (**5-a**).

| Run | Isolated yield (%) |
|---|---|
| 1 | 92 |
| 2 | 92 |
| 3 | 90 |
| 4 | 85 |
| 5 | 85 |

generality of the method, various aldehydes and amines were used in solvent-free conditions to obtain the corresponding tetra substituted imidazoles (Table 3).

Results with other catalysts and conditions for this one-pot four-component reaction for synthesis of 1,2,4,5-tetrasubstituted imidazoles pointed in (Table 4) for comparison. As it can be noticed, our catalyst and condition have advantages as far as times and yields of reactions are concerned. Reusability is a very important factor for heterogenous catalysts. After the first run of the reaction, the catalyst was separated by simple filtration and washed with $CH_2Cl_2$ and allowed to be dried at 100°C temperature for 1 h and used for similar reaction in 5 runs. The effect of reused catalyst on yields is shown in (Table 5).

## 4. Conclusions

We have established an efficient and facile one-pot synthesis of tetra substituted imidazole derivatives in solvent-free conditions. Catalyst handle and usable, high yields, easy workup, and purification of compound by nonchromatographic method (crystallization only) are other advantages of our work. This method is also environmentally friendly.

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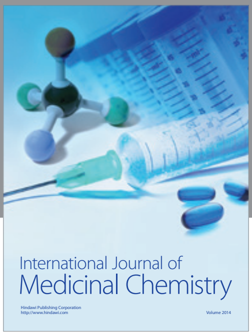
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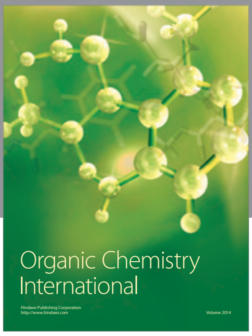
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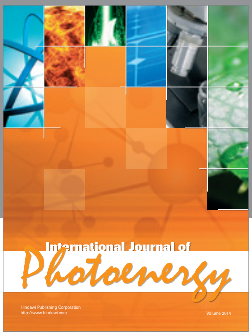
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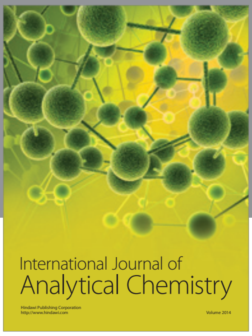
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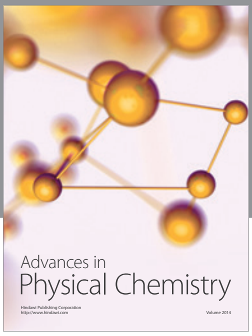
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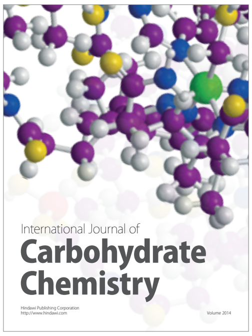
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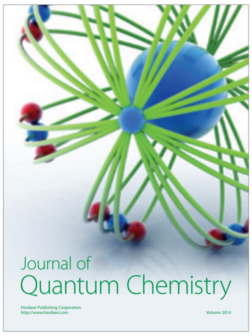
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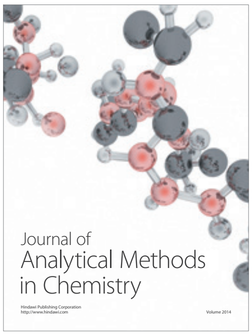
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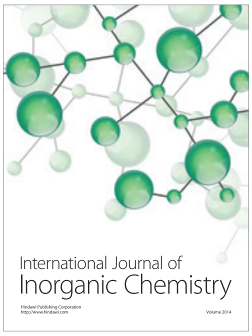
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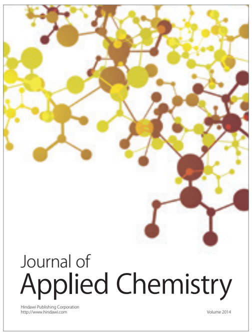
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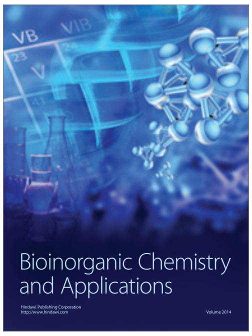
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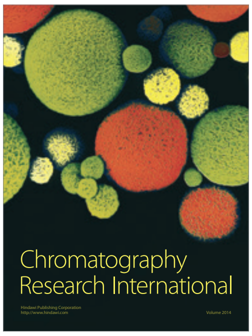
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