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SHORT COMMUNICATION

ONE-POT SYNTHESIS OF 2,4,5-TRISUBSTITUTED IMIDAZOLE DERIVATIVES CATALYZED BY BTPPC UNDER SOLVENT-FREE CONDITIONS

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ABSTRACT. A simple and efficient method for one-pot synthesis of lophine derivatives (2,4,5-trisubstituted imidazoles) by using the benzyltriphenylphosphonium chloride (BTPPC), as a catalyst, under solvent-free conditions is described. BTPPC is an available and inexpensive catalyst; also, it can be easily supplied. This procedure led to the corresponding 2,4,5-trisubstituted imidazoles products in high yields.

KEY WORDS: Lophine derivatives, BTPPC, Solvent-free, One-pot synthesis, Multi component synthesis

INTRODUCTION

Imidazole derivatives are an important class of heterocycles because of their applications in chemical processes and pharmaceuticals [1]. They have a wide range of biological activities and are well known analgesics, anti-inflammatory, antipar-asitic, anthelmintic, platelet aggregation inhibitors and antiepileptic agents [2]. Several methods such as the hetero-Cope rearrangement [3] and four-component condensation [4] for the synthesis of trisubstituted imidazoles are reported. In recent years, the synthesis of 2,4,5-trisubstituted imidazoles has been catalyzed by I_2 [5], $ZrCl_4$ [6], ionic liquid [7], L-proline [8], microwave irradiation [9], $Yb(OPf)_3$ [10], $InCl_3.3H_2O$ [11], $NiCl_2.6H_2O/Al_2O_3$ [12], DABCO [13], magnetic Fe_3O_4 nanoparticles [14], nano $MgAl_2O_4$ [15], ZrO_2-β-cyclodextrin [16], [EMIM]OAc [17], NaH_2PO_4 [18], *N*-methyl-2-pyrrolidone hydrogen sulfate [19], europium triflate [20], sulfated zirconia [21], *n*-Bu_4NBr [22], silica-supported Preyssler nanoparticles [23], $(NH_4)_6Mo_7O_{24}{\cdot}4H_2O$ [24], nano MgO [25], nano aluminium nitride [26], nano SiO_2-supported ferric hydrogen sulfate (FHS) [27] and KSF supported 10-molybdo-2-vanadophosphoric acid [28]. Although some of the methods are actually efficient from the synthetic chemist's points, many of the synthetic protocols for imidazoles reported above suffer from one or more disadvantages, such as harsh reaction conditions, poor yields, and prolonged reaction time, use of hazardous and often expensive acid catalysts. Therefore, the development of efficient, simple, environmentally friendly and high-yielding methods using new catalysts for the preparation of these compounds is still necessary.

BTPPC is a crystalline quaternary phosphonium salt which finds its application in Wittig reactions [29] and in phase transfer catalysis [30]. Very recently, we have reported the one-pot synthesis of dihydropyrimidinones/thiones under solvent-free conditions using BTPPC as a catalyst [31].

In this paper, we describe an efficient and practical route for the synthesis of lophine derivatives under solvent free conditions using BTPPC as catalyst (Scheme 1).

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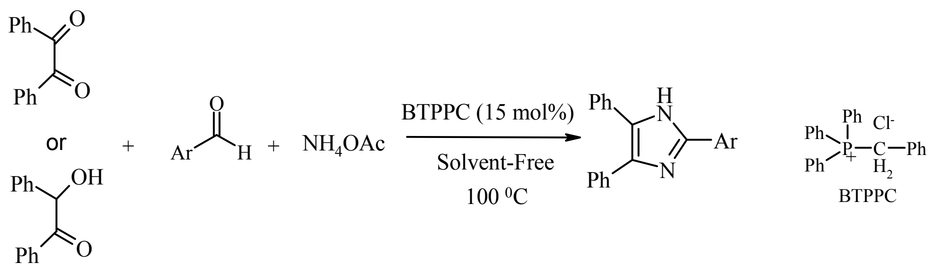
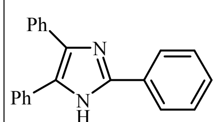
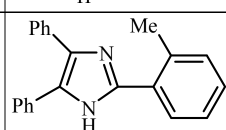
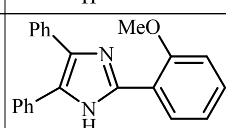
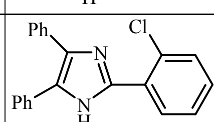
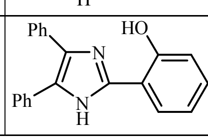


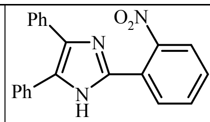
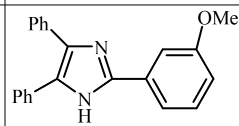
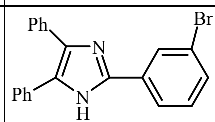
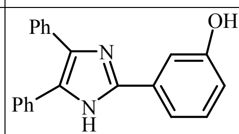
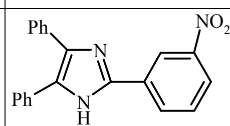
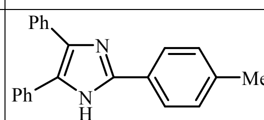
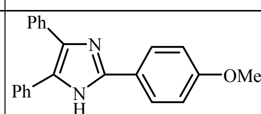
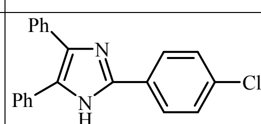
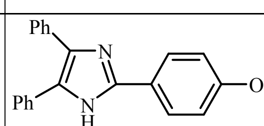
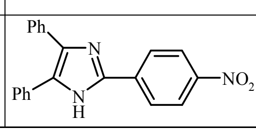
Scheme 1. Synthesis of lophine derivatives using BTPPC.

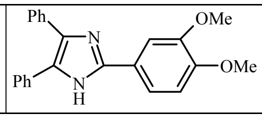
RESULTS AND DISCUSSION

In a model reaction, in the presence of the catalyst (15 mol%), the mixture of benzaldehyde (1 mmol), benzil (1 mmol) and NH_4OAc (2 mmol) as ammonia source, stirred at 100 °C under solvent free conditions. The 2,4,5-triphenyl imidazole are obtained in 92% yield. Various kinds of substituted benzaldehydes were also subjected in the presence of BTPPC at 100 °C under solvent free conditions (Table 1).

Table 1. BTPPC-catalyzed the synthesis of lophine derivatives[a].

Entry	Ar-CHO	Product	Time (min)		Yield (%)[b]		Mp/°C[c]		Ref.
			Benzyl	Benzoin	Benzyl	Benzoin	Found	Reported	
1	C_6H_5-		10	12	92	90	276-278	274-276	8a
2	2-MeC_6H_4-		10	13	89	86	208-210	207-208	23
3	2-$MeOC_6H_4$-		9	11	88	83	208-210	209-211	23
4	2-ClC_6H_4-		10	12	89	85	186	183-184	32
5	2-HOC_6H_4-		9	12	86	81	204-205	200-203	33

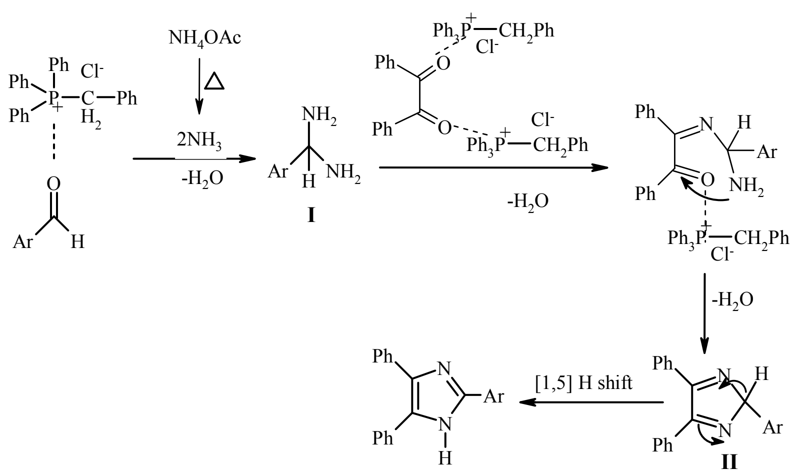
6	2-$NO_2C_6H_4$-		9	11	90	85	230-232	231-232	34
7	3-$MeOC_6H_4$-		12	14	86	82	260-262	266-268	35
8	3-BrC_6H_4-		10	12	89	85	198-200	199-200	36
9	3-HOC_6H_4-		10	13	86	81	257-258	258-260	8b
10	3-$NO_2C_6H_4$-		9	12	88	83	>300	313-315	12
11	4-MeC_6H_4-		10	12	92	89	231-232	234-236	23
12	4-$MeOC_6H_4$-		10	12	89	86	228-230	229-231	8a
13	4-ClC_6H_4-		9	13	90	86	264-266	262-264	8a
14	4-HOC_6H_4-		9	12	89	85	265-267	268-270	8a
15	4-$NO_2C_6H_4$-		9	12	90	86	232-233	235-238	12

16	3,4-MeOC$_6$H$_3$-		11	14	89	83	215-217	215	11

[a]Reaction conditions: benzyl/benzoin (1 mmol), aldehyde (1 mmol), NH_4OAc (2 mmol), BTPPC (15 mol%), 100 °C under solvent-free conditions. [b]Isolated yield. [c]Melting points are uncorrected.

We found that for aldehydes bearing either electron withdrawing or electron-releasing substituents in the *ortho*, *meta* or *para* positions; the reaction proceeded very efficiently in all cases. This procedure provides 2-aryl-4,5-diphenyl imidazoles directly, in relatively short reaction times and high yields. Furthermore, we used benzoin instead of benzyl and in this case corresponding products were achieved in good yields. In all cases, complete conversion was observed after appropriate time and the products were readily isolated in very high yields. A reasonable reaction mechanism for the BTPPC catalyzed is shown in Scheme 2.

BTPPC catalyst facilitates the formation of diamine intermediate [**I**] by increasing the electrophilicity of the carbonyl group of the aldehyde. Intermediate [**I**], in the presence of BTPPC, condenses with benzyl or benzoin to form intermediate [**II**], which in turn rearranges to the trisubstituted imidazole by a hydrogen shift [1, 5].



Scheme 2. A plausible mechanism for the formation of lophine derivatives.

In summary, a one-pot, multicomponent methodology has been developed for the synthesis of lophine derivatives catalyzed by BTPPC in high yields. Moreover, easy work-up, clean reaction profiles, low cost, availability, low toxicity, stable under normal temperatures and pressures of the catalyst, and short reaction time make this methodology a valid contribution to the existing processes in the field of 2-aryl-4,5-diphenyl imidazole derivatives synthesis.

EXPERIMENTAL

All the chemicals were obtained from Merck and Fluka Company. The melting points were obtained using an Electrothermal IA 9100 digital melting point apparatus. NMR spectra were recorded on a 400 MHz spectrometer using TMS as internal standard.

Preparation of BTPPC. To prepare BTPPC, 10 mmol (3.26 g) of PPh_3 and 10 mmol (1.26 g) of benzyl chloride were carefully dissolved in 15 mL of DMF, and the mixture was stirred at 80 ^oC. After 2 the resulting white precipitate was collected and washed with Et_2O (3×20 mL) and dried in a desiccator to afford a white solid. M.p. 335 ^oC [37].

General procedure for preparation of 2-aryl-4,5-diphenyl derivatives. A mixture of aldehyde (1 mmol), benzyl/benzoein (1 mmol) and NH_4OAc (2 mmol), as ammonia source, and BTPPC (15 mol %) stirred at 100 ^oC under solvent free conditions. The progress of the reaction was monitored by TLC. After completion of the reaction, the reaction mixture was dissolved in ethanol and poured into water. The resulting precipitate was filtered and purified by recrystallization from ethanol to afford the desired compound in pure form. All products were identified by comparison of their physical and spectroscopic data with those reported for authentic samples.

Triphenyl-1H-imidazole (Entry 1). Solid, m.p. = 276-278 ^oC. ^1H NMR (400 MHz, DMSO-d_6) δ_H (ppm): 7.56-7.22 (m, 15 arom. H), 12.69 (s, 1H, -NH).

2-(4-Methoxylphenyl)-4,5-diphenyl-1H-imidazole (Entry 12). Solid, m.p. = 228-230 ^oC. ^1H NMR (400 MHz, DMSO-d_6) δ_H (ppm): 3.81 (s, 3H, -OMe), 7.04 (d, J = 9.2 Hz, 2 arom. H), 7.55-7.28 (m, 10 arom. H), 8.02 (d, J = 9.2 Hz, 2 arom. H), 12.52 (s, 1H, -NH).

2-(3,4-Dimethoxylphenyl)-4,5-diphenyl-1H-imidazole (Entry 16). Solid, m.p. 215-217 ^oC. ^1H NMR (400 MHz, DMSO-d_6) δ_H (ppm): 3.81 (s, 3H, -OMe), 3.85 (s, 3H, -OMe), 7.06 (d, J = 8.0 Hz, 1 arom. H), 7.67-7.21 (m, 12 arom. H), 12.52 (s, 1H, -NH). ^{13}C NMR (100 MHz, DMSO-d_6) δ_H (ppm): 55.0, 55.0, 109.3, 112.3, 118.3, 123.6, 126.8, 127.5, 128.1, 128.6, 128.8, 129.1, 131.7, 135.7, 137.2, 146.1, 149.2, 149.5.

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