Efficient Multi-component Synthesis of Highly Substituted Imidazoles Utilizing P_2O_5/SiO_2 as a Reusable Catalyst

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Phosphorus pentoxide supported on silica gel (P_2O_5/SiO_2) has been used as an efficient and reusable catalyst for the one-pot pseudo four-component synthesis of 2,4,5-trisubstituted imidazoles from benzil or benzoin, aldehydes, and ammonium acetate. It was also used for four-component preparation of 1,2,4,5-tetrasubstituted imidazoles from benzil or benzoin, aldehydes, primary amine, and ammonium acetate under thermal solvent-free conditions. The remarkable features of this new procedure are high conversions, cleaner reaction, simple experimental and work-up procedures and also the catalyst can be easily separated from the reaction mixture and reused several times without any loss of its activity.

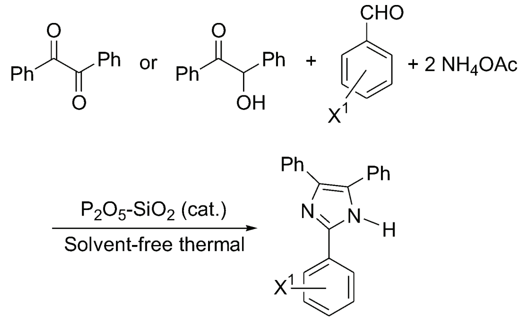
Keywords multicomponent reactions, domino reactions, phosphorus pentoxide supported on silica gel, 2,4,5-trisubstituted imidazoles, 1,2,4,5-tetrasubstituted imidazoles, heterogeneous catalysis

Introduction

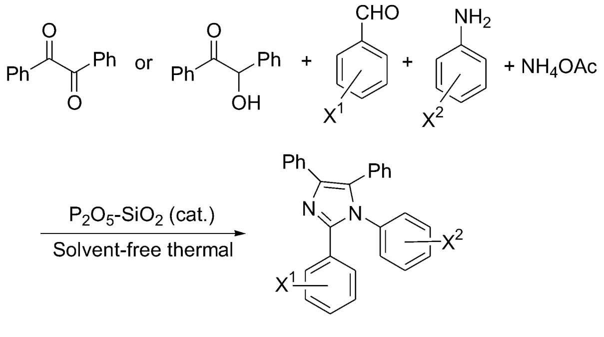
Multi-component reactions (MCRs) are powerful tools in generating products in organic and medicinal chemistry for their high degree of atom economy and application in the diversity-oriented convergent synthesis of complex organic molecules from simple and readily available substrates in a single synthetic operation.[1,2]

Imidazoles are an important class of heterocycles being the core fragment of different natural products and biological systems.[3,4] Substituted imidazoles used as light-sensitive materials in photography are known as inhibitors, fungicides and herbicides,[5] plant growth regulators and therapeutic agents.[6] Phosphorus pentoxide-methanesulfonic acid was used for the first time as a convenient alternative to polyphosphoric acid by Eaton *et al.* to escape the difficulties encountered with polyphosphoric acid (PPA).[7] In continuation of researcher works[8] on supported materials, P_2O_5/SiO_2 as an inexpensive, heterogeneous stable, free flowing, and white powder was prepared. P_2O_5/SiO_2 was used for several transformations.[9-15] We report herein, a simple pseudo four-component synthesis of 2,4,5-trisubstituted imidazoles from benzil or benzoin, aldehydes, and ammonium acetate (Scheme 1) and also four-component preparation of 1,2,4,5-tetrasubstituted imidazoles from benzil or benzoin, aldehydes, primary amine, and ammonium acetate (Scheme 2) in high yields using catalytic amount of P_2O_5/SiO_2 as a catalyst under solvent-free conditions for the first time.

Scheme 1



Scheme 2



Experimental

All reagents were purchased from Merck and Aldrich and used without further purification. P_2O_5/SiO_2 was prepared according to the reported procedure.[16] All yields refer to isolated products after purification.

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Products were characterized by comparison with authentic samples and by spectroscopy data (IR, ^{1}H NMR and ^{13}C NMR spectra). The NMR spectra were recorded on a Bruker Avance DPX 500 MHz instrument, and measured in DMSO-d_6 relative to TMS. FT-IR spectra were recorded on a JASCO FT-IR 460 plus spectrophotometer. Mass spectra were recorded on an Agilent technologies 5973 network mass selective detector (MSD) operating at an ionization potential of 70 eV. TLC was performed on a silica-gel Poly Gram SIL G/UV 254 plates.

Preparation of P_2O_5/SiO_2

A mixture of SiO_2 (2 g) and P_2O_5 (1 mmol, 0.142 g) was ground vigorously to give P_2O_5/SiO_2 catalytic system as a white powder (2.142 g).[16]

General procedure for preparation of 2,4,5-trisubstituted imidazoles

Benzil or benzoin (1 mmol), aldehyde (1 mmol) and ammonium acetate (4 mmol) were added to P_2O_5/SiO_2 (0.05 g, 7 wt%)[16] in an oil bath at room temperature. The resulting mixture was heated to 100 ℃ for the appropriate time reported in Table 1. Completion of the reaction was indicated by TLC. After completion of the reaction, the mixture was cooled to room temperature, and the crude solid product was dissolved in ethylacetate. The mixture was filtered for separation of the catalyst. The catalyst was washed twice with ethylacetate (5 mL×2), and then recovered catalyst was dried in oven at 100 ℃ for 3 h. The filtrate organic solution was concentrated. The solid product was purified by recrystallization procedure in ethanol. All of the desired product(s) were characterized by comparison of their physical data with those of known compounds. Some characterization data for selected known products are given below:

2-(4-Methoxyphenyl)-4,5-diphenylimidazole (Table 1, Entry 4): ^{1}H NMR (DMSO-d_6, 500 MHz) δ: 12.48 (s, 1H), 8.00 (dt, J=8.80, 2.0 Hz, 2H), 7.51 (d, J=7.2 Hz, 4H), 7.35 (t, J=7.2 Hz, 4H), 7.27 (t, J=7.2 Hz, 2H), 7.04 (dt, J=8.8, 2.0 Hz, 2H), 3.80 (s, 3H); ^{13}C NMR (DMSO-d_6, 125 MHz) δ: 55.2, 114.1, 122.9, 126.7, 127.0, 127.7, 128.4, 145.6, 159.4; IR (KBr) v: 3400, 3060, 1611, 1490, 1179, 1028, 830, 761 cm^{-1}.

2-(2,4-Dichlorophenyl)-4,5-diphenylimidazole (Table 1, Entry 18): ^{1}H NMR (DMSO-d_6, 500 MHz) δ: 12.8 (s, 1H), 7.80 (d, J=8.4 Hz, 2H), 7.81 (s, 1H), 7.61—7.22 (m, 12H); ^{13}C NMR (DMSO-d_6, 125 MHz) δ: 126.7, 127.2, 127.4, 127.9, 128.2, 128.3, 128.7, 128.8, 129.6, 130.6, 132.4, 132.6, 133.9, 134.8, 137.1, 142.4; IR (KBr) v: 3427, 3068, 1593, 824, 767 cm^{-1}.

General procedure for preparation of 1,2,4,5-tetrasubstituted imidazoles

Benzil or benzoin (1 mmol), aldehyde (1 mmol), amine (1 mmol), and ammonium acetate (1 mmol) were added to P_2O_5/SiO_2 (0.05 g, 7 wt%)[16] in an oil bath at room temperature. The resulting mixture was heated to 100 ℃ for the appropriate time reported in Table 2. Completion of the reaction was indicated by TLC. After completion of the reaction, the mixture was cooled to room temperature, and the crude solid product was dissolved in ethylacetate. The mixture was filtered for separation of the catalyst. The catalyst was washed twice with ethylacetate (5 mL×2), and then recovered catalyst was dried in an oven at 100 ℃ for 3 h. The filtrate organic solution was concentrated. The solid product was purified by recrystallization procedure in ethanol. All of the desired product(s) were characterized by comparison of their physical data with those of known compounds. Some characterization data for selected known products are given below.

1,4,5-Triphenyl-2-*p*-tolyl-1*H*-imidazole (Table 2, Entry 6): ^{1}H NMR (DMSO-d_6, 500 MHz) δ: 2.26 (s, 3H), 7.08 (d, J=8.0 Hz, 2H), 7.16—7.18 (m, 1H), 7.22—7.25 (m, 6H), 7.26—7.28 (m, 5H), 7.31—7.32 (m, 3H), 7.50 (d, J=8.0 Hz, 2H); ^{13}C NMR (DMSO-d_6, 125 MHz) δ: 21.5, 127.2, 128.4, 128.9, 129.0, 129.2, 129.50, 129.58, 129.6, 129.9, 131.3, 131.9, 132.0, 135.3, 137.60, 137,63, 138.6, 147.0; IR (KBr) v: 2935, 1590, 1577, 1492 cm^{-1}.

1,2-Bis(4-chlorophenyl)-4,5-diphenyl-1*H*-imidazole (Table 2, Entry 10): ^{1}H NMR (DMSO-d_6, 500 MHz): δ: 7.17—7.19 (m, 1H), 7.23—7.25 (m, 4H), 7.29—7.32 (m, 5H), 7.38—7.42 (m, 6H), 7.50 (d, J=7.0 Hz, 2H); ^{13}C NMR (DMSO-d_6, 125 MHz) δ: 127.2, 127.4, 129.44, 129.49, 129.9, 130.1, 130.8, 130.9, 131.3, 132.0, 132.3, 134.1, 134.3, 135.0, 136.2, 145.8; IR (KBr) v: 2987, 1596, 1499, 1411 cm^{-1}.

Results and discussion

Owing to the versatile biological activities of substituted imidazoles, numerous classical methods for the synthesis of these compounds have been reported.[3-6] In a typical procedure, benzil or benzoin, aldehydes, amines, and ammonium acetate are condensed in the presence of strong protic acid such as H_3PO_4,[17] H_2SO_4,[18] HOAc[19] as well as organo catalyst in HOAc[20] under reflux conditions and silica structure MCM-41 or *p*-toluenesulfonic acid (*p*-TsOH).[21] These catalysts present limitations due to the use of corrosive reagents and the necessity of neutralization of the strong acid media. In addition, the synthesis of these heterocycles in polar organic solvents such as ethanol, methanol, acetic acid, DMF and DMSO leads to complex isolation and recovery procedures.

In our research for selection of appropriate reaction conditions, we chose the reaction of benzil (1 mmol), benzaldehyde (1 mmol) and ammonium acetate (2 mmol) as a model with 0.05 g of P_2O_5/SiO_2 as catalyst at 100 ℃ under solvent-free conditions. We avoid using pure phosphorus pentoxide (P_2O_5) for the reaction, because it is a flammable, dangerous, corrosive to metal and extremely deliquescent compound. It reacts vigor-

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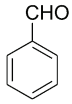
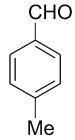
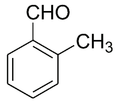
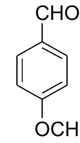
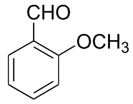
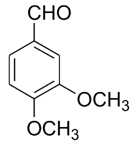
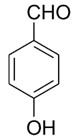
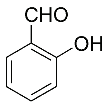
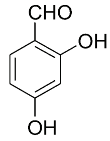
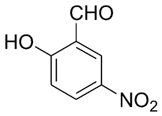
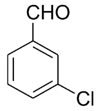
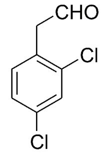
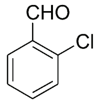
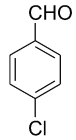
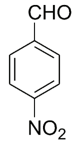
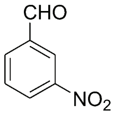
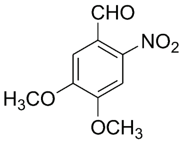
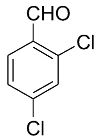
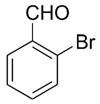
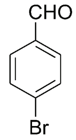
ously with water and water-containing substances, liberates much heat and may even cause fire. P_2O_5 is difficult to handle due to moisture sensitivity. However, the preparation of P_2O_5 on silica gel (P_2O_5/SiO_2) is straight forward to escape from disadvantages of P_2O_5. The stoichiometric amount of ammonium acetate in preparation of 2,4,5-trisubstituted imidazoles is 2. We can prepare 2,4,5-trisubstituted imidazoles using two equivalent of ammonium acetate, but we observed in our experiment and other published papers, if we use inexpensive and available ammonium acetate having more than two equivalent, the reaction will show better results. Thus, a slight excess of the ammonium acetate was found to be advantageous and hence the molar ratio of benzil or benzoin to ammonium acetate was kept at 1∶4. So, the condensation reaction of benzil or benzoin (1 mmol), aldehydes (1 mmol), and ammonium acetate (4 mmol) in the presence of P_2O_5/SiO_2 as the catalyst for synthesis of 2,4,5-trisubstituted imidazoles under solvent-free conditions at 100 ℃ was studied (Scheme 1, Table 1).

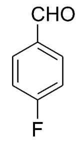
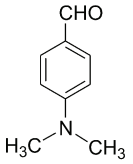
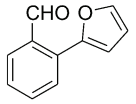
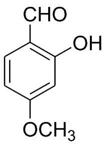
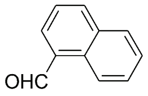
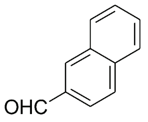
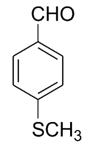
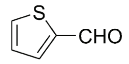
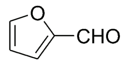
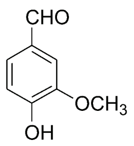
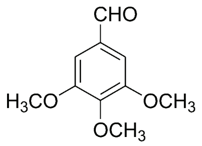
Table 1 Synthesis of 2,4,5-trisubstituted imidazoles via a one-pot pseudo four-component condensation reaction in the presence of P_2O_5/SiO_2 (0.05 g) as heterogeneous catalyst under thermal solvent-free condition at 100 ℃

Entry	Aldehyde	Reaction time/h		Yield^a/%		m.p./℃	
		Benzil	Benzoin	Benzil	Benzoin	Found	Reported [Ref.]
1		1	1.5	95	90	270	269 [24]
2		1.5	2	85	80	233—236	232—235 [25]
3		1.5	2.5	87	80	203—206	205—207 [26]
4		2	3	80	76	231—233	230—232 [26]
5		2	2.5	72	78	210	210—210.5 [26]
6		2	3.5	80	85	217—220	216—218 [27]
7		1	2.5	90	93	233	233 [24]
8		1	1.5	94	97	204—207	205 [28]
9		1	1.5	98	95	270—273	272 [22]

Continued

Entry	Aldehyde	Reaction time/h		Yield[a]/%		m.p./℃	
		Benzil	Benzoin	Benzil	Benzoin	Found	Reported [Ref.]
10		1	2	95	96	261—263	260.5—262 [29]
11		1	1.5	92	98	287—289	285—287 [23,30]
12		1	1.5	97	98	176—178	176.5—177 [31]
13		1	1.5	94	90	196—198	195—197 [32]
14		1	1.5	95	97	263	262—264 [26]
15		1	1.5	97	95	242—243	241—242 [23]
16		1	2	92	95	>265	>260 [23]
17		1.5	2.5	90	89	269—271	270 [22]
18		1	1.5	89	90	176—178	176.5—177 [31]
19		1.5	2	87	93	200—202	201—202 [33]
20		1	1.5	92	96	261—263	261.5—263.5 [26]

Continued

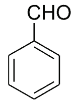
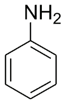
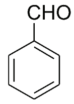
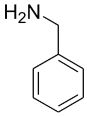
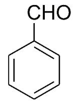
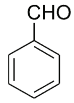
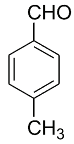
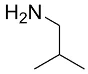
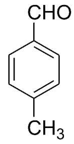
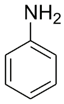
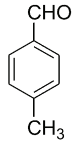
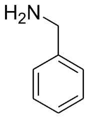
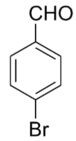
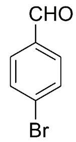
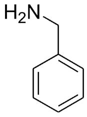
Entry	Aldehyde	Reaction time/h		Yield[a]/%		m.p./℃	
		Benzil	Benzoin	Benzil	Benzoin	Found	Reported [Ref.]
21		1.5	2	95	89	189—191	189—190 [32]
22		2	3	92	92	256—258	257—258 [32]
23		1	2	88	95	202—204	202—203 [33]
24		2	2.5	94	98	242—243	243 [22]
25		1.5	2	89	92	290—292	291.5—292 [26]
26		1	2	85	95	242—243	241—242 [34]
27		1.5	2	96	98	243—244	242—244 [35]
28		2	3	92	90	260—262	260—261 [32]
29		1.5	2	90	94	200—201	200—201 [32]
30		1.5	2	89	90	199	197 [36]
31		2	3	84	80	261—262	261 [22]

[a] Yields refer to isolated pure products. The known products were characterized by comparing their physical properties (m.p., ^{1}H NMR, ^{13}C NMR and FT-IR) with authentic samples.

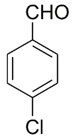
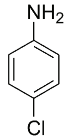
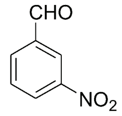
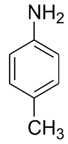
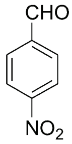
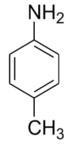
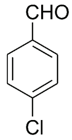
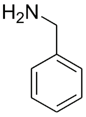
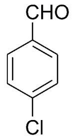
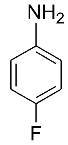
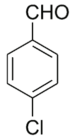
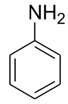
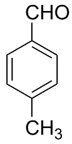
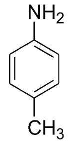
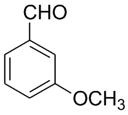
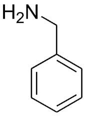
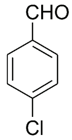
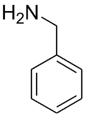
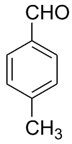
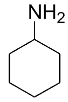
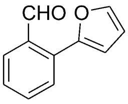
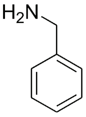
The efficiency and versatility of the P_2O_5/SiO_2 as catalyst for the preparation of 2,4,5-trisubstituted imidazoles were demonstrated by the wide rang of substituted and structurally divers aldehydes to synthesize the corresponding products in high to excellent yields. The presence of electron donating groups on the aromatic aldehyde resulted in the corresponding products in low yields and the reaction was sluggish, however the presence of electron-withdrawing groups afforded the corresponding 2,4,5-trisubstituted imidazoles in shorter reaction time with higher yields. Also, benzil in comparison with benzoin gives corresponding products in shorter reaction time but there was no effect in the yield of corresponding 2,4,5-trisubstituted imidazoles.

Next, as part of our program aimed at developing useful new synthesis methods based on the use of mentioned P_2O_5/SiO_2 as a catalyst, we have studied the four-component one-pot synthesis of 1,2,4,5-tetrasubstituted imidazoles by condensing benzil or benzoin (1 mmol), aldehyde (1 mmol), primary amines (1 mmol), and ammonium acetate (1 mmol) using a catalytic amount of P_2O_5/SiO_2 (0.05 g) under solvent-free conditions at 100 ℃ (Scheme 2, Table 2).

Table 2 Synthesis of 1,2,4,5-tetrasubstituted imidazoles via a one-pot four component condensation reaction in the presence of P_2O_5/SiO_2 as heterogeneous catalyst (0.05 g) under thermal solvent-free conditions at 100 ℃

Entry	Aldehyde	Amine	Reaction time/min		Yield[a]/%		m.p./℃	
			Benzil	Benzoin	Benzil	Benzoin	Found	Reported [Ref.]
1			35	40	89	87	215—217	218 [37]
2			30	45	88	83	161—163	158—160 [37]
3		CH_3NH_2	40	50	90	85	212—214	208—211 [38]
4		$C_2H_5NH_2$	45	55	80	89	121—123	124—126 [38]
5			25	60	85	80	156—157	151—153 [38]
6			20	55	78	85	192—193	189 [37]
7			15	20	96	98	164—165	165—166 [37]
8		CH_3NH_2	30	40	95	89	197—198	201—202 [37]
9			25	35	87	93	174—177	170—172 [39]

Continued

Entry	Aldehyde	Amine	Reaction time/min		Yield[a]/%		m.p./℃	
			Benzil	Benzoin	Benzil	Benzoin	Found	Reported [Ref.]
10			15	25	98	96	189—202	187—189 [38]
11			30	35	92	89	152—154	149—151 [38]
12			25	30	95	90	222—225	219—220 [38]
13			20	25	89	93	160—162	156—158 [38]
14			15	20	95	97	198—200	196—197 [38]
15			20	25	92	85	167—169	163—165 [39]
16			45	50	85	78	192—194	188—191 [38]
17			45	60	87	90	132—135	128—130 [39]
18			30	35	93	95	149—152	146—148 [39]
19			55	60	87	82	164—167	164 [39]
20			20	25	89	92	156—158	156—157 [39]

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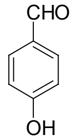
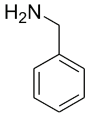
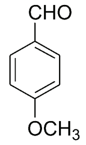
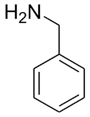
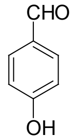
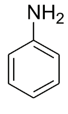
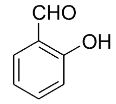
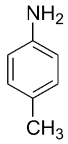
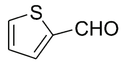
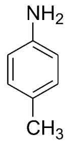
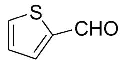
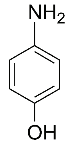
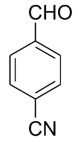
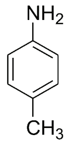
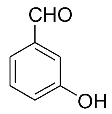
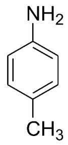
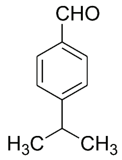
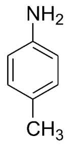
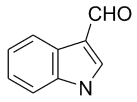
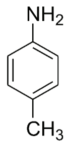
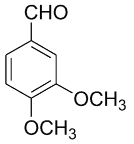
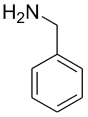
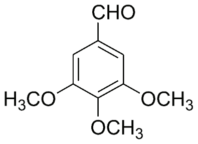
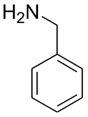
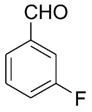
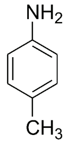
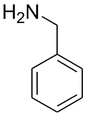
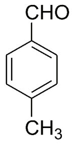
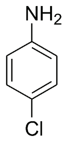
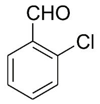
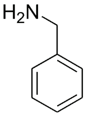
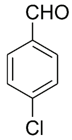
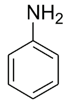
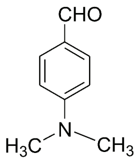
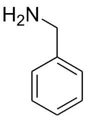
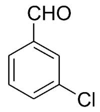
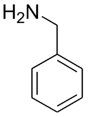
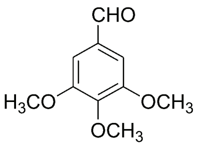
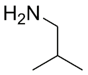
Continued

Entry	Aldehyde	Amine	Reaction time/min		Yield^a/%		m.p./℃	
			Benzil	Benzoin	Benzil	Benzoin	Found	Reported [Ref.]
21			20	30	92	97	135—137	134—135 [39]
22			40	45	80	85	155—158	157—160 [40]
23			20	25	92	96	283—286	280—281 [41]
24			30	40	95	92	227—230	226—228 [42]
25			30	40	90	95	203—205	199—202 [21]
26			30	35	89	87	294—297	289—290 [21]
27			30	45	90	92	205—207	198—201 [21]
28			45	50	95	92	227—229	230—232 [42]
29			45	60	89	87	214—216	215—217 [21]
30			35	45	82	89	219—221	218—220 [21]

Continued

Entry	Aldehyde	Amine	Reaction time/min		Yield[a]/%		m.p./℃	
			Benzil	Benzoin	Benzil	Benzoin	Found	Reported [Ref.]
31			45	60	75	78	188—190	185—187 [42]
32			50	60	72	76	188—190	184—186 [39]
33			30	40	85	89	250—253	248—251 [42]
34	CH_3CH_2CHO		50	60	80	87	100—104	104—105 [39]
35			25	30	92	92	162—165	165—167 [38]
36			25	35	90	85	145—147	140—142 [39]
37			20	30	87	85	154—155	150—152 [39]
38			25	40	85	86	180—182	183—185 [42]
39			25	30	92	92	140—141	144—146 [42]
40			50	60	89	83	117—119	112—115 [38]

[a] Yields refer to isolated pure products. The known products were characterized by comparing their physical properties (m.p., ^{1}H NMR, ^{13}C NMR and FT-IR) with authentic samples.

To show the merit of the present work in comparison with reported results in the literature, we compared results of P_2O_5/SiO_2 with reported catalysts in the synthesis of 2,4,5-trisubstituted imidazoles and 1,2,4,5-tetrasubstituted imidazoles. As shown in Tables 3 and 4, the P_2O_5/SiO_2 can act as a suitable catalyst with respect to reaction times and yields of the products.

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Table 3 Comparison of the efficiency of various catalysts with P_2O_5/SiO_2 in the synthesis of 2,4,5-trisubstituted imidazoles[a]

Entry	Catalyst	Condition	Time/min	Yield/%	Ref.
1	$InCl_3•3H_2O$	MeOH/r.t.	492	76	43
2	KH_2PO_4	EtOH/Reflux	60	89	44
3	$Yb(OPf)_3$	$C_{10}F_{18}$/80 ℃	360	80	45
4	$Zr(acac)_4$	EtOH/Reflux	120	95	46
5	*L*-Proline	Methanol/60 ℃	540	87	28
6	$[Hbim]BF_4$	100 ℃	60	94	24
8	$NiCl_2•6H_2O/Al_2O_3$	EtOH/Reflux	90	89	47
9	P_2O_5/SiO_2	100 ℃	60	98	Present work

[a] Based on the preparation of 2-(4-nitrophenyl)-4,5-diphenyl-*1H*-imidazole (Table 1, Entry 15).

Table 4 Comparison of the efficiency of various catalysts with P_2O_5/SiO_2 in the synthesis of 1,2,4,5-tetrasubstituted imidazoles[a]

Entry	Catalyst	Condition	Time/min	Yield/%	Ref.
1	BF_3/SiO_2	140 ℃	120	92	45
2	SiO_2	CH_2Cl_2/r.t.	120	90	48
3	$NaHSO_4/ SiO_2$	140 ℃	120	92	38
4	$InCl_3•3H_2O$	140 ℃	440	79	41
5	*L*-Proline	Methanol/r.t.	9h	88	28
6	$AlCl_3$	Methanol/ 60 ℃	120	53	45
7	$MgCl_2$	140 ℃	120	50	45
8	$SnCl_4$	140 ℃	120	60	37
9	$K_5CoW_{12}O_{40}•3H_2O$	140 ℃	120	95	42
10	P_2O_5/SiO_2	100 ℃	15	98	Present work

[a] Based on the preparation of 1-benzyl-4,5-diphenyl-2-*p*-tolyl-*1H*-imidazole (Table 2, Entry 7).

We also investigated the recycling of the catalyst under solvent-free conditions using a model reaction of 4-methylbenzaldehyde, benzylamine, benzil and ammonium acetate (Table 2, Entry 7). In this procedure, after completion of the reaction, the mixture was cooled to room temperature, and the crude solid product was dissolved in ethylacetate. The mixture was filtered for separation of the catalyst. The catalyst was washed twice with ethylacetate (5 mL×2), and then recovered catalyst was dried in an oven at 100 ℃ for 3 h. The recovered catalyst was used for the subsequent catalytic runs. The recovered catalyst could be recycled six times without significant loss of catalytic activity (Figure 1)

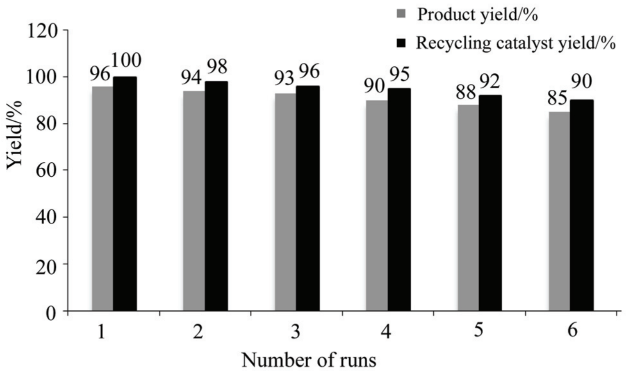

Figure 1 The study of the recovered catalyst, P_2O_5/SiO_2, activities

Conclusions

P_2O_5/SiO_2 efficiently catalyzes the condensation reaction of benzil or benzoin, aldehydes and ammonium acetate in a pseudo four-component reaction and affords the corresponding 2,4,5-trisubstituted imidazoles in high yields. Also this recyclable catalyst has been used for preparation of 1,2,4,5-tetrasubstituted imidazoles by one-pot condensation reaction of benzil or benzoin, aldehydes, primary amines and ammonium acetate in excellent yields under solvent-free and conventional heating conditions. The recovered catalyst was reused for six cycles without loss of its activities.

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