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Ultrasound based method for one-pot synthesis of substituted imidazoles using SiO_2-$OSbCl_2$ as highly effective and reusable catalyst

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ABSTRACT

A simple highly versatile and efficient sonochemical synthesis of 1,2,4,5-tetrasubstituted and 2,4,5-trisubstituted imidazoles is achieved by condensation of 1,2-dicarbonyl compound, aldehyde, ammonium acetate and primary amine using antimony trichloride immobilized on silica gel (SiO_2-$OSbCl_2$) as a catalyst in ethanol at moderate temperature. Operational simplicity, practicability and applicability to various substrates make this method an interesting alternative to previously applied procedures. From the environmental standpoint, this eco-friendly green catalyst is stable, highly active, easy to prepare and handle.

***Keywords**: SiO_2-$OSbCl_2$, Lewis acid, Heterogeneous catalyst, Imidazoles, Sonication.*

1. Introduction

Heterogeneous organic transformations have shown useful to chemists in the laboratories as well as in the modern industrials. These reactions are affected by the catalysts immobilized on the porous solid supports [1,2]. Lewis acids supported on "inert" carriers have received considerable importance in organic synthesis [3]. The individual features of supported Lewis acids such as their ease of handling, enhance reaction rates, greater selectivity, simple workup, and recoverability of catalysts make the use of supported Lewis acids as attractive alternatives to conventional individual reagents [4].

Antimony (III) trichloride is among the most widely used Lewis acids due to its accessibility as an inexpensive commercial reagent and easier handling as compared to other metal halides [5-7], Recently, Silica-bonded antimony (III) trichloride has been utilized as highly recoverable and reusable heterogeneous catalyst in various organic transformation including Knoevenagel condensation [8], bis(indolyl)methanes synthesis [9], Michael addition of indoles to α,β-unsaturated ketones [10], ring opening of epoxides [11], Biginelli reaction [12], and Paal-Knorr pyrrole synthesis [13].

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Imidazoles and their derivatives are important compounds that are found in nature and also in biological and chemical systems [14]. They play a major role in pharmacology, for instance histidine, histamine, and biotin [15]. These aromatic heterocyclic compounds operate as active parts in drugs like Losartan and eprosartan [16]. The substituted imidazoles perform various biological activities such as anti-inflammatory [17], anti-allergic [18] and analgesic activities [19].

Due to their wide range of biological, industrial and synthetic applications, recently, a number of methods have been developed for the synthesis of 2,4,5-trisubstituted and 1,2,4,5-tetrasubstituted imidazoles. Generally 2,4,5-trisubstituted imidazoles are synthesized by three-component cyclocondensation of 1,2-diketone, α-hydroxyketone or a α-ketomonoxime with an aldehyde and ammonium acetate, which comprise the use of ionic liquids [20], refluxing in acetic acid [21], silica immobilized sulfuric acid [22], $InCl_3.3H_2O$ [23], ceric ammonium nitrate (CAN) [24], $NiCl_2.6H_2O/Al_2O_3$ [25] and microwave irradiation[26]. On the other hand, the synthesis of 1,2,4,5-tetrasubstituted imidazoles have been carried out by four-component condensation of a 1,2-diketone, α-hydroxyketone or α-ketomonoxime with an aldehyde, primary amine and ammonium acetate using microwaves [27], heteropolyacid [28], $BF_3.SiO_2$ [29], silica gel/$NaHSO_4$ [30] or $HClO_4$-SiO_2 [31]. In

addition, they can also be accessed by the condensation of a 1,2-diketone with an aryl nitrile and primary amine under microwave irradiation [32], by hetero-Cope rearrangement [33], and by N-alkylation of trisubstituted imidazoles [34]. These methods are suitable for certain synthetic conditions, however, many of these procedures are associated with one or more disadvantages such as expensive reagents, longer reaction times, tedious work-up procedure, low selectivity, and large amounts of catalysts which would eventually result in the generation of large amounts of toxic waste.

In this context and in continuation of our studies on the applications of new catalysts in the development of synthetic methodologies [15,35,36], and for extension of cleaner processes, we wish to describe an efficient approach for the synthesis of polysubstituted imidazoles, using a catalytic amount of silica-bonded $SbCl_3$ (SiO_2-$OSbCl_2$) as a heterogeneous catalyst under ultrasound irradiation which, to the best of our knowledge, this has not been used earlier in this reaction (Scheme 1).

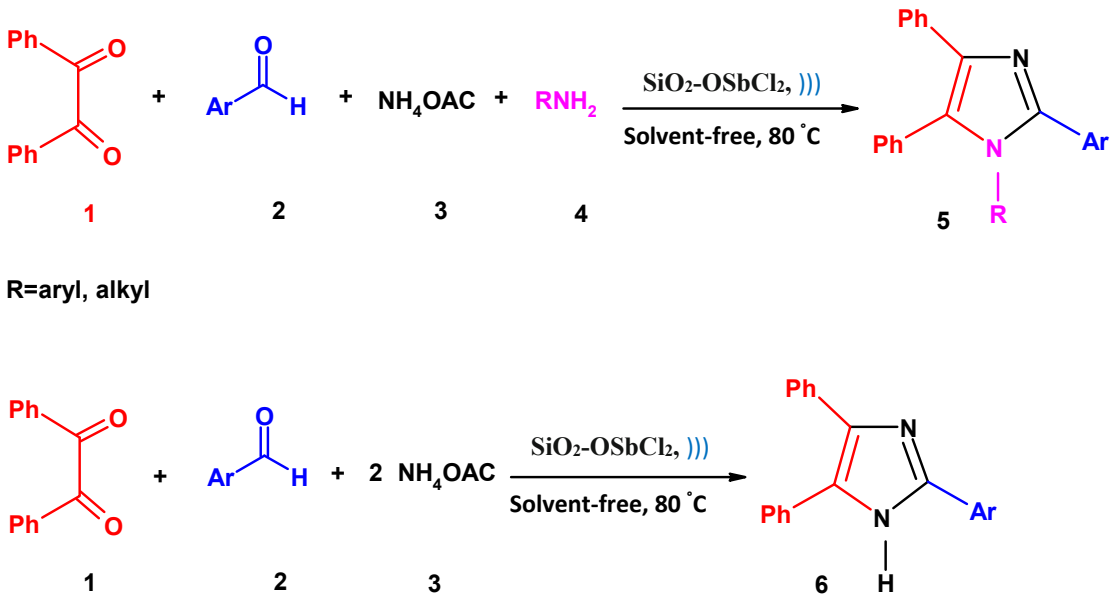
2. Experimental

2.1. Chemicals and apparatus

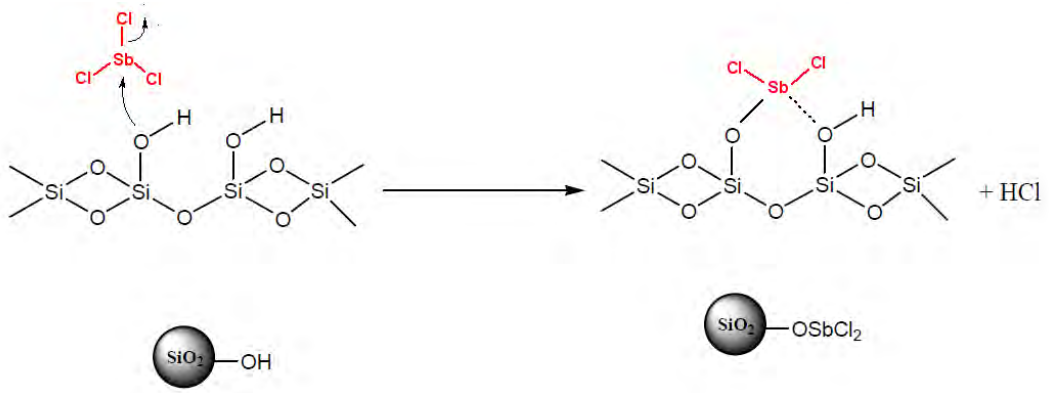
Chemical reagents in high purity were purchased from the Merck Chemical Company. All materials were of commercial reagent grade. Melting points were determined in open capillaries using an Electrothermal Mk3 apparatus and are uncorrected.^{1}H NMR and ^{13}C NMR spectra were recorded with a Bruker DRX-400 spectrometer at 400 and 100 MHz respectively and reported as parts per million (ppm) downfield from tetramethylsilane as internal standard. FT-IR spectra were obtained with potassium bromide pellets in the range 400-4000 cm^{-1} with a Perkin- Elmer 550 spectrometer. An ultrasound bath (water) dental scaler (with a frequency of 35 kHz and a nominal power 100 W) was used. A circulating water bath (DC2006, Shanghai Hengping Apparatus Factory) with an accuracy of 0.1 K was adopted to keep the reaction temperature at a constant.

2.2. Preparation of SiO_2-$OSbCl_2$ catalyst

Catalyst was prepared according to literature method [6]. Twenty grams of silica gel (80–200 mesh) was activated by refluxing with 150 mL of 6 mol/L hydrochloric acid under stirring for 24 h. The activated silica gel was filtered and washed with double distilled water to neutral and dried overnight at 70 °C in vacuum to give preconditioned silica gel. Antimony trichloride (1.99 g) was added to a suspension of activated silica gel (1.54 g) in EtOH (50.0 mL). The mixture was stirred at room temperature for 1 h. The solvent was removed, and the residue was washed three times with absolute ethanol and heated at 100°C under vacuum for 5 h to furnish silica-supported antimony (III) chloride (2.83 g) as a white free-flowing powder (Scheme 2).



Scheme 1. Synthesis of substituted imidazoles by SiO_2-$OSbCl_2$ catalyst under sonic conditions.



Scheme 2. Reaction of $SbCl_3$ with silica gel to form a highly effective catalyst.

2.3. Preparation of 1,2,4,5-tetrasubstituted imidazoles by use of SiO_2-$OSbCl_2$ catalyst

To a solution of aldehyde (1 mmol), benzil (1 mmol), ammonium acetate (0.4 g, 5 mmol) and primary aliphatic and aromatic amine (1 mmol) in 10 ml ethanol, SiO_2-$OSbCl_2$ (0.1 g) was added in a 25 mL Erlenmeyer flask and the reaction mixture was exposed to ultrasound irradiation at 80 °C for appropriated time. After the reaction was completed (monitored by TLC), the reaction mixture was dissolved in acetone and filtered. The solid product obtained was washed with water and recrystallized from acetone–water 9:1 (v/v) to offer the pure 1,2,4,5-tetrasubstituted imidazole derivatives as colorless crystals. Typical heating method was carried out under similar conditions.

2.4. Preparation of 2,4,5-trisubstituted imidazoles by use of SiO_2-$OSbCl_2$ catalyst

A 25 mL Erlenmeyer flask was charged with benzil (1 mmol), aldehyde (1 mmol), ammonium acetate (0.4 g, 5 mmol), and SiO_2-$OSbCl_2$ (0.1 g). The reaction flask was located in the ultrasonic bath, where the surface of reactants is slightly lower than the level of the water, the reaction mixture was exposed to ultrasound irradiation at 80 °C for appropriated time. The pure 2,4,5-trisubstituted imidazole derivatives were obtained by ultrasonic method. Typical heating method was carried out under similar conditions. The spectral data for some selected compounds were given below.

Selected spectral data

*2-(3,4-dimethoxyphenyl)-1,4,5-triphenyl-1H-imidazole (**$C_{29}H_{24}N_2O_2$, 5h**):*

m.p.= 178-180 °C; IR (KBr): $\bar{\nu}$ = 3045, 1617, 1578 cm^{-1}. ^{1}HNMR (400 MHz, DMSO-d_6): δ= 3.61 (s, *J* = 8.4 Hz, 6H), 6.85 (d, *J* = 8.8 Hz, 2H), 7.15-7.33 (m, 16H) ppm. ^{13}CNMR (100 MHz, DMSO-d_6): δ= 55.6, 55.6, 115.2, 124.5, 127.2, 128.5, 128.6, 129.1, 129.2, 129.2, 129.4, 130.1, 131.1, 132.30, 132.48, 136.50, 136.55, 136.61, 140.49, 145.29 ppm. UV-Vis (EtOH): λ_{max}= 311 nm.

*2,4,5-triphenyl-1-propyl-1H-imidazole (**$C_{24}H_{22}N_2$, 5k**)*:

m.p.= 87-89 °C; IR (KBr): $\bar{\nu}$ = 3025, 1597, 1479 cm^{-1}. ^{1}HNMR (400 MHz, DMSO-d_6): δ = 0.51 (t, *J* = 6.8 Hz, 3H), 1.32 (m, *J* = 6.8 Hz, 2H), 3.81 (t, *J* = 7.2 Hz, 2H), 7.10-7.55 (m, 13H), 7.7 (d, *J* = 6.8 Hz, 2H) ppm. ^{13}CNMR (100 MHz, DMSO-d_6): δ= 11.1, 23. 6, 46.3, 126.5, 128.5, 129.1, 129.2, 129. 3, 129.6, 130.3, 131.3, 131.6, 131.8, 135.2, 137.0, 147.2 ppm. UV-Vis (EtOH): $\lambda_{max=}$ 284 nm.

*2-(4-chlorophenyl)-4,5-diphenyl-1-propyl-1H-imidazole (**$C_{24}H_{21}N_2Cl$, 5l**)*:

m.p.= 85-87 °C; IR (KBr): $\bar{\nu}$ = 3025, 1645, 1489 cm^{-1}. ^{1}HNMR (400 MHz, DMSO-d_6): δ = 0.51 (t, *J* = 6.8 Hz, 3H), 1.34 (m, *J* = 6.8 Hz, 2H), 3.81 (t, *J* = 7.2 Hz, 2H), 7.15-7.30 (m, 12H), 7.34 (d, *J* = 7.2 Hz, 2H) ppm. ^{13}CNMR (100 MHz, DMSO-d_6):δ= 11.1, 23.5, 46.39, 126.5, 126.6, 128.5, 129.2, 129.4, 129.6, 130.52, 130.6, 130.9, 131.3, 131.3, 134, 137.1, 146 ppm.. UV-Vis (EtOH): λ_{max}= 294 nm.

*2-(4-methylphenyl)-4,5-diphenyl-1-propyl-1H-imidazole (**$C_{25}H_{24}N_2$, 5m**)*:

m.p.= 78-83 °C; IR (KBr): $\bar{\nu}$ =3028, 1620, 1497 cm^{-1}. ^{1}HNMR (400 MHz, DMSO-d_6): δ= 0.52 (t, *J* = 6.8 Hz, 3H), 1.30 (m, *J* = 6.8 Hz, 2H), 2.50 (s, 3H), 3.80 (t, *J* = 7.2 Hz, 2H), 7.12-7.35 (m, 12H), 7.50 (d, *J* = 8 Hz, 2H) ppm. ^{13}CNMR (100 MHz, DMSO-d_6): δ= 11.1, 21.4, 23. 6, 46.3, 125.6, 126.5, 126.5, 128.5, 129, 129.1, 129.3, 129.6, 129.7, 130.1, 131.6, 135.2, 136.9, 138.6, 147.3 ppm. UV-Vis (EtOH): λ_{max}= 294 nm.

*2-(4-methoxyphenyl)-4,5-diphenyl-1-propyl-1H-imidazole (**$C_{25}H_{24}N_2O$, 5n**)*:

m.p.= 76-80 °C; IR (KBr) $\bar{\nu}$ = 3016, 1628, 1510 cm^{-1}. ^{1}HNMR (400 MHz, DMSO-d_6): δ= 0.50 (t, *J* = 6.8 Hz, 3H), 1.33 (m, *J* = 6.8 Hz, 2H), 3.05 (s, 3H), 3.81 (t, *J* = 7.2 Hz, 2H), 7.10-7.30 (m, 12H), 7.46 (d, *J*=8 Hz, 2H) ppm. ^{13}CNMR (100 MHz, DMSO-d_6): δ= 21.2, 61.5, 127.1, 129.6, 133.6, 138, 158, 174.8 ppm. UV-Vis (EtOH): λ_{max}:= 305 nm.

3. Results and Discussion

Many recent papers describing the use of $SbCl_3/SiO_2$ in organic transformations pointed out that its use due to inexpensive, low toxicity, ease of handling and high catalytic activity is a potential green catalyst [8-13]. In continuation of our work to develop new and eco-friendly synthetic methodologies [37,38], herein we report a green, facile and efficient method for the synthesis of 2,4,5-trisubstituted and 1,2,4,5-tetrasubstituted imidazole derivatives catalyzed by SiO_2-$OSbCl_2$ under ultrasound irradiation at 80 °C ambient temperature (Scheme 1). To the best of our knowledge no report is available in the literature using ultrasonic-assisted for this transformation.

To achieve suitable conditions for the synthesis of polysubstituted imidazoles, various reaction conditions have been investigated in the reaction of benzaldehyde, aniline, benzil and ammonium acetate as a model reaction. In the initial experiments, we screened different common Lewis acids for their ability to catalyze the four-component cyclocondensation at 80 °C under ultrasound irradiation. As shown in Table 1, SiO_2-$OSbCl_2$ has a good efficiency compared to other Lewis acid catalysts such as $FeCl_3$, $AlCl_3$ and $ZnCl_2$. According to the obtained data, the use of 0.1 g of SiO_2-$OSbCl_2$ afforded 93 % yield (Table 1, entry 9) of the desired product. In contrast, the physical mixture of silica and $SbCl_3$ (Table 1, entry 13) exhibited an intermediate level of activity, which was lower than the silica supported antimony $SbCl_3$. This reveals that SiO_2-$OSbCl_2$ is presumably acting in a synergistic fashion to catalyze the reaction. Moreover, the experimental results show that the reaction times are shorter and the yields of the products are higher under sonication. As shown, in the absence of catalyst the yield of the product was found to be low (Table 1, entry 14), which indicates that the catalyst is obviously necessary for the reaction. We examined the effect of different solvents such as EtOH, MeOH, THF, DMF, CH_3CN, and DCM on the model reaction under ultrasound irradiation (35 kHz) at 80 °C. The results were summarized in Table 2. It was found that EtOH was the optimum solvent as well as solvent-free conditions. On the other hand, because of the toxicity of organic solvents, organic reactions under solvent free media have attracted the attention of researchers. So the reactions were conducted under solvent-free conditions, the expected products were obtained with excellent yields in short reaction times.

Table 1. Synthesis of 1,2,4,5-tetrasubstitutedimidazole catalyzed by different catalysts under sonication at 80 °C.

Entry	Catalyst (g)	Time (min)	Yield (%)[a]
1	0.02 (SiO_2-$FeCl_3$)	25	20
2	0.02 ($SiO2$-$AlCl_3$)	25	10
3	0.02 (SiO_2-$ZnCl_2$)	25	20
4	0.02 (SiO_2-$OSbCl_2$)	25	28
5	0.04 (SiO_2-$OSbCl_2$)	25	43
6	0.06 (SiO_2-$OSbCl_2$)	25	66
7	0.08 (SiO_2-$OSbCl_2$)	25	79
8	0.09 (SiO_2-$OSbCl_2$)	25	84
9	0.1 (SiO_2-$OSbCl_2$)	25	93
10	0.1 (SiO_2-$OSbCl_2$)[b]	45	87
11	0.11 (SiO_2-$OSbCl_2$)	25	93
12	0.1 (SiO_2)	25	30
13	0.1 ($SbCl_3/SiO_2$)	25	80
14	-	40	10

[a]Isolated yield.
[b]Without sonication.

Table 2. Screening of solvent effect on model reaction.

Entry	Solvent	Time (min)	Yield (%)[a]
1	Ethanol	25	93
2	Methanol	25	70
3	DCM	25	20
4	DMF	25	45
5	THF	25	50
6	Acetonitrile	25	60
7	Ethyl acetate	25	70
8	Chloroform	25	43
9	H_2O	25	20
10	Solvent-free	25	93

[a]Isolated yield.

To synthesize 1,2,4,5-tetrasubstitutedimidazole, we carried out the reactions of benzil, aliphatic and aromatic amines and ammonium acetate with various aromatic aldehydes carrying either electron-releasing or electron-withdrawing substituent under sonic conditions.

As shown in Tables 3, the nature of the functional group on the aromatic ring of the aldehyde exerted a strong influence on the time and the reaction yield. Aldehydes with electron-withdrawing groups afford more pure products compared with electron-donor groups in higher yields. Moreover, the results showed that the reaction worked well in the presence of aliphatic amine and the yield of product was higher than aromatic ones maybe because of the higher nucleophilicity of aliphatic amines.

Using the optimized reaction conditions, this process was demonstrated by the wide range of substituted and structurally divers aldehydes to synthesize 2,4,5-trisubstituted imidazole derivatives in high to excellent yields (Table 4). Aldehydes bearing either electron-withdrawing or electron-donating groups perform equally well in the reaction and all imidazoles were obtained in high yields.

Table 3. Synthesis of 1,2,4,5-tetrasubstituted imidazole derivatives catalyzed by SiO_2-$OSbCl_2$.

Entry	R	Amine	Product	Time (min)/Yield (%)[a]	Time (min)/Yield (%)[b,c]	m.p. (°C)	Ref.
1	H	Ph-NH_2	5**a**	45/87	25/93	216-218	[39]
2	4-Me	Ph-NH_2	5**c**	60/80	28/90	186-188	[40]
3	4-Cl	Ph-NH_2	5**d**	35/92	15/95	160-163	[41]
4	3-NO_2	Ph-NH_2	5**e**	30/93	30/80	243-246	[42]
5	2-OH	Ph-NH_2	5**f**	70/73	30/75	252-254	[41]
6	3,4-$(OMe)_2$	Ph-NH_2	5**h**	75/75	33/80	178-180	[43]
7	2-OMe	Ph-NH_2	5**i**	20/85	20/84	207-211	[41]
8	4-OMe	Ph-NH_2	5**j**	65/87	28/87	177-180	[39]
9	H	n-Pr-NH_2	5**k**	40/90	20/94	88-90	[43]
10	4-Cl	n-Pr-NH_2	5**l**	35/93	13/95	86-87	[43]
11	4-Me	n-Pr-NH_2	5**m**	50/82	25/91	81-83	[43]
12	4-OMe	n-Pr-NH_2	5**n**	55/80	25/89	76-78	[43]

[a]Without sonication.

[b]Under sonication.

[c]Isolated yield.

Table 4. Synthesis of 2,4,5-trisubstituted imidazole derivatives catalyzed by SiO_2-$OSbCl_2$.

Entry	R	Product	Time (min)/Yield (%)[a,c]	Time (min)/Yield (%)[b,c]	m.p. (˚C)	Ref.
1	H	6a	35/87	15/97	272–273	[40]
2	4-Me	6b	40/80	15/98	226-227	[40]
3	4-OMe	6c	34/78	15/97	227-228	[40]
4	4-Cl	6d	25/90	10/90	272–273	[40]
5	3-Cl	6e	20/93	15/92	226-227	[40]
6	2-OMe	6f	45/73	15/95	227-228	[40]
7	3-NO_2	6g	20/95	15/89	259-261	[40]
8	2-OH	6h	45/70	20/97	282-284	[40]
9	3-OH	6i	35/85	10/95	260-261	[40]
10	3,5-$(OMe)_2$	6j	45/80	20/99	269-271	[40]
11	4-Br	6k	25/92	15/95	203-205	[40]
12	4-$N(Me)_2$	6l	40/77	10/97	260-261	[40]

[a]Without sonication

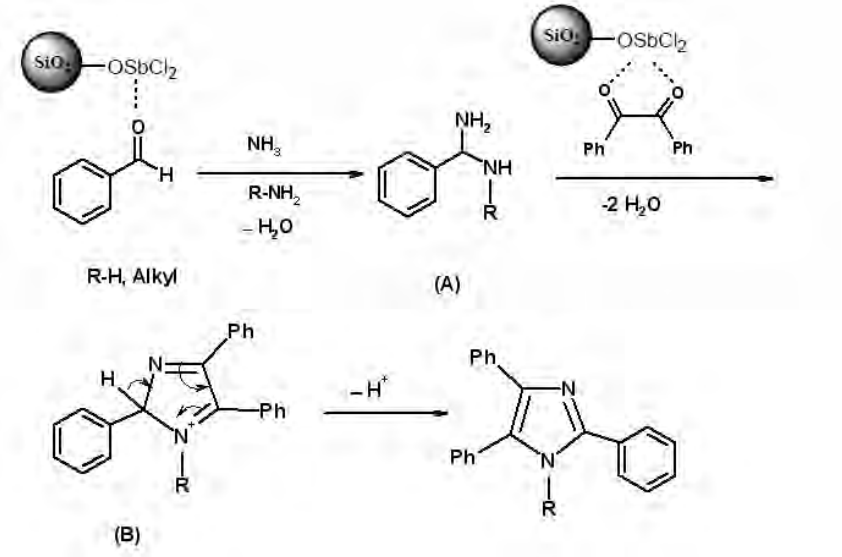
[b]Under sonication.

[c]Isolated yield.

Also, as shown in Tables 3 and 4, when the reaction was carried out without sonic conditions, it gave comparatively low yields of products and took longer reaction times, therefor, this protocol is more environmentally friendly, particularly when considering the basic green chemistry concepts.

The plausible mechanism for the synthesis of highly substituted imidazoles in the presence of SiO_2-$OSbCl_2$ is outlined in Scheme 3. The reaction proceeds via the diamine intermediate [A], which is formed by the activation of aldehyde carbonyl group by the Lewis acids on the surface of SiO_2.

Condensation of diamine with 1,2-diketone followed by dehydration, and then rearrangement through the imino intermediate [B] yielded the desired product. Lewis acid site on silica-supported antimony(III) chloride acts as a highly efficient catalyst, even for low activity substrates, in the preparation of imidazoles derivatives. Moreover, the higher catalytic activity of SiO_2-$OSbCl_2$ is due to a good dispersion of $SbCl_3$ over high surface area silica support. The results indicated that dispersed $SbCl_3$ coordinates with surface hydroxyl groups leading to formation-O–Sb–Cl as stable Lewis acid sites under the reaction condition [6].



Scheme 3. Proposed mechanism for the formation of substituted imidazoles in the presence of SiO_2-$OSbCl_2$.

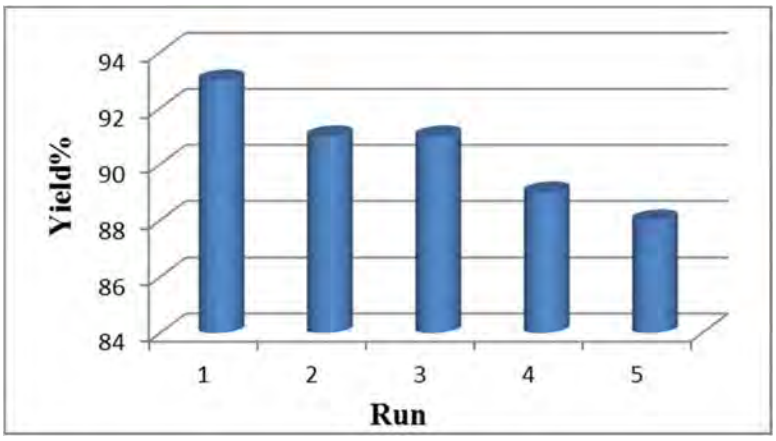


Fig. 1. Recyclability of SiO_2-$OSbCl_2$ in the model reaction by ultrasonic technique.

We also examined the possibility for recycling the catalyst in the model reaction. When the reaction was complete, ethyl acetate was added to the reaction mixture, the mixture was stirred, and the $SbCl_3/SiO_2$ was filtered off, washed with ethyl acetate, and activated by heating for 2 h at 110 °C. The recovered catalyst was used in another reaction with the same substrates without significant loss of activity after five cycles (Fig. 1).

4. Conclusions

In conclusion, we have perceived that SiO_2-$OSbCl_2$ can catalyzed efficiently the one-pot synthesis of a large number of tri- and tetrasubstituted imidazoles at moderate temperature under sonic conditions. Present methodology offers very attractive features such as reduced reaction times, higher yields, and economic viability of the catalyst, when compared with conventional methods as well as with other catalysts, which will have wide scope in organic synthesis. We expect this method will find extensive applications in the field of combinatorial chemistry, diversity-oriented synthesis, heterogeneous catalytic systems, and drug discovery.

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