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Simple and accurate method for synthesis of 2,4,5-triary-1H-imidazole derivatives using SiO_2-$NaHSO_4$ under solvent-free condition

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Abstract
This paper presents a mixture of benzyl, aromatic aldehyde and ammonium acetate in presence of SiO_2-$NaHSO_4$ under solvent-free condition were converted to 2,4,5-Triaryl-1H-Imidazoles. The short reaction time, cleaner reaction, and easy workup make this protocol practical and economically attractive.

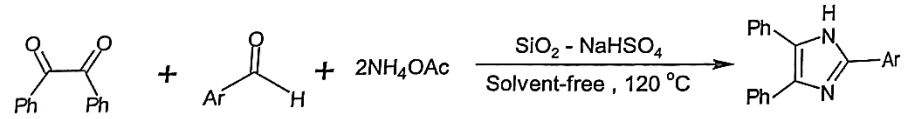
Keywords: SiO_2-$NaHSO_4$, aromatic aldehyde, heterocyclic compounds

Introduction
Since the multi-component reactions for the synthesis of organic compounds and these compounds can be used as a drug and precursor multicomponent reactions, so to investigate them out is important [1-4].
Imidazoles are important heterocycle compounds in medicinal chemistry. Imidazoles widely have been used of biological activity which has made them privileged structures in combinational drug discovery libraries. They have the biological roles and also found application as a chromophore with high extinction coefficient, readily tunable absorption wavelength, and fluorophoric properties and was desirable as a large planer synthetic building block in supramolecular chemistry. Recently, several improved methodologies have been developed that use HY/silica gel, acidic Al_2O_3, AcOH, ionic liquid, NH_4OAc, sodium bisulfate among others [5-8]. Previously, we have synthesized a number of heterocyclic compounds [9-17].
In this study, we have used of SiO_2-$NaHSO_4$ as a catalysts to develop a new and easy methodology for the synthesis of 2,4,5-triaryl imidazole derivatives, The experiments were started with the study of one-pot, a simple, mild and efficient method for the preparation of the 2,4,5-triarylimidazoles by using Sio_2-$NaHSO_4$ as a catalyst (Scheme1).

Experimental
All chemicals were obtained from Merck of Fluka without further purification. Silica gel SILG/UV 254 plates were used for TLC. IR spectra were measured on a Shimadzu IR-470 Spectrophotometer. ^{1}H NMR spectra were determined on Bruker 500 DRX AVANCE instrument at 500 MHz, respectively'.

General procedure for preparation of 2a-I
A mixture of aldehyde (1 mmol), benzyl (1 mmol), ammonium acetate (3 mmol) and SiO_2-$NaHSO_4$ (10 mol %) as a catalyst was stirred at 120 ºC for 30 min. The progress of reaction was monitored by TLC. After finishing, recrystallized from ethanol 95% to give pure products (A1-A4).

Spectral data 2,4,5-Triphenyl-1H-imidazole(A1)
Pale yellow crystals, Tield: 0.26 δ(89%) mp: 274-276 ºC. IR (λ_{max}∫cm^{-1})(KBr): 3400(NH Str.); ^{1}H NMR(400.13 MHZ $CDCI_3$)δ(ppm): 7.39-7.85(10H, m, CH_{arom}); 7.98-8.0(2H, d, j=8HZ, 2CH); 8, 10(1H, s,CH).



Results and Discussion

We have been able to introduce an efficient and environmentally friendly for the synthesis of imidazole derivatives via condensation of benzyl with various aromatic aldehydes and ammonium acetate. 3000(CHarom Str.); 1600(C=C Str.); 1470(C=NStr.). ^{1}H NMR(400. 13MHz $CDCI_3$)δ (ppm): 7.22-7.49(14H, m, CH_{arom}), 7.94(2H, d, J=8HZ,2CH), 9.40(H,s, NH).

2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazole(A2)

Bright crystals, Yield: 0.28 g(85%, mp: 260-262 °C. IR(λ_{max}∫cm^{-1}) (KBr): 3400(NH Str.); 3000(CHarom Str.); 1600(C=C Str.); 1500(C=N Str.). ^{1}H NMR (400.13 MHZ $CDCI_3$) δ(ppm): 7.09-7.84 (11H,m,CH_{arom}), 7.82-7.84 (2H,d,j=8Hz 2Ch), 9.53(H,S, NH).

2-(4-Methoxyphenyl)-4,5-diphenyl-1H-imidazole(A3)

Pala yellow crystals, Yield: 0.28 g(86%) mp: 227-230 °C. IR(λ_{max}∫cm^{-1})(Kbr): 3442(NH Str.); 3070(CHarom Str.); 1599(C=C Str.); 1450(C=N Str.). ^{1}H NMR(400.13 MHZ $CDCI_3$) δ(ppm): 3.84(3H,s,CH_3);6.93(2H,d,j=8Hz, 2CH); 7.27-7.53 (10H,m, 10CH), 7.82(2H,d,^{3}J=8Hz, 2CH).

2-(3-Nitrophenyl)-4,5-diphenyl-1H-imidazole(A4)

Pale yellow crystals, Yield: 0.3 g(88%) mp>300 °C. IR(λ_{max}∫cm^{-1})(KBr): 3400(NH); 3055 (CHarom); 1600(C=C); 1475(C=N); 1420,1375 (NO_2).

Therefore, reported SiO_2-$NaHSO_4$ as catalyst which could provide an efficient, cheap, easy separation high yield and simple route under solvent-free condition for the synthesis of imidazoles.

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