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Transition Metal Oxide (TiO_2) Catalyst Promoted by the Synthesis of Imidazoles

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Abstract: ***A one pot, three-component condensation of benzil, substituted aromatic aldehydes, and ammonium acetate using as a mild oxidant is achieved to form trisubstituted imidazole compounds. A series of 2, 4, 5-triarylimidazole derivatives are obtained from the simple and highly efficient three components such as benzil, aromatic aldehydes and ammonium acetate promoted by TiO_2 in presence ethanol solvent. The study explores the scope and limitation of TiO_2 as an oxidant and suggests advantages, viz., simplicity of operation, reduction in time, and an increase in product yields. These are used in the catalysis. This reaction produces the 2, 4, 5-triarylimidazole derivatives with good yield and purity. The newly synthesized derivatives determined by advanced spectroscopic data viz; IR, ^{1}HNMR, ^{13}CNMR and LCMS and also determined by elemental analysis.***

Keywords: Multicomponent reactions (MCRs), Benzil, benzaldehyde, NH_4OAc, 2, 3, 4 -trisubstituted Imidazole, TiO_2

1. Introduction

One-pot processes involving multiple components are interesting because they produce a single, high-yield output. [1-5]. These reactions are useful and effective methods for creating a small number of chemical molecules with biological and pharmacological activity. It was discovered that numerous physiologically significant natural compounds had substituted imidazole structures. The primary structural component of several commonly used medications, including ketoconazole, metronidazole, clotrimazole, omeprazole, metidine, and a possible inhibitor of P38 kinase, as well as therapeutic medicines, HIV-I protease, and three proton pump inhibitors, are tri aryl imidazole.1.Now days, imidazole plays an important key role in the field of organic synthesis and medicinal chemistry because of an extensive range of biological activities including antidepressant activity (6), Antitubercular Agents (7), Mycobacterium Tuberculosis (8), anti-inflammatory (9), Antifungal Activity (10), Antileishmanial activity (11), antioxidant (12), Antibacterial Agents (13), antimicrobial activity (14).These catalysts and their methods were all plagued by drawbacks, such as the requirement for specialized equipment, time-consuming procedures for recovering and reusing the catalysts, expensive, poisonous, and moisture-sensitive catalysts, as well as volatile organic solvents and labor-intensive setup.

Despite the great potential of these techniques, the reactions suffer from low yields, prolonged reaction durations, costly reagent usage, mixed product associations, and limited generalizability. As a result, they weren't suitable for the synthesis of imidazoles with different structures. There is still a great need and desire for the development of clean, highly productive, and environmentally friendly methods.

The effectiveness of TiO_2 catalysts for the synthesis of trisubstituted imidazoles has not been previously studied, according to our ongoing investigation into the catalytic activity of magnetically separable substituted TiO_2 for various organic transformations. Therefore, using the cyclocondensation of benzil, benzaldehyde, and ammonium acetate, an effort has been made to recognize the effects of TiO_2 catalyst promoted the one-pot multicomponent synthesis of 2, 4, 5, -trisubstituted imidazoles.

The present research work, our aimed to main focus on thedevelopment of straightforward and an efficient process for the synthesis of different substituted titled derivatives and it would to report on a simple, economical, and efficient one-pot method for the synthesis of 2, 4, 5-trisubstituted imidazoles from benzil, ammonium acetate, aromatic aldehydes, and TiO_2 catalysts (scheme-1).

2. Methods and Materials

Experimental:
All air reactions were carried out in oven dried (100^0C) or flame dried glassware and commercially substituted aryl benzaldehyde, benzil, NH4OAc and IPA were bought from Merck and used as such without further purification. All melting points the newly analogous were measured in open capillaries and are uncorrected. ^{1}H and ^{13}C NMR spectra were recorded on a BRUKER 400 and 100MHz spectrometer in $CDCl_3$and using TMS an internal standard. Elemental analyses were carried out in Perkin Elmer 240 CHN elemental analyzer. Analytical thin layer chromatography was performed with Merck silica gel plates with PF254 indicator. Compounds were visualized under UV lamp. Column chromatography was carried out using 100-200 mesh silica gel and technical grade solvents.

The methods of procedure for the synthesis of 2, 4, 5-trisubstituted imidazoles:
The dry and clean 50mL RBF and charged themixture of starting material benzili (1.12 mmol) and substituted aromatic aldehydes (1.25mmol)was dissolved in ethanol (25mL) followed by the ammonium acetate (4.50 mmol), and TiO_2 (2.12 mmol) was added to the reaction mixture and continuously stirred the reaction mixture at 70ºC for 5 hours.The completion of the reaction was identified by TLC and the reaction mixture was poured out on ice cooled saturated $NaHCO_3$ solution (25 mL) and the aqueous phase was extracted with ethyl acetate ,. The pure compound was

obtained by the column chromatography of the crude product on 100-150 mesh silica (Ethyl acetate and Hexane – 5:5) and purified 2, 4, 5-trisubstituted imidazole derivatives.

1) 2,4,5-triphenyl-1H-imidazole (4a):
Pale yellow solid, Yield-86%.m.p:251-253°C.1HNMR(400MHz,$CDCl_3$,)δppm:10.541(s,1H, NH-imidazole),8.214 (d, J = 8.8 Hz, 2H), 7.651 (d, J = 7.6 Hz, 4H), 7.450(t, J = 8.0 Hz, 2H), 7.384 (t, J = 9.2 Hz, 6H) 7.328 (d, J = 6.8 Hz, 2H). ^{13}C NMR (100MHz, $CDCl_3$) δppm: 175.26,135.45, 130.09, 129.68, 129.17, 128.92, 128.55, 128.08, 127.74, 127.13; Molecular weight (m/z): 297.38(M+H) ; Formula of compound: $C_{21}H_{16}N_2$;

2) 4-(4,5-Diphenyl-1H-imidazol-2-yl)-phenol (4b):
Light- yellow; Mp 252–254 ◦C.Yield-90%, FTIR (KBr,cm−1): 3554, 3441, 3286, 3056, 1702,1280; 1H NMR (400 MHz, DMSO-d6): 11.840 (s, 1H), 9.670 (s, 1H,-OH),7.790 (d,8.8 Hz,2H),7.654–7.321(m,10H), 6.964(d,8.0Hz,2H); 13CNMR (400MHz,CDCl3):173.26, 155.35, 136.09,130.19,129.44, 128. 88,128.24, 127.65, 120.33, 118.68; Molecular weight (m/z): 313.56(M+H) ; Formula of compound: $C_{21}H_{16}N_2O$;

3) 2-(4-methoxyphenyl)-4,5-diphenyl-1H-imidazole (4c):
Light- yellow m.p. -220-222°C.Yield-89%; ^{1HNMR} (400 MHz CDCl3) δppm: 11.246(s, 1H, NH-imidazole), 7.894 (d, J = 10.2 Hz, 2H), 7.596-7.412 (m, 6H, Ar-H), 7.736-7.114(m, 6H, Ar-H), 3.774(OCH3, s, 3H). 13C NMR (100MHz, CDCl3) δppm: 172.28, 157.35, 136.69, 130.09, 129.57, 128.69, 128.12, 120.64, 118.55, 54.86; Molecular weight (m/z):327.44(M+H); Formula of compound: $C_{22}H_{18}N_2O$;

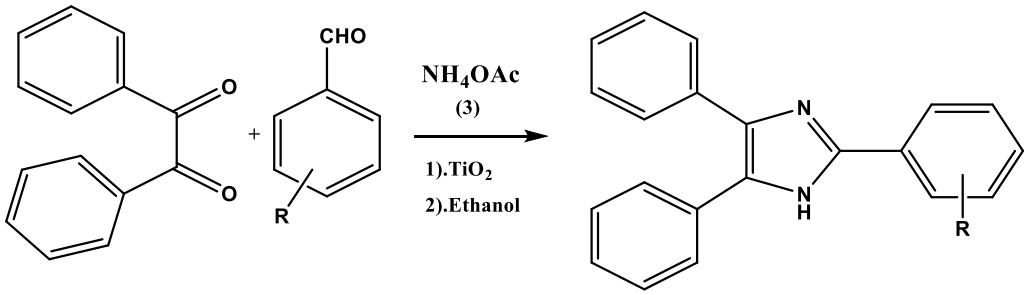
4) 4,5-diphenyl-2-(3,4,5-trimethoxyphenyl)-1H-imidazole (4d):
White sold,Mp:231–233◦C.Yield-89%;^{1}HNMR(400MHz,CDCl3):11.354(s,1H,NH-imidazole),7.482–7.287(m,10H,Ar-H), 7.148 (d,J=7.2Hz,2H, Ar-H),3.784(s,6H,(-OCH_3)$_2$), 3.627(s,3H,-OCH_3), ^{13}CNMR(400MHz,CDCl3):171.15, 155.85, 152.47, 137.07, 129.48, 128.81, 128.34, 127.55, 116.41, 112.26, 109.54,56.17,53.28;Molecular weight (m/z): 387.32(M+H) ; Formula of compound: $C_{24}H_{22}N_2O_3$;

5) 5-(dimethylamino)-2-(4,5-diphenyl-1H-imidazol-2-yl)phenol(4e):
Pale yellow, Mp:215–217◦C.Yield-89;1HNMR(400MHz,CDCl3):11.887(s,1H,NH-imidazole), 9.128(s,1H,-OH),7.501–7.293 (m,10H,Ar-H),7.258(d,J=7.2Hz,1H,Ar-H),6.874(d,J=9.2Hz, 1H,Ar-H),6.741(s,1H,Ar-H),2.284(s,6H,(-CH3)2), 13CNMR (400MHz, CDCl3):167.15, 153.07,150.12,136.04,129.13,128.77,128.26, 127.18, 114.75, 108.95,103.17,40.45;Molecular weight (m/z): 356.07(M+H); Formula of compound: $C_{23}H_{21}N_2O$;

6) 2-(2-bromo-3,4-dimethoxyphenyl)-4,5-diphenyl-1H-imidazole (4f) :
Palered;Mp:265–267°C.Yield-89%;^{1}HNMR($CDCl_3$,400MHz):11.456(NH-imidazole,s,1H), 7.524–7.385(m,10H,Ar-H),7.324 (d,J=8.0,1H, Ar-H),7156(d,J=8.8,1H,Ar-H),3.810(s,3H,-OCH_3),3.684(s,3H,-OCH_3),^{13}CNMR(400MHz,$CDCl_3$): 170.65, 151.58, 148.73,136.44, 132.51, 129.14, 128.88, 128.17, 127.91, 125.69, 120.24, 110.07, 58.34,55.71;Molecular weight (m/z): 436.56(M+2); Formula of compound: $C_{23}H_{19}BrN_2O_2$;

7) 2.3.4. 2-(4-Nitrophenyl)-4,5-diphenyl-1H-imidazole (4g)
Whitesold,Mp 245–247◦C.Yield-86%; FTIR (KBr, cm−1): 3457, 3058, 1676, and 1270; ^{1}HNMR (400 MHz, $CDCl_3$):11.991(s,1H,NH-imidazole),8.232(d,J=10.4,2H,Ar-H),8.076(d,J=8.0,2H,Ar-H),7.452–7.290(m,10H), ^{13}CNMR (400MHz,$CDCl_3$): 178.44, 147.06, 137.69, 134.36, 129.75, 129.16, 128.65, 128.19, 127.59, 125.28;; Molecular weight (m/z): 342.72(M+H) ; Formula of compound: $C_{21}H_{15}N_3O_2$;

3. Results and discussion



(1) (2) (4)

(4a-4l)

R= H, 4-OH, 4-OH-3-OC_2H_5, 4-OMe , 3,4,5$(OMe)_3$,
(4a) (4b) (4c) (4d) (4e)

4-C_2H_5, 2-OH-4-$N(CH_3)_2$, 4-Cl,
(4f) (4g) (4h)

4-Br, 2-Br-3,4$(OCH_3)_2$, 2-I-3,5$(OCH_3)_2$, 4-NO_2,
(4i) (4j) (4k) (4l)

(scheme-1)

Our research initially begins with a reaction between 1.125 mmol of benzil, 1.125 mmol of substituted aromatic aldehyde, and 2.50 mmol of ammonium acetate heated in ethanol under reflux in the presence of supported TiO_2 as catalyst. This reaction is produced the appropriate derivatives of 2,4,5-trisubstituted imidazole (Scheme-1). To get the ideal conditions, the reaction was preceded by variation of temperatures, solvent compositions, catalyst concentrations, time dependent and also nature of the substituted aryl aldehyde . After the reaction was finished, the TiO_2 catalyst was separated from the crude products. It was then cleaned with a diluted acid solution, water, and acetone before being vacuum-dried. The reusability of the catalyst for the performed to create of the model analogous "4e" under ideal conditions was investigated.

The spectroscopic methods are submitted in the characterization of the material for this article and also each of the derivatives can be explained and showed above data. The ^{1}HNMR spectra exhibited a peak at 11.974-10.541 δppm shown the presence of N-H proton, a peak between 8.232-6.584 δppm showed aromatic proton, a peak exhibited between 3.842-3.581 δppm represents methoxy protons, a peak shown between 9.851-9.128 δppm indicates hydroxyl proton(-OH) and a peak exhibited between 3.784-3.715 δppm represents $–CH_2$-protons.The methyl protons appears at1.123ppm appear at The ^{13}CNMR spectra recorded a peak maximum at 178.44 δppm is belongs to the carbonyl group in nitro substituted group. The LCMS spectra recorded the molecular weight of halogen substituted such as 4h,4i,4J and 4k showed (M+2) peak. The derivatives “4a,4b,4c,4d, 4e,4f and 4g” exhibited (M^{+}+H) peaks.

In order to optimize the catalyst and reaction conditions, the reaction was initially carried out without catalyst and without solvent. However, even after accelerating the reaction time, the desired product was not formed. Therefore, in order for the reaction to continue, the catalyst must be used. Considering the importance of catalyst, we chose a mild, inexpensive, nontoxic, very stable, and readily available transition metal catalyst. Throughout the reaction, different types of metal oxide catalysts were added at different temperatures and with different solvents, and these conditions were maintained until the reaction was finished. After the reaction was finished, it was found that TiO2 was the most profitable transition metal element in terms of producing the majority of the product, with the remaining catalyst producing fewer products than the catalyst with the same name. The remaining catalysts, which include MgO, TiO_2, SnO_2, ZnO, and others, contain non-transition metal particles more so than the others.

Table 1: Effective of catalyst for synthesis of 2, 4, 5-triaryl imidazoles (4d):

Entry	catalyst	Time (min)	Isolated Yields (%)
1	SnO_2	120	72
2	ZnO	120	68
3	TiO_2	120	94
4	MgO	110	45

Through the use of multiple mole ratios ranging from 0.5 to 2.5mmol, the catalyst loading was assessed in a solvent-free setting. When operating at room temperature without a catalyst, there was no observed product yield. However, after progressively raising the reaction temperature from room temperature to 50^{o}C, traces of the product were found again. The desired chemicals in the final product grew as the catalyst was added up to 2.5mmol at 70°C. A 3.0 mmole increase in catalyst concentration did not result in any appreciable improvement in the rat reaction. Therefore, Table-2 demonstrates that 2.5mmole equivalent of TiO_2 is adequate to finish the reaction and that there is no increase in product formation beyond the catalyst's saturation quantity.

Table 2: Effect of the loaded catalyst for the synthesis of 2, 4, 5-triphenyl-1H-imidazole (4d):

Entry	Amount of the catalyst (%mmole)[a]	Time (min)	Yield (%)[b]
1	0.5	120	Traces
2	1.0	120	41
3	1.5	120	68
4	2.0	120	73
5	2.5	120	94
6	3.0	125	94

a. Isolated product, **b**. 3, 4, 5-trimethoxybenzaldehyde (1.125 mmol), ammonium acetate (3 mmol), benzil (1.25 mmol).

After the aforementioned catalyst was used in the reaction, we used a range of solvents, such as H2O, CH3CN, DMF, ethanol, MeOH, and THF, to screen for solvent effects. According to Table-3, our observations indicate that the optimal circumstances for a reaction are those in which solvents are not used, and that the reaction is completed. Additionally, the yield of the desired product is higher in these settings than it is in any of the solvents studied.

Table 3: The solvent comparison for the synthesis of 2,4,5-triphenyl-1H-imidazole (4d):

Entry	solvent	Time(min)[c]	Yield (%)[d]
1	Water	120	Traces
2	Acetonitrile	120	68
3	DMF	120	52
4	Ethanpl	120	94
5	Methanol	120	65
6	THF	120	50

c. isolated product, **d**. 3, 4, 5-trimethoxybenzaldehyde (1.125 mmol), ammonium acetate (3 mmol), benzil (1.25 mmol)

First, substituted aromatic aldehydes were selected for the reaction with benzil and ammonium acetate in order to study the catalytic activity of nano catalysts such as TiO2. Improvements in reaction conditions allowed for the effective synthesis of 2,4,5-triphenyl-1H-imidazole in a solvent-free environment, even at temperatures as high as 100^{0}C, with a catalytic amount of TiO2. The outcomes, though, fell short. In order to conduct reactions at different temperatures, we added TiO2 to a variety of solvents (Table 1). In the ethanol-lactic acid system, we were able to get 94% of the product yield through experimentation. At 70°C (120 minutes), the optimal results are obtained with benzil, an aromatic aldehydes, and ammonium acetate in 1:1:3 equivalent ratios.

Table-3: Effect of temperature for synthesis of 2, 4, 5-triaryl imidazoles (4e)

Entry	Temperature (^{0}C)^e	Solvent	Time (min)	Yield (%)^f
1	RT	Ethanol	240	40
2	70	Ethanol	120	95
3	100	Ethanol	180	50
4	Above 100	Ethanol	180	45

e. isolated product, f. 3, 4, 5-trimethoxybenzaldehyde (1.125 mmol), ammonium acetate (3 mmol), benzil (1.25 mmol)

As the amount of TiO_2 in isopropanol increases, the product yield does not rise, according to the results (Table 1). Additionally, we limited our reactions to the TiO_2 medium, and the outcomes indicate a rise in the yield product in 25 mmole of the transition metal catalyst. To showcase the efficacy of this catalyst, a range of aryl aldehydes were subjected to optimal reaction conditions including 1,2-diketone and ammonium sources, resulting in the synthesis of 2, 4, 5-triaryl imidazole derivatives (4a-4g). This process is effective for both electron-donating and electron-withdrawing aromatic aldehydes. When the reaction time is right, the aromatic aldehyde with the electron withdrawing group yields a good yield, but the one with the electron donating group yields a little lower yield. Excellent yields were achieved in all reactions, which were finished within 30 minutes of the reaction period, and the specifics were stated in Table -1.

4. Conclusion

We have effectively developed synthesis of tri substituted imidazoles in this work. Additionally, the appropriate chemical shift values derived from an NMR spectroscopy result provided insight into the molecular structure. It was found that there was a good degree of agreement between the published literature and the experimental results. This method has a number of notable advantages, including as high yields of the final product, short reaction times, high efficiency, and ethanol as solvent reaction conditions. Its simplicity makes it a desirable alternative to the clean synthesis of biologically and medicinally significant 2, 4, 5-trisubstituted imidazoles. We have created the three basic and incredibly effective components—benzil, aromatic aldehydes, and ammonium acetate—that are supported by TiO2 when IPA is used as a solvent.

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