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Nano Titanium Dioxide: Efficient and Reusable Heterogeneous Nano Catalyst for Synthesis of 1,8-Dioxo-Decahydroacridines

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Abstract

3,3,6,6-Tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8-(2H,5H)-dione derivatives are synthesized by nano titanium dioxide as an efficient and reusable heterogeneous nano catalyst in a one pot multi component reaction. Easy preparation and separation of catalyst, simply workup procedure, clean reaction and reusability of the catalyst up to 7 times without appreciable loss of its catalytic activity and high yields are some advantages of this work.

1. Introduction

Nano titanium dioxide has been widely investigated in the past decades due to its multiple potential applications such as catalytic activity for esterification [1], rhodamine B degradation [2], crosslinking of cotton cellulose with succinic acid under UV [3], synthesis of β-acetamido ketones [4], synthesis of dibenzo[a,j]xanthenes [5], polyhydroquinoline [6] and 1,8-dioxo-octahydroxanthenes [7].

Acridines and their fused derivatives possess wide spectrum of biological activities, viz. antibacterial [8], fungicidal [9], anti-tubercular [10, 11], antimalarial [12], anti-tumor [13], anti-cancer [14], acetylcholine esterase inhibitory [15], vasorelaxing [16] and anti-viral [17]. Therefore, the synthesis of acridine derivatives has great importance. 1,8-Dioxodecahydroacridines and their derivatives are generally synthesized in a three-component cyclo condensation of dimedone (2 equiv.), aromatic aldehydes (1 equiv.) and ammonium acetate or amines in the present of several

catalysts such as $[B(C_6F_5)_3]$ [18], p-dodecylbenezenesulfonic acid (DBSA) [19], brønsted acidic imidazolium salts [23]. This work report synthesis of 1,8-dioxo-decahydroacridine derivatives catalyzed by nano TiO_2 as an efficient, high reusable and highly active heterogeneous nano catalyst in solvent free condition.

2. Experimental procedure

2.1. General.

Chemicals were purchased from the Merck and Aldrich chemical companies. Thin Layer Chromatography (TLC) on commercial aluminum backed plates of silica gel 60 F254 was used to monitor the progress of reactions. The products were characterized by FT-IR spectra, ^{1}H NMR, ^{13}C NMR. FT-IR spectra were recorded on Shimadzu FT-IR-8400 instrument. ^{1}H and ^{13}C NMR spectra were recorded on Bruker Advance Spectrometer 400 & 500 MHz using DMSO-d^6 as solvent. The chemical shifts are expressed in parts per million (ppm) and tetramethylsilane (TMS) was used as an internal reference. Elemental analyses were performed by Perkin Elmer CHN analyzer, 2400 series II. Melting points were recorded on a THERMO SCIENTIFIC 9100 apparatus. X-ray diffraction analyses were conducted using a Bruker D8 X-ray diffractometer (Cu-Ka radiation, k = 1.54) were obtained for characterization of the heterogeneous catalyst. Transmission electron microscopy (TEM) was carried out with Philips CM10 instrument. Thermo gravimetric analyses (TGA) were conducted on a Du Pont 2000 thermal analysis apparatus under air atmosphere at a heating rate of 5 °C/min.

proline [20], amberlyst-15 [21], NH_4Cl [22] and

2.2. Preparation of nano TiO_2.

Nano TiO_2 was prepared with hydrothermal method. The pH value of $TiCl_4$ solution was adjusted to 1.8 with $NH_3.H_2O$. After vigorously stirring at 70 °C for 2 h, the final pH of the solution was adjusted to 6. Then the resulting suspension was cooled down to room temperature and kept for 24 h. After filtration, the solid was firstly washed with NH_4Ac-HOAc until no Cl^- was detected, then separated out with a centrifuge and washed with ethanol, at last dried in vacuum. After 2 h treatment at 650 °C, nano TiO_2 material was obtained [24].

2.3. General procedure for the synthesis of 1,8-dioxo-decahydroacridine derivatives in presence of nano TiO_2.

A mixture of different aromatic aldehydes (1 mmol), 1,3-diketone (2 mmol), ammonium acetate (1 mmol) as the nitrogen source and nano TiO_2 (10 mol%) as catalyst were poured into a test tube in EtOH as solvent under reflux condition were stirred for an appropriate time. The progress of the reaction was monitored by TLC. After completion of the reaction, because of the heterogeneous catalyst was insoluble in ethanol, it was removed by centrifuged, filtration and the solution was cooled to room temperature and pure product was crystallized in ethanol to form crystals.

2.4. Spectral data of some selected compound.

*3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8-(2H,5H)-dione (**1**):*

IR (KBr, Cm^{-1}) ν max: 3279, 3209, 3063, 2955, 2932, 1636, 1605, 1481, 1365, 1219, 1142.^{1}H NMR (400 MHz, DMSO-d^6): δ 0.84 (s, 6H), 0.99 (s, 6H), 1.96 (d, J= 21.5 Hz, 2H), 2.15 (d, J= 21.5

Hz, 2H), 2.30 (d, J= 22.8 2H), 2.40-2.49 (m, 2H), 4.8 (s, 1H), 7.00-7.14 (m, 5H), 9.28 (s, 1H).^{13}C NMR (100 MHz, DMSO-d^6): δ26.45, 29.13, 32.14, 32.86, 50.24, 111.47, 125.46, 127.56, 127.63, 147.15, 149.32, 194.36. Anal. Calc. for $C_{23}H_{27}NO_2$; C 79.05, H 7.79, N 4.01, O 9.16; Found: C 79.08, H 7.73, N 4.06, O 9.11.

9-(4-fluorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione ***(4):***

FT-IR (KBr, Cm^{-1}) ν max: 3280, 3197, 3070, 2954, 2869, 1635, 1608, 1570, 1437, 1364, 1256, 1145.. ^{1}H NMR (400 MHz, DMSO-d^6): δ: 0.99 (s, 6H), 1.11 (s, 6H), 2.15-2.26 (dd, J= 28, 16 Hz, 4H), 2.46 (s, 4H), 4.73 (s, 1H), 6.88-6.92 (t, J= 8.8 Hz, 2H), 7.24-7.27 (d, J= 10.8 Hz, 2H), 9.33 (s, 1H).^{13}C NMR (100 MHz, DMSO-d^6): δ 27.29, 29.26, 31.22, 32.20, 50.73, 111.94,126.07, 127.80, 127.87, 147.17, 149.30, 149.60,194.46. Anal. Calc. for $C_{23}H_{26}FNO_2$; C 75.18, H 7.13, N 3.81, O 8.71; Found: C 75.13, H 7.18, N 3.78, O 8.76.

9-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione ***(5):***

FT-IR (KBr, Cm^{-1}) ν max: 3288, 3205, 3068, 2956, 2869, 1643, 1608, 1475, 1363, 1223, 1171. ^{1}H NMR (500 MHz, DMSO-d^6): δ: 0.91 (s, 6H), 1.04 (s, 6H), 2.08-2.11 (d, J= 16, 2H), 2.26-2.29 (d, J= 16, 2H), 2.50-2.60 (dd, J= 14.65, 4H), 4.51 (s, 1H), 7.18-7.20 (d, J= 8.2 Hz, 2H), 7.28-7.29 (d, J= 8.2 Hz, 2H), 9.32 (s, 1H). Anal. Calc. for $C_{23}H_{26}ClNO_2$; C 71.96, H 6.83, N 3.65, O 8.33; Found: C 71.90, H 6.89, N 3.59, O 8.32.

9-(4-bromophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione ***(6):***

FT-IR (KBr, Cm^{-1}) ν max: 3274, 3176, 3058, 2954, 2921, 1649, 1608, 1491, 1364, 1221, 1171. ^{1}H NMR (400 MHz, DMSO-d^6): δ: 1.03 (s, 6H), 1.23 (s, 6H), 2.24-2.30 (dd, J= 14.8, 8.8 Hz, 4H), 2.44 (s, 4H), 4.84 (s, 1H), 7.16 (d, J= 6.8 Hz, 2H), 7.43 (d, J= 17.6 Hz, 2H), 9.30 (s, 1H). Anal. Calc. for $C_{23}H_{26}BrNO_2$; C 64.49, H 6.12, N 3.27, O 7.47; Found: C 64.52, H 6.17, N 3.31, O 7.49.

3,3,6,6-tetramethyl-9-(4-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione ***(7):***

FT-IR (KBr, Cm^{-1}) ν max: 3384, 2958, 2908, 1643, 1602, 1514, 1447, 1364, 1221, 1171. ^{1}H NMR (500 MHz, DMSO-d^6) δ: 0.91 (s, 6H), 1.05 (s, 6H), 2.08-2.11 (d, J= 6.5 Hz, 2H), 2.27-2.30 (d, J= 6.5 Hz, 2H), 2.50-2.63 (m, 4H), 4.63 (s, 1H), 7.45-7.48 (m, 2H), 8.10-8.12 (m, 2H), 9.32 (s, 1H). Anal. Calc. for $C_{23}H_{26}N_2O_4$; C 70.03, H 6.64, N 7.10, O 16.22; Found: C 70.08, H 6.69, N 7.07, O 16.26.

3,3,6,6-tetramethyl-9-(3-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione ***(8):***

FT-IR (KBr, Cm^{-1}) ν max: 3282, 3193, 3070, 2957, 2930, 2889, 2870, 1649, 1612, 1577, 1434, 1364, 1225, 1171. ^{1}H NMR (500 MHz, DMSO-d^6): δ: 0.91 (s, 6H), 1.05 (s, 6H), 2.09-2.12 (d, J= 4.4 Hz, 2H), 2.27-2.30 (d, J= 6.4 Hz, 2H), 2.55-2.63 (dd, J= 7.2 Hz,4H), 4.64 (s, 1H), 7.55-7.58 (t, J= 3.1 Hz, 1H), 7.65-7.67 (d, J= 3 Hz, 1H), 7.99-8.02 (d, J= 5.1 Hz, 2H), 9.31 (s, 1H). Anal. Calc. for $C_{23}H_{26}N_2O_4$; C 70.03, H 6.64, N 7.10, O 16.22; Found: C 70.06, H 6.70, N 7.08, O 16.24.

9-(4-methoxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione ***(9):***

IR (KBr, Cm^{-1}) ν max: 3277, 3203, 3070, 2959, 2870, 1643, 1606, 1508, 1483, 1367, 1223, 1171.^{1}H NMR (500 MHz, DMSO-d^6): δ 0.90 (s, 6H), 1.04 (s, 6H), 2.08 (d, J= 9.7 Hz, 2H), 2.26 (d, J= 9.9 Hz, 2H), 2.48-2.52 (m, 2H), 2.57 (d, J= 10.8 Hz, 2H), 3.68 (s, 3H), 4.7 (s, 1H), 6.77-6.78 (t, J=

4.1 2H), 7.06-7.08 (t. J= 5.12 Hz, 2H), 9.21 (s, 1H). Anal. Calc. for $C_{24}H_{29}NO_3$; C 75.96, H 7.70, N 3.69, O 12.65; Found: C 75.90, H 7.72, N 3.62, O 12.69.

3,3,6,6-tetramethyl-9-(naphthalen-2-yl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione ***(11)****:*

IR (KBr, Cm^{-1}) ν max: 3274, 3176, 3059, 2954, 2922, 1649, 1608, 1491, 1364, 1221, 1171.. ^{1}H NMR (400 MHz, DMSO-d^6): δ 0.99 (s, 6H), 1.12 (s, 6H), 2.13-2.18 (d, J= 18 Hz, 2H), 2.21-2.23 (d, J= 9.6 Hz, 2H), 2.52 (s, 4H), 4.92 (s, 1H), 7.34-7.38 (d, J= 16, 1H), 7.46-7.47 (d, J= 6.8 Hz, 2H), 7.73-7.79 (m, 4H), 9.12 (s, 1H). Anal. Calc. for $C_{27}H_{29}NO_2$; C 81.17, H 7.32, N 3.51, O 8.01; Found: C 81.22, H 7.35, N 3.54, O 8.08.

9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione ***(12)****:*

IR (KBr, Cm^{-1}) ν max: 3273, 3196, 3080, 1624, 1601, 1483, 1218, 1140. ^{1}H NMR (400 MHz, DMSO-d^6): δ 1.90-2.09 (m, 4H), 2.26-2.28 (m, 2H), 2.30-2.38 (m, 2H), 2.51-2.64 (m, 4H), 4.79 (s, 1H), 7.05-7.09 (t, J= 7.6, 2H), 7.11-7.12 (d, J= 8.4 Hz, 1H), 7.23-7.31 (dd, J= 8, 24.2, 2H), 9.17 (s, 1H). Anal. Calc. for $C_{19}H_{19}NO_2$; C 77.79, H 6.53, N 4.77, O 10.91; Found: C 77.73, H 6.59, N 4.71, O 10.92.

9-(4-bromophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione ***(14)****:*

IR (KBr, Cm^{-1}) ν max: 3340, 3244, 2958, 2927, 1647, 1627, 1549, 1450, 1367, 1227, 1171. ^{1}H NMR (400 MHz, DMSO-d^6): δ 1.92-2.08 (m, 4H), 2.28-2.32 (m, 2H), 2.34-2.40 (m, 2H), 2.53-2.65 (m, 4H), 4.76 (s, 1H), 7.13-7.27 (m, 2H), 7.33-7.43 (dd, J= 30.6, 8.4 Hz, 2H), 9.38 (s, 1H).^{13}C NMR (100 MHz, DMSO-d^6): δ 20.10, 20.27, 27.13, 31.39, 36.76, 36.90, 112.08, 117.66, 127.83, 129.43, 146.47, 151.59, 171.19, 194.58. Anal. Calc. for $C_{19}H_{18}BrNO_2$; C 61.30, H 4.87, N 3.76, O 8.60; Found: C 61.33, H 4.81, N 3.71, O 8.69.

9-(p-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione ***(15)****:*IR (KBr, Cm^{-1}) ν max: 3286, 3203, 3068, 2943, 2887, 1639, 1608, 1458, 1364, 1232, 1176.^{1}H NMR (400 MHz, DMSO-d^6): δ 1.96-2.07 (m, 4H), 2.26 (s, 3H), 2.29-2.42 (m, 4H), 2.52-2.69 (m, 4H), 4.79 (s, 1H), 7.03-7.05 (d, J= 8 Hz, 2H), 7.12-7.21 (d, J= 7.6 Hz, 2H), 9.18 (s, 1H).^{13}C NMR (100 MHz, DMSO-d^6): δ 20.31, 21.06, 27.15, 31.22, 36.98, 55.14, 113.52, 117.03, 128.25, 128.84, 129.33, 135.86, 146.36, 150.53, 169.57, 194.52. Anal. Calc. for $C_{20}H_{21}NO_2$; C 78.15, H 6.89, N 4.56, O 10.41; Found: C 78.19, H 6.83, N 4.58, O 10.45.

3. Results and discussion

Initially, nano TiO_2 powder was easily prepared according to the reported procedure [24]. Figure 1 shows the XRD patterns of prepared nano TiO_2 powder.

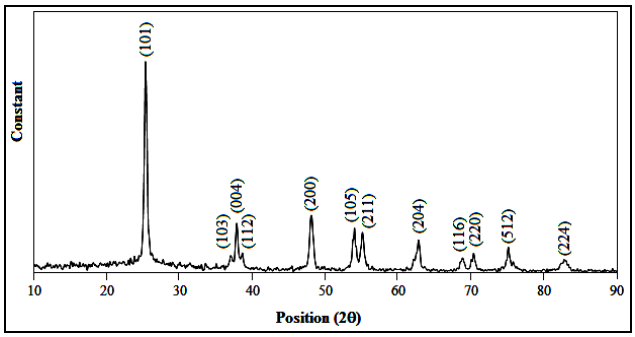


Fig. 1. The X-ray diffraction patterns of the nano TiO_2.

As it can be seen in Figure 1, the following peak signals at (101), (103), (004), (112), (200),(105), (211), (204), (220), (215) and (224) planes confirm that the formation of anatase crystal phase mostly, which coincides with JCPD 89-4921 standard. Moreover, the main phase was nano TiO_2 powder and any other phase was not observed. The size of the nano TiO_2 powder was also determined by X-ray line broad using the Debye-Scherrer formula given as D =

$0.9\lambda/\beta\cos\theta$, where D is the average crystalline size, λ the X-ray wavelength used, β the angular line width at half maximum intensity and θ the Bragg's angle. The average size of the nano TiO_2 powder for $2\theta = 25.303^o$ was estimated to be around 21.88 nm.

Transmission Electron Microscopy (TEM) analysis was used for characterization of nano TiO_2 powder (Figure 2). The TEM image reveals the spherical nano TiO_2 powder with average size 20-30 nm.

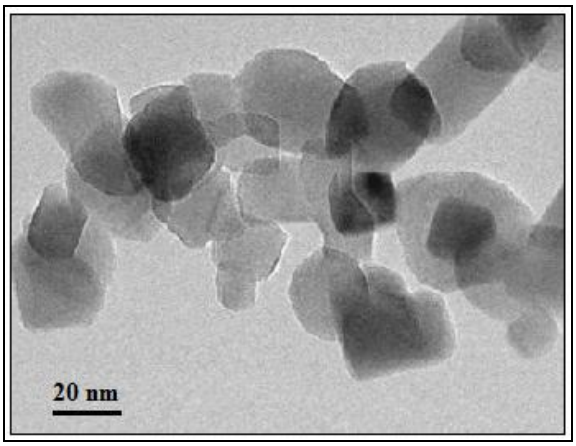


Fig. 2. The TEM image of the nano TiO_2.

The FT-IR spectra of nano TiO_2 powder are shown in Figure 3. The bending vibrations of adsorbed water molecules and stretching vibrations of O-H around 1630 and 3350-3450 cm^{-1}, respectively [25]. The broad intense band at 520 and 689 cm^{-1} as due to Ti-O-Ti vibration which is consistent with the reported IR spectra for nano TiO_2 [25].

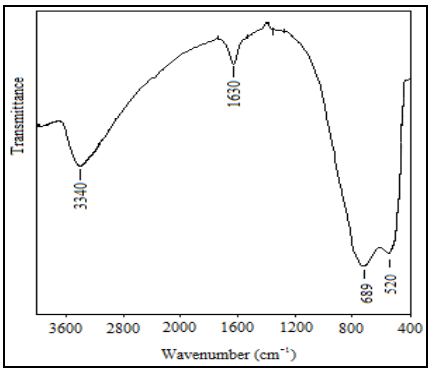


Fig. 3. The FT-IR spectra of the nano TiO_2 powder.

Thermo gravimetric analysis (TGA) of nano TiO_2 is shown in Figure 4. The TGA curve displays a weight loss (4 wt.%) below 100 °C which corresponds to the loss of the physically adsorbed water. Also, there is a slight weight loss (1 wt.%) between 100 °C and 800 °C, which possibly corresponds to the dehydroxylation of nano TiO_2. Also, from the TGA, we understood that nano TiO_2 has a greater thermal stability (up 400 °C) confirming that it could be safely used in organic reactions at temperatures in the range of 80-150 °C.

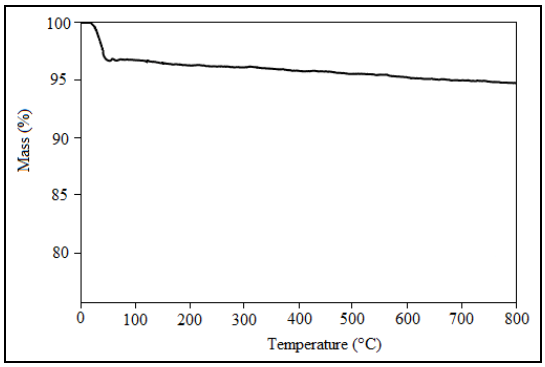
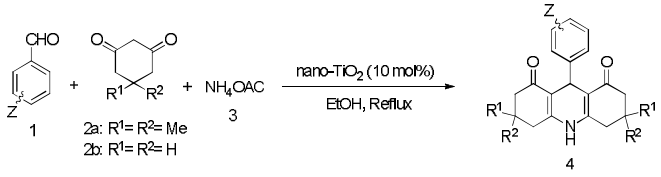


Fig. 4. TGA cure of the nano TiO_2 powder.

To examine catalytic activity of the nano TiO_2, the condensation of benzaldehyde (**1**) (106.0 mg, 1 mmol), dimedone (**2**) (280.0 mg, 2 mmol) and ammonium acetate (**3**) (77.1 mg, 1 mmol) was investigated (Scheme 1).



Scheme 1

To optimize the reaction conditions, amount of the nano TiO_2, different temperature and solvents were screened. Initially, to investigate the effect of solvent, it has been decided to carry out model reaction in present different solvents such as H_2O,

EtOH, CH_3CN, $CHCl_3$, Toluene under reflux conditions (Table 1, entries **1-5**) and solvent free condition at 100 °C in present 10 mol% of nano TiO_2 (Table 1, entry **6**). It was found that H_2O, CH_3CN, $CHCl_3$, Toluene and the solvent free system were unfavorable for the formation of the product (Table 1, entries **1-4** and **6**). Then the best yield of product was provided in EtOH under reflux condition.

Table 1. The Effect of different conditions on synthesis of 1,8-dioxo-decahydroacridines[a].

Entry	Nano TiO_2 (mol%)	Temp. (°C)	Solvent[b]	Time (h)	Yield[c] (%)
1	10	Reflux	H_2O	3	18
2	10	Reflux	CH_3CN	3	37
3	10	Reflux	$CHCl_3$	3	48
4	10	Reflux	Toluene	3	56
5	10	Reflux	EtOH	1	90
6	10	100	-	3	58
7	-	Reflux	EtOH	1	5
8	4	Reflux	EtOH	1	67
9	8	Reflux	EtOH	1	83
10	12	Reflux	EtOH	1	89
11	10	25	EtOH	3	34
12	10	40	EtOH	3	51
13	10	60	EtOH	3	76

[a] Reaction Condition: **1** (1 mmol), **2** (2 mmol) and **3** (1 mmol).
[b] 2 ml solvent.
[c] Isolated yield.

In the next step, the amount of catalyst was examined. The results showed that by increasing the catalytic load of nano TiO_2 from 0 to 10 mol%, the yield improved from 5 to 90% (Table 1, entries **5**, **7-9**). But more increasing in the catalyst amount did not increase the obtained yield (Table 1, entry **10**).

Eventually, the effect of temperature in present 10 mol% of nano TiO_2 was investigated. Obtained results showed that the product was obtained in excellent yield if the reaction was carried out in EtOH at reflux condition (Table 1, entry **6**).

Now, with different optimized conditions in hand [nano TiO_2 (10 mol%) as catalyst in EtOH under reflux conditions], some experiences were investigated to study the scope of this procedure. For this purpose, condensation between various aromatic aldehydes (including aldehydes with electron-releasing substituents, electron-withdrawing substituents and halogens on the aromatic ring), cyclic 1,3-diketone compounds and ammonium acetate were carried out to provide corresponding products in good to excellent yields. Entertainingly, synthesized acridine derivatives were prepared without any byproducts. The results are summarized in Table 2.

Table 2. Synthesis of 1,8-dioxodecahydroacridines (4) by nano TiO_2 as a reusable catalyst[a].

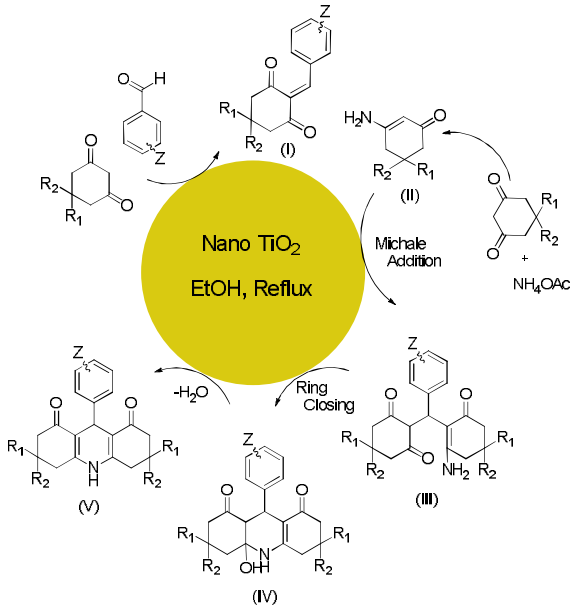
Entry	Ar	2	Time (min)	Yield (%)	Mp (°C)
1	$-C_6H_5$	**2a**	90	85	190-191 [9]
2	2-Cl-C_6H_4	**2a**	85	89	310-312 [26]
3	4-HO-C_6H_4	**2a**	95	86	>300 [26]
4	4-F-C_6H_4	**2a**	75	89	292-294
5	4-Cl-C_6H_4	**2a**	80	89	299-302 [8]
6	4-Br-C_6H_4	**2a**	85	87	308-313 [26]
7	4-O_2N-C_6H_4	**2a**	60	91	284-286 [8]
8	3-O_2N-C_6H_4	**2a**	60	90	299-301 [8]
9	4-H_3CO-C_6H_4	**2a**	105	82	300-301 [8]
10	4-H_3C-C_6H_4	**2a**	100	87	268-270 [8]
11	2-Naphtyl	**2a**	95	88	>330 [26]
12	$-C_6H_5$	**2b**	90	86	278-280 [9]
13	4-Cl-C_6H_4	**2b**	85	88	277-279[27]
14	4-Br-C_6H_4	**2b**	85	89	310-312
15	4-H_3C-C_6H_4	**2b**	100	86	254-256
16	4-H_3CO-C_6H_4	**2b**	100	87	302-305 [27]

[a] reaction condition: **1** (1 mmol), **2** (2 mmol), **3** (1 mmol), n-TiO_2 (10 mol%) in 2ml ethanol under reflux condition.
[b] Isolated yield.

As indicated in Table 2, the reaction works easily for a vast variety of aromatic aldehydes with both electron-donating and electron-withdrawing groups and different cyclic diketone to give corresponding 1,8-dioxo-decahydroacridine derivatives in good to excellent yields. In almost all cases, the reactions proceeded smoothly within

60-105 min. However, it is notable that substituted aromatic aldehydes with electron-withdrawing groups increase the rate of reaction (Table 2 entries **4**, **5**, **6**, **7**, **8**, **13** and **14**) probably by activating the carbonyl group as electrophile center. Contrarily in the case of electron-donating groups, the reaction was more slowly and also the obtained yields were decreased (Table 2, entries **3**, **9**, **10**, **15** and **16**).

A proposed mechanism for the synthesis of 1,8-dioxo-decahydroacridines is shown in scheme 2.



Scheme 2

In this proposed mechanism, 1 mmol of cyclic diketone reacts with 1 mmol aromatic aldehyde to produce intermediate **I**. On the other hand, 1 mmol of dimedone reacts with ammonium acetate to produce enamine **II**. Then, Michael addition of intermediate **I** with enamine **II**, gives adduct **III**. After the ring closing reaction, adduct **III** changes to intermediate **IV**. Finally, by elimination of water acridine **V** is obtained.

For testing the reusability of catalyst concerning green chemistry aspects, the mixture of reaction was washed with hot ethanol, catalyst separated with simple filtration and dried at 50 ºC for 2h. The condensation of **1** (1 mmol), **2** (2 mmol) and **3** was repeated with recovered catalyst without any loss of activity. The catalyst could be efficiently recovered and recycled up to seven times without suffering any significant drop in its catalytic activity or the yield for the reaction (Figure 5).

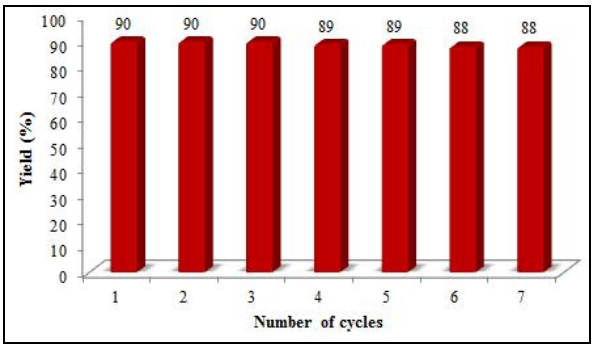


Fig. 5. Recycled nano TiO_2 catalyzed condensation between dimedone (1mmol), benzaldehyde (1 mmol) and ammonium acetate (1 mmol) within 90 minute.

4. Conclusion

In summary, nano TiO_2 as a heterogeneous nano catalyst has been used for the synthesis of different acridines under reflux conditions. This reaction leads to different 3, 3, 6, 6-tetramethyl-9-phenyl-3, 4, 6, 7, 9, 10-hexahydroacridine-1, 8-(2H, 5H)-diones in good to excellent yields. The present reported conditions have several advantages such as easy preparation of catalyst, high yields of products, having no environmental hazards and reusability of the catalyst for at least 7 times.

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