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Nanocrystalline TiO_2 as an efficient and reusable catalyst for the one-pot synthesis of polyhydroquinolien derivatives via Hantzsch reaction

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ABSTRACT

An efficient synthesis of polyhydroquinoline derivatives was reported via four-component coupling reactions of aldehydes, 1,3-dicarbonyl ketones (dimedone or 1,3-cyclohexanedione), ethyl acetoacetate or methyl acetoacetate and ammonium acetate in the presence of a catalytic amount of nanocrystalline TiO_2 under solvent free conditions. The reported method is mild, rapid and has the advantages such as heterogeneous catalysis, simple work-up procedure, recyclability of the catalyst and purification of products without chromatographic methods.

Keywords: Nanocrystalline TiO_2, Hantzsch reaction, Polyhydorquinolie derivatives, Heterogeneous catalysis, one-pot condensation reaction.

1. Introduction

Multi-component reactions have emerged as an efficient and powerful tool in modern synthetic organic chemistry allowing the facile creation of several new bonds in a one-pot reaction. Polyhydroquinoline derivatives compounds are well known as calcium channel modulators and have emerged as one of the most important classes of drugs for the treatment of cardio-vascular diseases [1,2]. Cardiovascular agents such as nifedipine, nicardipine, amlodipine, and other related derivatives are dihydropyridyl compounds, effective in treatment of hypertension [3,4]. Extensive studies have revealed that these compounds exhibit various medicinal functions such as neuroprotectant, platelet anti-aggregatory activity, cerebral antischemic activity in the treatment of Alzheimer's disease, and chemosensitizer in tumor therapy [5-8].

Several methods have been developed for the preparation of polyhydroquinoline derivatives by using various catalysts, including the use of microwave and ultrasound [9,10] ionic liquids [11,12], $Bu_4N^+HSO_4^-$ [13], in situ generated HCl [14], $K_7[PW_{11}CoO_{40}]$ [15], I_2 [16], silica-supported acids [17, 18], silica perchloric acid ($HClO_4$–SiO_2) [19], ZnO-nanoparticles [20], $Sc(OTf)_3$ [21] HY-Zeolite [22], montmorillonite K-10 [23], *p*-TSA [24], on grinding [25] and in EtOH [26]. Therefore, the development of an efficient method for the synthesis of these compounds is an active ongoing research area and there is scope for further improvement toward milder reaction conditions and higher yields.

In recent years, there has been considerable growth of interest in the catalysis of organic reactions by solid acid catalysts. Solid acid catalysts provide numerous opportunities for recovering and recycling catalysts from reaction environments. These features can lead to the improvement of the processing steps, better economical processes, and environmentally friendly industrial manufacturing. After mesoporous TiO_2 was first synthesized by a sol–gel process with phosphorous surfactants as templates by Antonelli and Ying [27], various methods of surfactant templating have been developed for the preparation of mesoporous structures of TiO_2 [28]. Since mesoporous materials normally possess large surface area and narrow pore size distribution, which advantageously make them a versatile candidate in the catalysis field, the utilization of mesoporous TiO_2 in many catalytic reactions becomes feasibly attractive [29-33].

2. Experimental

2.1. General

Chemicals were purchased from Fluka, Merck, and Aldrich chemical companies. Melting points (Mp) were recorded on a Barnstead electrothermal instrument and are uncorrected. The IR and FT-IR spectra (cm^{-1}) were recorded on Shimadzu Corporation 200-91-527 and Perkin Elmer RX1 spectrophotometers as pallets on KBr discs. The ^1H and ^{13}C NMR spectra were recorded on Bruker AVANCEIII-400

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spectrometer in $CDCl_3$ using TMS as an internal reference. The purity determination of the substrate and reaction monitoring were accompanied by TLC on silica-gel polygram SILG/UV 254 plates. All yields refer to the isolated products and the known products were characterized by their physical constants and comparison with authentic samples.

2.2. General procedure

A mixture of aldehyde (1 mmol), dimedone or 1,3-cyclohexanedione (1 mmol), ammonium acetate (1.1 mmol), ethylacetoacetate or methylacetoacetate (1 mmol), and TiO_2 (5 mg, It is a mixture of 80% anatase and 20% rutile and it has a particle size of 30 nm and BET specific area 34 m^2g^{-1}) was heated at 70 °C with stirring for 25-50 min. After completion of the reaction as indicated by TLC, the reaction mixture was washed with cool water then hot ethanol was added to the mixture and the catalyst was filtered off. The pure product was obtained by recrystallization from ethanol. The catalyst recovered by filtration, was washed with ethanol (10 mL), dried at room temperature and reutilized four times for the same reaction.

The Selected spectral data:

The physical (Mp) and spectral data (IR, ^1H and ^{13}C NMR) of polyhydroquinolie derivatives (5a-o) are presented below:

5a: Pale yellow solid, m.p. 224-226 ^oC; IR (KBr): 3295, 2995, 1695, 1643, 1605, 1460, 1375, 1220, 1180 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ= 7.1-7.34 (m, 5H, Ar-H), 6.70 (br s, 1H, NH), 5.08 (s, 1H), 4.09 (q, *J*=7.2 Hz, 2H), 2.36 (s, 3H), 2.15-2.35 (m, 4H) , 1.24 (t, *J*=7.2 Hz, 3H), 1.08 (s, 3H), 0.95 (s, 3H).

5b: Pale yellow solid, m.p. 243-245 ^oC; IR (KBr): 3295, 2995, 1685, 1650, 1604, 1470, 1378, 1220, 1180 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$, ppm) δ: 7.26 (d, J=8.4 Hz, 2H, Ar–H), 7.18 (d, *J*=8.4 Hz, 2H, Ar–H), 6.50 (br s,1H, NH), 5.05 (s, 1H), 4.08 (q, *J*=7.2 Hz, 2H), 2.38 (s,3H), 2.14-2.34 (m, 4H), 1.21 (t, *J*=7.2 Hz, 3H), 1.09 (s, 3H), 0.94 (s, 3H).

5c: Pale yellow solid, m.p. 254-256 ^oC; IR (KBr): 3295, 2990, 1692, 1645, 1604, 1475, 1375, 1220, 1180 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$, ppm) δ: 7.34 (d, *J*=8.4 Hz, 2H, Ar–H), 7.21 (d, *J*=8.4 Hz, 2H, Ar–H), 6.12 (br s,1H, NH), 5.04 (s, 1H), 4.08 (q, *J*=7.2 Hz, 2H), 2.40 (s, 3H), 2.15-2.37(m, 4H) , 1.22 (t, *J*=7.2 Hz, 3H), 1.09 (s, 3H), 0.95 (s, 3H).

5d: Yellow solid, m.p. 174-176 ^oC; IR (KBr): 3298, 2995, 1695, 1645, 1610, 1480, 1375, 1220, 1180 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$, ppm) δ: 8.14 (s, 1H, Ar–H), 7.99 (dd, J_1=8 Hz, J_2= 1.2 Hz, 1H, Ar–H), 7.74 (d, *J*=8 Hz,1H, Ar–H), 7.40 (t, *J*=8 Hz, 1H, Ar–H), 6.70 (br s,1H, NH), 5.18 (s, 1H), 4.08 (q, *J*= 7.2 Hz, 2H), 2.40 (s, 3H), 2.14-2.32 (m, 4H), 1.24 (t, *J*=7.2 Hz, 3H), 1.10 (s, 3H), 0.94 (s, 3H).

5e: Yellow solid, m.p. 241-243 ^oC; IR (KBr): 3290, 2985, 1697, 1645, 1610, 1460, 1378, 1220, 1180 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$, ppm) δ: 8.10 (d, *J*=8.8 Hz, 2H, Ar–H), 7.51 (d, *J*=8.8 Hz, 2H, Ar–H), 6.72 (br s, 1H, NH), 5.18 (s, 1H), 4.07 (q, *J*=7.2 Hz, 2H), 2.40 (s, 3H), 2.13-2.37 (m, 4H), 1.24 (t, *J*=7.2 Hz, 3H), 1.09 (s, 3H), 0.92 (s, 3H).

5f: Pale yellow solid, m.p. 255-257 ^oC; IR (KBr): 3287, 2995, 1695, 1645, 1605, 1487, 1378, 1220, 1180 cm^{-1}.^1H NMR (400 MHz, $CDCl_3$, ppm) δ: 7.23 (d, *J*=8.4 Hz, 2H, Ar–H), 6.75 (d, *J*=8.4 Hz, 2H, Ar–H), 6.62 (br s, 1H, NH), 5.02 (s, 1H), 4.09 (q, *J*=7.2 Hz, 2H), 3.74 (s, 3H), 2.36 (s, 3H), 2.18-2.27 (m, 4H), 1.24 (t, *J*=7.2 Hz, 3H), 1.07 (s, 3H), 0.95 (s, 3H).

5g: Pale yellow solid, m.p. 179-181 ^oC; IR (KBr): 3292, 2995, 1698, 1650, 1610, 1478, 1370, 1220, 1180 cm^{-1}. ^1H NMR (400 MHz , $CDCl_3$, ppm) δ: 7.22 (d, *J*=8 Hz, 2H, Ar–H), 7.06 (d, *J*=8 Hz, 2H, Ar–H), 6.77 (br s, 1H, NH), 5.05 (s, 1H), 4.09 (q, *J*=7.2 Hz, 2H), 2.79-2.86 (m,1H), 2.16-2.37 (m,7H), 1.19-1.28 (m, 9H), 1.09 (s, 3H), 0.98 (s, 3H).

5h: Yellow solid, m.p. 245-247 ^oC; IR (KBr): 3295, 2970, 1690, 1605, 1480, 1375, 1220, 1179 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$, ppm) δ: 7.21 (d, *J*=0.8 Hz,1H), 6.23 (t, *J*=3.2 Hz, 1H), 6.04 (d, *J*=3.2 Hz, 1H), 6.0 (br s, 1H, NH), 5.28 (s, 1H), 4.12-4.22 (m, 2H), 2.20-2.41 (m,6H), 1.25-1.30 (m, 4H), 1.12 (s, 3H), 1.05 (s, 3H).

5i: Pale yellow solid, m.p. 254-256 ^oC; IR (KBr): 3295, 2970, 1695, 1610, 1475, 1375, 1220, 1170 cm^{-1}. ^1H NMR (400 MHz ,$CDCl_3$, ppm) δ: 7.26 (d, *J*=8.4 Hz, 2H, Ar–H), 7.19 (d, *J*=8.4 Hz, 2H, Ar–H), 6.05 (br s, 1H, NH), 5.06 (s, 1H), 3.64 (s,3H), 2.41 (s, 3H), 2.16-2.39 (m, 4H), 1.10 (s, 3H), 0.95 (s, 3H).

5j: Pale yellow solid, m.p. 256-258 ^oC; IR (KBr): 3298, 2980, 1695, 1600, 1480, 1375, 1220, 1180 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$, ppm) δ: 7.18 (d, *J*=8.8 Hz, 2H, Ar–H), 6.64 (d, *J*= 8.8 Hz, 2H, Ar–H), 6.09 (br s, 1H, NH), 4.99 (s, 1H), 3.64 (s, 3H), 2.90 (s, 6H), 2.39 (s, 3H), 2.17-2.34 (m, 4H), 1.09 (s, 3H), 0.98 (s, 3H).

5k: White solid, m.p. 250-251 ^oC; IR (KBr): 3298, 2995, 1698, 1645, 1604, 1478, 1378, 1220, 1180 cm^{-1}.^1H NMR (400 MHz, $CDCl_3$, ppm) δ: 7.26 (d, *J*=8.4 Hz, 2H, Ar–H), 7.19 (d, *J*=8.4 Hz, 2H, Ar–H), 6.01 (br s, 1H, NH), 5.09 (s, 1H), 4.08 (q, *J*=7.2 Hz, 2H), 2.41 (s, 3H), 2.33-2.48 (m, 7H), 1.94-2.05 (m, 2H), 1.23 (t, *J*=7.2 Hz, 3H).

5l: Pale yellow solid, m.p. 230-232 ^oC; IR (KBr): 3295, 2995, 1695, 1643, 1604, 1400, 1377, 1220, 1175 cm^{-1}.^1H NMR (400 MHz, $CDCl_3$, ppm) δ: 7.34 (d, *J*=8.4 Hz, 2H), 7.19 (d, *J*=8.4 Hz, 2H), 6.29 (br s, 1H, NH), 5.08 (s, 1H), 3.67 (s, 3H), 2.30-2.47 (m, 7H), 1.99-2.05 (m, 1H), 1.91-1.97 (m, 1H). ^{13}C NMR (100 MHz, $CDCl_3$,ppm) δ: 195.86, 167.70, 149.95, 146.03, 144.03, 131.11, 129.69, 119.90, 113.03, 105.23, 51.17, 36.99, 35.91, 27.41, 21.0, 19.46.

5m: Pale yellow solid, m.p. 210-215 ^oC; IR (KBr): 3295, 2970, 1695, 1604, 1480, 1378, 1220, 1179 cm^{-1}. ^1H NMR(400 MHz, $CDCl_3$, ppm) δ: 7.24 (d, *J*=8.8 Hz, 2H, Ar–H), 6.74-6.78 (m, 3H), 5.06 (s, 1H), 4.08 (q, *J*=7.2 Hz, 2H), 3.75 (s, 3H), 2.29-2.42 (m, 7H) , 1.91-2.02 (m, 2H), 1.24 (t, *J*=7.2 Hz, 3H).

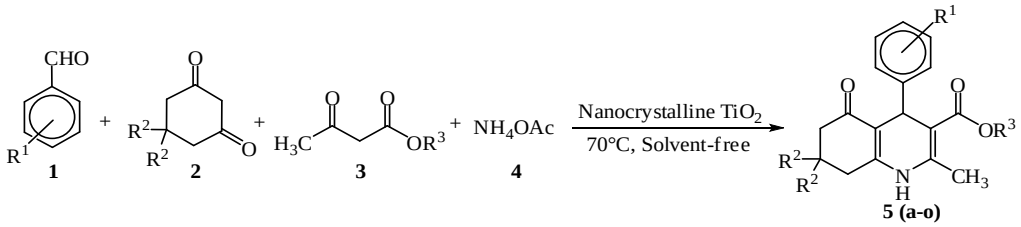

Fig. 1 Synthesis of polyhydroquinoline derivatives.

5n: Yellow solid, m.p. 239-240 ^{o}C; IR (KBr): 3295, 2995, 1695, 1643, 1603, 1477, 1375, 1218, 1180 cm^{-1}. ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ: 7.23 (d, *J*=0.8 Hz,1H), 6.23 (t, *J*=2.8 Hz, 1H,), 6.14 (br s, 1H, NH), 6.01(d, *J*=2.8 Hz, 1H), 5.31 (s, 1H), 4.11-4.21 (m, 2H), 2.36-2.51 (m,7H), 2.02-2.06 (m, 2H), 1.27 (t, *J*=8.8 Hz, 3H).

5o: Yellow solid, m.p. 252-253 ^{o}C; IR (KBr): 3298, 2995, 1680, 1648, 1600, 1478, 1378, 1221, 1178 cm^{-1}. ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ: 7.18 (d, *J*=8.8 Hz, 2H, Ar–H), 6.62 (d, *J*=8.8 Hz, 2H, Ar–H), 6.45 (br s, 1H, NH), 5.02 (s, 1H), 4.09 (q, *J*=7.2 Hz, 2H), 2.89 (s, 6H), 2.35-2.42 (m, 7H), 1.98 (m, 2H), 1.24 (t, *J*=7.2Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$,ppm) δ: 196.08, 167.77, 149.83, 149.02, 143.05, 135.95, 128.60, 113.63, 112.36, 106.38, 59.77, 40.78, 37.14, 35.17, 27.37, 21.11, 19.34, 14.30.

3. Results and Discussion

In continuation of our investigation on the use of new catalysts for chemical transformations [34] and our interest in the use of nanocrystalline TiO_2 as a solid acid catalyst [29], herein, we wish to report the one-pot condensation of 1,3-dicarbonyl compounds with various aldehydes and methyl or ethyl acetoacetate and NH_4OAc in the presence of catalytic amounts of TiO_2, as an inexpensive and recyclable catalyst (Fig. 1). In order to optimize the reaction conditions, the reaction of 4-chlorobenzaldehyde, 1,3-cyclohexanedione, ethylacetoacetate and ammonium acetate was studied with stirring at 70°C in a few drops of ethanol. After 3 hours, only 50% of product was realized after recrystallization of the crude product from ethanol. To improve the product yield and to optimize the reaction conditions, nano TiO_2 was used in catalytic amount (5 mg) and a reaction was carried out under similar conditions. A significant improvement in the yield of the product (87%) was observed. With this optimistic result in hand, we further investigated for the best

Table 1. Nanocrystalline TiO_2 catalyzed the synthesis of polyhydroquinoline derivatives 5(a-o).

Products [a]	R^1	R^2	R^3	Time(min)	Yield [b] (%)	Mp (°C)	
						Found	Lit [ref]
5a	H	Me	Et	35	90	224-226	203-204 [19]
5b	4-Cl	Me	Et	40	89	243-245	245-246 [19]
5c	4-Br	Me	Et	45	85	254-256	252-253 [19]
5d	3-NO_2	Me	Et	45	82	174-176	178-179 [19]
5e	4-NO_2	Me	Et	50	83	241-243	243-244 [19]
5f	4-OMe	Me	Et	40	86	255-257	256-257 [19]
5g	4-iPr	Me	Et	35	90	179-181	180-182 [26]
5h	2-Furyl	Me	Et	25	84	245-247	247-248 [19]
5i	4-Cl	Me	Me	45	88	254-256	245-246 [19]
5j	4-$N(CH_3)_2$	Me	Me	30	90	256-258	257-258 [19]
5k	4-Cl	H	Et	45	87	250-251	234-235 [20]
5l	4-Br	H	Me	50	84	230-232	—
5m	4-OMe	H	Et	45	85	212-215	193-195 [20]
5n	2-Furyl	H	Et	30	83	239-240	210- 212 [20]
5o	4-$N(CH_3)_2$	H	Et	35	89	252-253	—

[a.] All products were characterized from their spectroscopic (IR, ^{1}HNMR and ^{13}CNMR) data comparision with authentic samples,
[b.] Isolated yield.

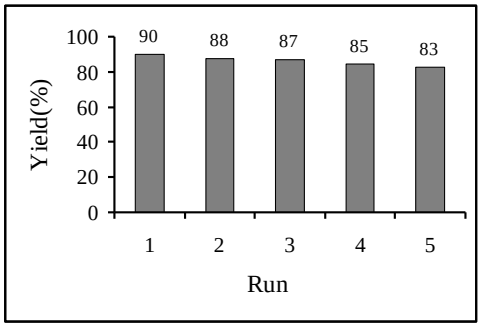

Fig 2. Recyclability of TiO_2 for the synthesis of 5a.

reaction conditions. In this connection, we investigated the reaction outcome using different amounts of TiO_2. An increase in the quantity of TiO_2 from 30 to 50 mg decreased the reaction time from 100 min to 40 min, with same yield (87 %). More increasing at the amount of TiO_2 not affected on the time and yield of the reaction. After optimization of the reaction conditions various aromatic and heterocyclic aldehydes were subjected to preparation of polyhydroquinolines under the selected conditions. The results are summarized in Table 1. We investigated 1,3-cyclohexanedione and dimedone as diketone compounds for the synthesis of polyhydroquinolines. Aromatic aldehydes containing electron withdrawing and electron donating substituents reacted with ethyl or methyl acetoacetate and ammonium acetate in the presence of 5 mg of nanocrystalline TiO_2 to afford the products in good to high yields (Table 1). It is noteworthy to mention that, the effect of the nature of the substituents on the aromatic ring showed no obvious effect on this conversion, because they were obtained in high yields in relatively short reaction times. In this study, the catalyst was recovered and reused in another run. The catalyst was recovered by a simple filtration and washed with ethanol and reused during four consecutive runs without any apparent loss of activity for the same reaction (Fig. 2). The possible mechanism for the formation of polyhydroquinolines in the presence of nano TiO_2 as a promoter is shown in Fig. 3. On the basis of this mechanism, TiO_2 catalyzes the reaction by the electrophilic activation of aldehyde for coupling with active methylene compounds and also it catalyses the Michel type addition of intermediates I, II and III, IV to give the product. To illustrate the efficiency of the present method, Table 2 compares our results in the preparation of polyhydroquinolines with the same results reported by the relevant reagents in the literature.

3. Conclusion

In conclusion, we have developed a facile and efficient method for the synthesis of a variety of polyhydroquinoline derivatives via an improved Hantzsch reaction catalyzed by nano TiO_2. The reaction conditions are mild and the reaction gives excellent yields of the products. This method does not involve the use of toxic solvents thus it is an environmentally friendly process.

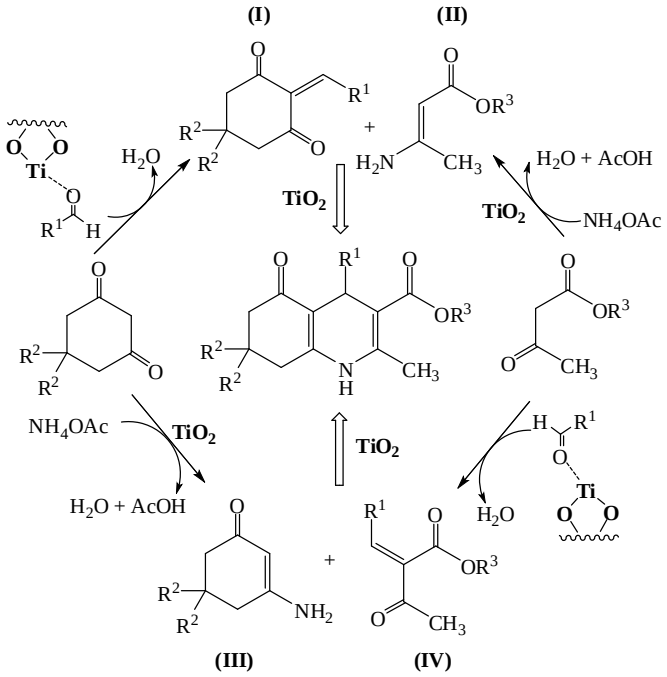

Fig 3. The plausible mechanism of the reaction.

Table 2. Comparison of some of the results obtained by the present method (I) with those reported using $K_7[PW_{11}CoO_{40}]$ (II), [15] I_2 (III), [16] ZnO-nanoparticles (IV), [20] $Sc(OTf)_3$ (V) [21] and HY-Zeolite (VI) [22].

	Time (h)/ Yield (%)/ Loading of catalyst (mg)					
Product	**I**	**II**	**III**	**IV**	**V**	**VI**
5a	0.58/90/5	0.58/80/30	1.5/93/76 (25°C)	0.33/ 98/5	4/93/77	2/93/100
5b	0.66/89/5	0.5/85/30	2.5/92/76 (25°C)	0.33/98/5	—	2.5/92/100
5e	0.8/83/5	0.5/80/30	—	0.25/89/5	—	2.5/88/100
5h	0.42/84/5	—	—	0.33/ 91/5	2/91/77	2.25/90/100

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