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Benign synthesis of *N*-aryl-3,10-dihydroacridin-1(2H)-one derivatives *via* ZnO nanoparticle-catalyzed Knoevenagel condensation/intramolecular enamination reaction

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Abstract

An efficient construction of 2-(*N*-arylamino)benzaldehydes and *N*-aryl-3,10-dihydroacridin-1(2H)-one derivatives starting from 2-hydroxybenzaldehydes has been developed. The synthesis of *N*-aryl-3,10-dihydroacridin-1(2H)-ones is based on the Knoevenagel condensation of dimedone to various 2-(*N*-arylamino)benzaldehydes, followed by an intramolecular enamination in the presence of 20 mol% of nanocrystalline ZnO. Moderate to high yields, operation simplicity, and cheap starting materials are the key features of the present method. The structures of the products were confirmed by ^{1}H and ^{13}C NMR spectroscopy and mass spectrometry (EI). Probable mechanisms for the present reactions to account for the formation of 2-(*N*-arylamino)benzaldehydes **3a-h** and *N*-aryl-3,10-dihydroacridin-1(2H)-one derivatives **4a-h** are also reported.

Keywords: *N*-aryl-3,10-dihydroacridin-1(2H)-ones; 2-(*N*-arylamino)benzaldehydes; Knoevenagel condensation; intramolecular enamination; ZnO nanoparticles; Smiles rearrangement.

Introduction

1,2-Dihydroquinoline derivatives are versatile and valuable building blocks and intermediates in the synthesis of biologically and pharmaceutically active compounds and natural products [1-4]. These compounds are found in a large number of biologically active compounds, which exhibit a wide range of activity such as antidiabetic [5], antimalarial [6], anti-inflammatory [7], antithyroid [8], HMG-CoA reductase inhibitors [9], and lipid peroxidation inhibitors [10]. Furthermore, a number of title compounds serve as a precursor to heterocyclic systems and natural products [11-15]. On the other hand, dihydroacridines as a class of dihydroquinolines have been found as potential chemicals in organic synthesis [16-18]. To the best of our knowledge, no attention has been given to the synthesis of *N*-aryl-3,10-dihydroacridin-1(2H)-ones. Thus, it is desirable to design an efficient and convenient method to access such heterocyclic molecules. Recently, literature has highlighted the importance of nanosized materials in several scientific and technological areas, and many research organizations have intensified investments in nanotechnology for the coming years [19]. In metal oxides, surface atoms

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make a distinct contribution to its catalyst activity. Nanocrystalline zinc oxide is certainly one of the most interesting of metal oxides, because it has special surface properties, which suggest that a very rich organic chemistry may occur on ZnO surface [20–22]. High yield, selectivity, and recyclability have been reported for a variety of ZnO nanocatalyst-based organic reactions [23–30].

During our studies on the ZnO nanoparticle-catalyzed *O*-acylation of alcohols [31] and synthesis of β-acetamidoketones and β-esters [32] *via* a multicomponent reaction, we became interested in the synthesis of highly substituted *N*-aryl-3,10-dihydroacridin-1(2H)-one derivatives using ZnO nanoparticles (nano-ZnO) as a catalyst. In our earlier work, we reported the synthesis of 2-(*N*-arylamino)benzaldehydes from 2-hydroxybenzaldehydes *via* Smiles rearrangement [33]. These fine compounds can be used as useful synthones in the substrates of *N*-aryl 1,2-dihydroquinoline derivatives. Herein, we wish to report an efficient reaction of Knoevenagel condensation/intramolecular enamination reaction of 2-(*N*-arylamino)benzaldehydes and dimedone in the presence of ZnO nanoparticles, which leads to *N*-aryl-3,10-dihydroacridin-1(2H)-one derivatives.

Experimental

The ^{1}H and ^{13}C NMR spectra were recorded on a Bruker Avance DRX-400 instrument in $CDCl_3$ at ambient temperature. The EI (70 eV) mass spectra were registered on a HP6890 GC-MS instrument with a mass-selective detector HP5793, column HP-5 (30 m × 0.25 mm × 0.2 μm). XRD patterns were obtained on a Siemens D5000 X-ray diffractometer using graphite-monochromatized high-intensity CuKα radiation (λ = 1.5406 Å). TEM and SEM images were obtained on a JEOL JEM-2010 and Leica Stereo Scan 3360 instruments, respectively. Microwave heating was conducted in an Ethos MR apparatus (2.45 GHz, maximum power 1000 W). Centrifugation was carried out in a HettichMikro 200 centrifuge. All chemicals required for the synthesis of 2-(*N*-arylamino) benzaldehydes **3** and *N*-aryl-3,10-dihydroacridin-1(2H)-ones **5** were purchased from Sigma-Aldrich (St. Louis, MO, USA), Fluka (Neu-Ulm, Germany) and Merck (Darmstadt, Germany) companies and were used as received.

Preparation of KF/Al_2O_3

The KF/Al_2O_3 catalyst was prepared according to the previously our reported procedure [34]. A mixture of potassium fluoride (45 g) and basic alumina (55 g, type T, Merck) in water (100 mL) was stirred at room temperature for 10 min. The resulting suspension was concentrated in vacuum and dried in a vacuum oven at 120 °C for 15 h.

Preparation of ZnO nanoparticles

The ZnO nanoparticles were prepared according to our previously reported method [31]. In a typical procedure, $Zn(OAc)_2.2H_2O$ (0.22 g, 1 mmol) was suspended in 2-PrOH (120 mL) under vigorous stirring at 50 °C. Sodium hydroxide alcoholic solution was prepared by adding NaOH (0.08 g, 2 mmol) to 2-PrOH (30 mL) under vigorous stirring at 50 °C. The flasks containing zinc acetate and sodium hydroxide solutions were cooled in an ice water bath. The sodium hydroxide solution was then added to zinc acetate solution under vigorous stirring to give a total volume of 150 mL. Final solution was heated in a controlled microwave cavity for 5 min. During the microwave irradiation, the temperature

of the solution reached 80 °C. After 5 min, a transparent solution was obtained. The centrifugation of the transparent solution yielded white products, which were washed twice with absolute EtOH and dried at 70°C for 4 h. The obtained white powder was calcined at 600 °C for 1 h.

Synthesis of 2-(*N*-arylamino)benzaldehydes 3a–h (General method)

An appropriate 2-hydroxybenzaldehyde **1** (1.0 mmol) and 2-chloro-*N*-arylacetamide **2** (1.0 mmol) were added to a stirred suspension of KF/Al_2O_3(150 mg) in DMF (5 mL), and the reaction mixture was stirred at 120 °C for 14 h with progress of the reaction being monitored by TLC. After completion of the reaction, the mixture was poured into ice-cold H_2O (10–12 g), stirred for 15 min, then extracted with CH_2Cl_2 (3 × 10 mL). The organic phase was dried with Na_2SO_4, filtered, and concentrated under vacuum. The residue was purified by preparative TLC (eluent petrol ether–EtOAc, 6:1).

Spectroscopic data*:* 2-(4-methylphenylamino)benzaldehyde(3)

^{1}H NMR (400 MHz, $CDC1_3$): δ = 9.98 (brs, 1H, NH), 9.93 (s, 1H, CHO), 7.58 (d, *J* = 8.80 Hz, 1H), 7.38 (t, *J* = 7.80 Hz, 1H), 7.17-7.23 (m, 5H), 6.84 (t, *J* = 7.40 Hz, 1H), 2.39 (s, 3H, CH_3) ppm. ^{13}C NMR (100 MHz, $CDC1_3$): δ = 194.18, 148.39, 136.93, 136.61, 135.55, 135.35, 130.06, 123.69, 119.15, 116.77, 112.81, 21.00 ppm. MS (EI): *m/z* (%) = 211 (M^+, 92), 182 (100), 167 (43). Anal Calcd for $C_{14}H_{13}NO$: C, 79.59; H, 6.20; N, 6.63. Found: C, 79.50; H, 6.09; N, 6.70.

General procedure for the synthesis of *N*-aryl-3,10-dihydroacridin-1(2H)-one derivatives 5a–h (General method)

Dimedone (**4**) (1.0 mmol) and an appropriate 2-(*N*-arylamino)benzaldehyde **3a–h** (1.0 mmol) were added to a stirred suspension of nano-ZnO (16 mg) in toluene (5 mL). The reaction mixture was stirred at 110 °C for 15 h. The progress of the reaction was monitored by TLC. After completion of the reaction, the catalyst was recovered by centrifugation. The solvent was removed under reduced pressure, and the residue was separated by column chromatography on silica gel (eluent petrol ether–EtOAc, 5:1).

Representative spectroscopic data

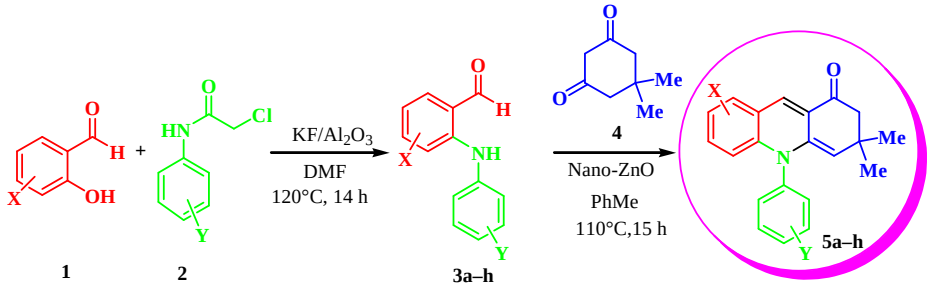
2,3-Dihydro-3,3-dimethyl-10-o-tolylacridin-1(10H)-one (**5c**): ^{1}H NMR (400 MHz, $CDC1_3$): δ = 7.27-6.90 (m, 8H), 6.04 (s, 1H), 5.05 (s, 1H), 2.19 (s, 3H), 2.00 (s, 2H), 1.26 (s, 6H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 196.91, 139.55, 139.33, 138.78, 138.72, 137.17, 131.02, 130.29, 129.74, 129.02, 128.68, 126.74, 123.88, 123.76, 115.15, 109.29, 107.38, 50.95, 32.38, 29.24, 17.47 ppm; MS (EI): *m/z* (%) = 315 (M^+, 14), 300 (100), 285 (2), 284 (3), 270 (4), 256 (6); Anal. Calcd for $C_{22}H_{21}NO$: C, 83.78; H, 6.71; N, 4.44. Found: C, 83.84; H, 6.76; N, 4.39. 7-Bromo-2,3-dihydro-3,3-dimethyl-10-p-tolylacridin-1(10H)-one (**5d**): ^{1}H NMR (400 MHz, $CDC1_3$): δ = 7.71-6.96 (m, 7H), 6.04 (s, 1H), 4.95 (s, 1H), 2.47 (s, 3H), 2.41 (s, 2H), 1.03 (s, 6H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 197, 140.09, 139.04, 136.54, 131.64, 130.84, 130.06, 129.60, 129.24, 129.03, 128.82, 120.83, 117.41, 116.10, 106.42, 50.84, 31.92, 29.50, 26.31 ppm; MS (EI): *m/z* (%) = 395 (12), 393 (M^+, 12), 381 (21), 380 (98), 379 (24), 378 (100); 299 (5) Anal. Calcd for $C_{22}H_{20}BrNO$: C, 67.01; H, 5.11; N, 3.55. Found: C, 67.09; H, 5.18; N, 3.62.

Results and discussion

ZnO nanoparticles were prepared from $Zn(OAc)_2.2H_2O$ according to the previously reported procedure [31] and

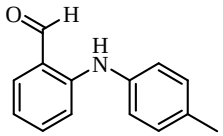
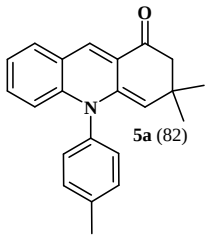
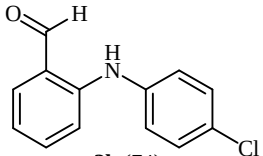
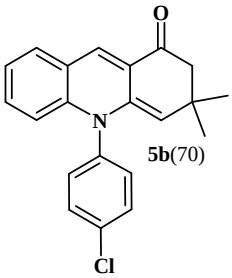
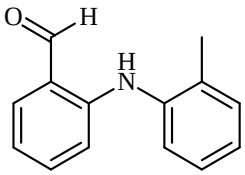
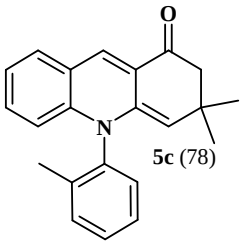
were characterized by X-ray diffraction (XRD), transmission electron microscopy (TEM), and scanning electron microscopy (SEM) (Figures 1S, 2S and 3S of supporting information). The mean particle size of nano-ZnO was shown to be 30 nm.

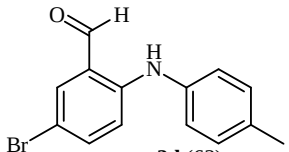
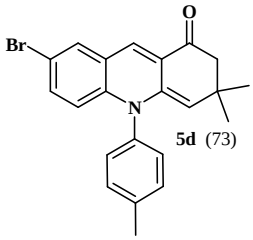
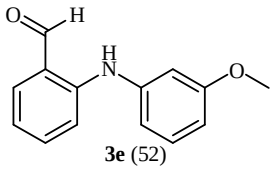
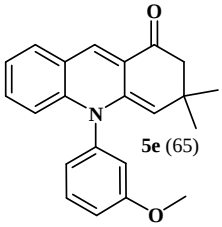
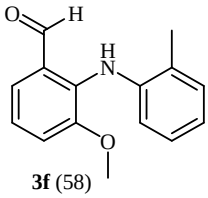
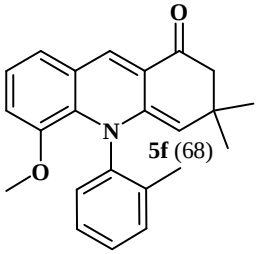
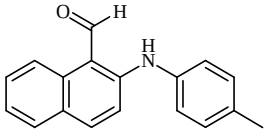
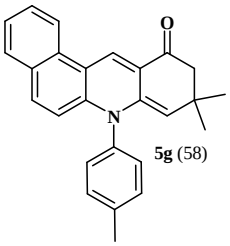
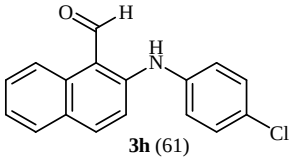
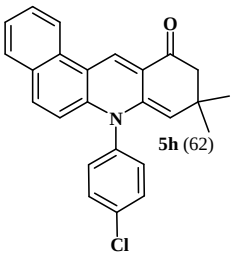
2-(*N*-arylamino)benzaldehydes **3a–h** were synthesized with moderate to good yields (43–74%) by reacting readily available 2-hydroxybenzaldehyde derivatives **1** with *N*-aryl 2-chloroacetamides **2** in DMF at 120 °C for 14 h using KF/Al_2O_3 as an efficient catalyst (Table 1). The ^{1}H and ^{13}C NMR spectra of compounds **3a–h** clearly indicated the formation of 2-arylaminobenzaldehydes **3a-h**.



Scheme 1.

Table 1. Synthesis of 2-(*N*-arylamino) benzaldehydes **3a-h** and *N*-aryl-3,10-dihydroacridin-1(2*H*)-one derivatives **5a-h**

Entry	2-(*N*-arylamino)benzaldehydes3a–h (Yield, %)	*N*-aryl-3,10-dihydroacridin-1(2*H*)-ones5a–h (Yield, %)*
1	 **3a** (72)	 **5a** (82)
2	 **3b** (74)	 **5b**(70)
3	 **3c** (55)	 **5c** (78)

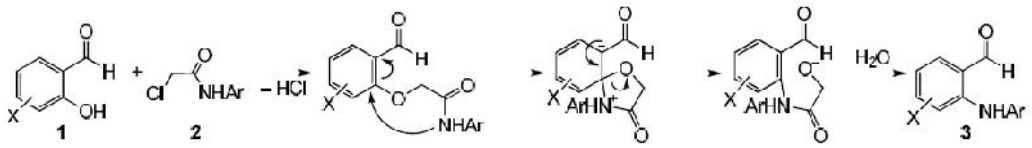
4	 **3d** (63)	 **5d** (73)
5	 **3e** (52)	 **5e** (65)
6	 **3f** (58)	 **5f** (68)
7	 **3g** (60)	 **5g** (58)
8	 **3h** (61)	 **5h** (62)

* Isolated yields in respect to compounds **3a–h**

The ^{1}H NMR spectrum of 2-(4-methylphenylamino)benzaldehyde **3a** (Entry 1, Table 1) contained a broad resonance at 9.98 ppm assigned to the NH, a sharp singlet for the aldehyde proton at 9.93 ppm and a singlet for the methyl protons at 2.39 ppm. Intramolecular hydrogen bonding between the amine proton and the carbaldehyde oxygen atom results in the deshielding of the NH proton. The ^{1}H-decoupled ^{13}C NMR spectrum of compound **3a** showed 12 distinct resonances in agreement with the proposed structure, with the aldehyde carbon signal appearing at 194.2 ppm, 10 distinct resonances for the aromatic carbons located between 112.8 and 148.4 ppm and a resonance at 21.0 ppm assigned to the methyl group. The electron ionization mass spectrum of compound **3a** clearly showed the

presence of the molecular ion (*m/z* 211) together with the base peak at *m/z* 182 attributed to the elimination of the formyl moiety. We used a series of 2-hydroxybenzaldehydes and 2-hydroxynaphthaldehydes **1** and *N*-aryl-2-chloroacetamides **2** to obtain a series of diverse 2-(*N*-arylamino)benzaldehydes and 2-(*N*-arylamino)naphthaldehydes **3** (Table 1). A possible reaction mechanism to account for the formation of the diarylamino linkage is proposed in Scheme 2 based on the Smiles rearrangement, in which the oxygen atom at the benzene ring was replaced with a nitrogen atom [35].

For the synthesis of *N*-aryl-3,10-dihydroacridin-1(2H)-one derivatives **5a–h**, we focused on systematic evaluation of different catalysts for the model reaction of 2-(4-methylphenylamino)benzaldehyde (**3a**) with dimedone (**4**). We have applied a wide range of catalysts including nano-CuO, KF/Al_2O_3, KF/nano-Al_2O_3, nano-α-Fe_2O_3 [36], $ZrOCl_2.8H_2O$ [37], HCl(1N) [38], *p*-TsOH, bulk ZnO, and nano-ZnO to improve the yield for the synthesis of 3,3-dimethyl-10-(4-methylphenyl)-3,10-dihydroacridin-1(2H)-one (**5a**) (Table 2). The reaction did not take place without any catalyst (Table 2, Entry 1).



Scheme 2.

The most interesting result was obtained with nano-ZnO as the catalyst. Commercially available ZnO was also evaluated for the synthesis of the title compounds. Using ZnO nanoparticles as a catalyst, the reaction time was reduced and the yield was increased in comparison with the use of bulk ZnO (Table 2, Entries 9 and 17).The higher catalytic activity of nano-ZnO over bulk ZnO may be attributed to the higher surface area, thus resulting in higher surface concentration of the catalytically active centers. The same reaction was carried out in various organic solvents in order to optimize the reaction conditions using nano-ZnO as catalyst (Table 2, Entries 10 and 11).

The highest yield was obtained with toluene as solvent. At room temperature, the desired product was not observed. However the product was observed in good overall yield when the reaction mixture was kept under reflux conditions in any of the solvents. While evaluating the necessary amount of catalyst, the best yield was observed in the presence of 20 mol% nano-ZnO with short reaction time. Using more than 20 mol% of the catalyst had no significant effect on the yield and time of the reaction. Therefore, 20 mol% of ZnO nanoparticles kept for 15 h in refluxing toluene was found to be the best conditions for this reaction (Table 2, Entry 17). Under the optimized conditions, the reaction of dimedone (**4**) with various 2-(*N*-arylamino)benzaldehydes **3a–h** proceeded smoothly to afford of diversely substituted *N*-aryl-3,10-dihydroacridin-1(2H)-one derivatives **5a–h**. All the substrates consistently furnished the desired products in moderate to high yields (Table 1). The nano-ZnO can be recovered and reused without any significant loss of activity.

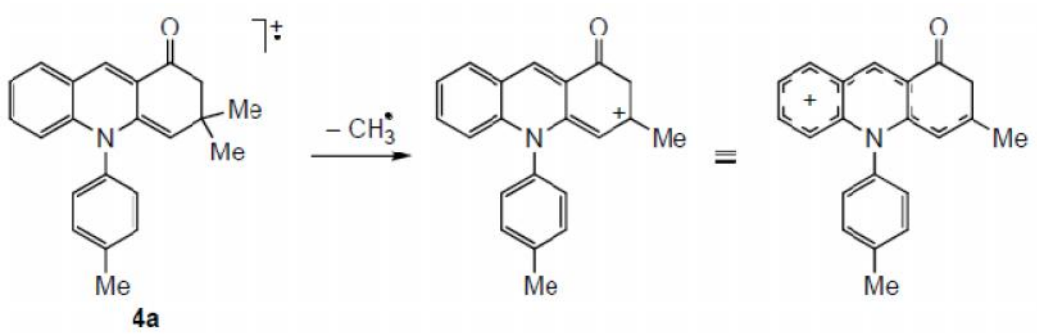
The structures of the products **5a–h** were confirmed by ^{1}H and ^{13}C NMR spectroscopy and mass spectrometry. For example, in the ^{1}H NMR spectrum of compound **5a**, the characteristic signals were a singlet at 0.87 ppm assigned to the protons of the dimedone ring methyl groups, a singlet of the methylene protons at 2.40 ppm, a singlet at 2.47 ppm for the methyl group of aromatic ring, and two singlets at 5.03 and 6.18 ppm for methine protons.The ^{13}C NMR spectrum of compound **5a** showed 19 distinct resonances in agreement with the proposed structure. The mass spectrum of compound **5a** clearly showed the presence of the molecular ion with moderate relative abundance and other expected fragments. Direct elimination of methyl moiety radical from molecular radical-ion yielded a fragment with *m/z* 300 as the base peak of the spectrum. This fragment as a carbocation could be stabilized by aromatic system (Scheme 3). A probable mechanism for the present reaction to account for the formation *N*-aryl-3,10-dihydroacridin-1(2H)-one derivatives **5** is shown in Scheme 4. In the first step, oxygen atom of the aldehyde group and aniline nitrogen atom of 2-(*N*-arylamino)benzaldehyde **3** are coordinated to active centers on the ZnO nanoparticle surface, and the carbonyl group is thus activated for Knoevenagel condensation with dimedone (**4**) to furnish the intermediate **6** with the release of H_2O [39]. Subsequently, the ring closure proceeds through an intramolecular enamination to give the desired tricyclic acridine ring system of compounds **5**.

Table 2. Optimization of the nano-ZnO-catalyzed synthesis of 3,3-dimethyl-10-(4-methylphenyl)-3,10-dihydroacridin-1(2*H*)-one (**5a**)*

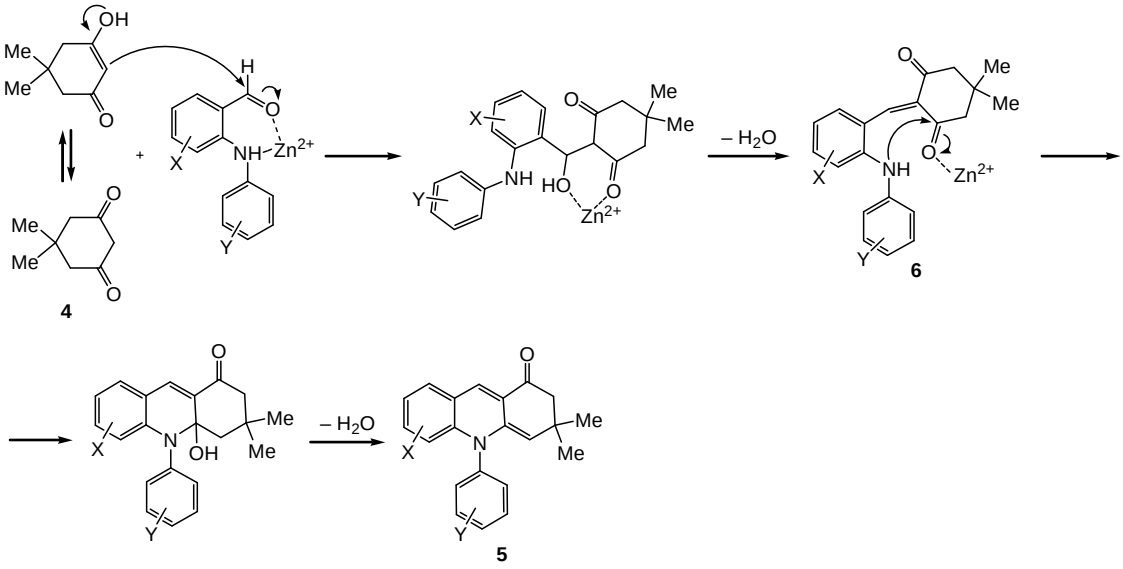
Entry	Catalyst	Solvent	Temperature, °C	Reaction time (h)	Yield, %
1	no catalyst	PhMe	110	15	0
2	nano-CuO	PhMe	110	15	23
3	KF/Al_2O_3	PhMe	110	15	trace
4	KF/nano Al_2O_3	PhMe	110	15	trace
5	nano-α-Fe_2O_3	PhMe	110	15	trace
6	$ZrOCl_2 \cdot 8H_2O$	PhMe	110	15	10
7	1N HCl	PhMe	110	15	trace
8	*p*-TsOH	PhMe	110	15	trace
9	bulk ZnO	PhMe	110	15	58
10	nano-ZnO	MeCN	80	15	56
11	nano-ZnO	EtOH	78	15	20
12	nano-ZnO	PhMe	80	15	65
13	nano-ZnO(0.05 mmol)	PhMe	110	15	54
14	nano-ZnO (0.1 mmol)	PhMe	110	15	60

15	nano-ZnO (0.4 mmol)	PhMe	110	15	85
16	nano-ZnO	PhMe	110	10	73
17	nano-ZnO	PhMe	110	15	82

*Reaction conditions: solvent (5 ml), 2-(4-methylphenylamino)benzaldehyde (**3a**) (1.0 mmol), dimedone (**4**) (1.0 mmol) and catalyst (0.2 mmol).



Scheme 3.



Scheme 4.

Conclusion

In summary, we have described a simple and efficient procedure for the synthesis of 2-(*N*-arylamino) benzaldehydes and *N*-aryl-3,10-dihydroacridin-1(2H)-one derivatives in moderate to high yields. Straightforward and easy work-up protocol and use of readily available

reagents are advantages of present method.

Supplementary Information

XRD, SEM and TEM of ZnO nanoparticles and copies of ^{1}H NMR, ^{13}C NMR, EI-MS spectra of **3** and **5** can be found *via* the "Supplementary Content" section of this article's webpage.

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