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ZnO nanoparticles: an efficient and reusable catalyst for one-pot synthesis of 1,8-dioxo-octahydroxanthenes

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Abstract Zinc oxide (ZnO) nanoparticles have been used as an efficient and recyclable catalyst for one-pot synthesis of 1,8-dioxo-octahydroxanthene derivatives via condensation of aromatic aldehydes and 5,5-dimethyl-1,3- cyclohexanedione under mild reaction conditions in good to excellent yields. ZnO nanoparticles were easily recycled from the reaction mixture and were reused three times without any loss in activity.

Keywords ZnO nanoparticles · 1,8-Dioxo-octahydroxanthenes · Aromatic aldehyde · 5,5-Dimethyl-1,3- cyclohexanedione · Solid acid · Reusable catalyst

Introduction

Xanthenes derivatives constitute an important class of heterocyclic compounds due to their various biological and pharmacological properties [1], such as antibacterial, [2] antiviral, [3] antiplasmodial [4], and anti-inflammatory [5] activities, while they are also being developed to act as new clinical agents in cancer therapy [6]. In addition, because of their useful spectroscopic properties, they have been widely used as dyes in laser technology [7], sensitizers in photodynamic therapy [8], and pH-sensitive fluorescent materials for visualization of biomolecules [9]. There are several methods for the synthesis of 1,8-dioxooctahydroxanthenes; these compounds conventionally prepared using condensation of aromatic aldehydes with

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5,5-dimethyl-1,3-cyclohexanedione (dimedone) in the presence of catalysts such as $InCl_3·4H2O$ [10], $FeCl_3·6H_2O$ [11], NH_2SO_3/SDS [12], $NaHSO_4/SiO_2$ [13], PPA·SiO_2 [14], ZnO–acetyl chloride [15], $BiCl_3$ [16], (L)-proline [17], trimethylsilyl chloride [18], amberlyst-15 [19], *p*-dodecylbenzene sulfonic acid [20], and tetrabutylammonium hydrogen sulfate [21].

Although, many of the reported methods are effective, but suffer from drawbacks such as the use of expensive or toxic catalysts, long reaction times, low yields, tedious work-up processes, and hazardous reaction conditions. Furthermore, the main disadvantage of some existing methods is that the catalysts are not recyclable and are destroyed during the work-up procedure. Therefore, the introduction of a mild, simple, efficient, and environmentally benign method to synthesize 1,8-dioxo-octahydroxanthene derivatives is still needed.

As reflected by the large number of publications in the past decade, nanomaterials have received much interest due to their potential application in many diverse areas, from basic research to various applications in electronics [22, 23], as biochemical sensors [24], in catalysis, and energy [25, 26]. Nanoparticles have emerged as sustainable alternatives to conventional materials, as robust high surface area heterogeneous catalysts and catalyst supports [27]. The nano-sized particles increase the exposed surface area of the active component of the catalyst. Therefore, nanoparticles can dramatically enhance the contact between reactants and catalyst and mimic the homogeneous catalysts [28]. Among several studies in this area, transition-metal oxide nanoparticles, specifically zinc oxide nanoparticles (ZnO NPs), have great potential as a professional catalysis in various organic transformations due to their high surface to volume ratio, easy recoverability, and their low cost [29–35].

In this work, we reveal zinc oxide (ZnO) nanoparticles as an efficient and reusable catalyst for one-pot multicomponent synthesis of 1,8-dioxo-octahydroxanthene derivatives via the condensation reaction of various aromatic aldehydes with dimedone in refluxing ethanol as a solvent (Scheme 1).

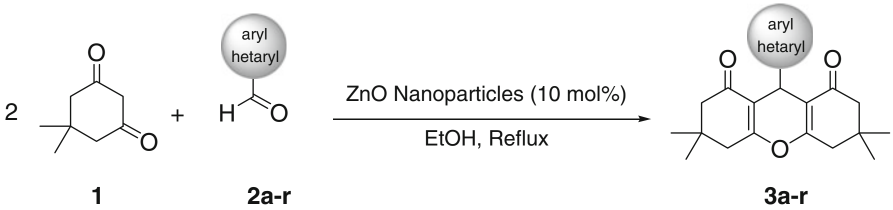
Experimental

Chemicals and materials

Melting points were determined with an Electrothermal 9100 apparatus. ^{1}H NMR spectra were recorded on Bruker Avance 400 MHz spectrometer, using deuterated chloroform ($CDCl_3$) as solvent. XRD patterns were recorded by a CBC MMA Instrument. Transmission electron microscopy (TEM) image was obtained on a Philips CM10 microscope. Materials were purchased from Fluka and Merck.

Preparation of ZnO nanocrystals

Aqueous solutions of $Zn^{2+}$and urea (molar ratio of Zn^{2+} to urea: 1/4) were placed in a flask under vigorous stirring (300 rpm). In order to inhibit the growth of the ZnO crystallite during the course of precipitation, a certain amount of surfactant SDS (0.001 M) was added into the reaction system. Then, the reaction system was heated



Scheme 1 ZnO nanoparticles-catalyzed synthesis of 1,8-dioxo-octahydroxanthenes

to 95 °C and stirred for 2 h. The precipitates were then filtered, washed with distilled water and ethanol four times, dried in air at 80 °C, and calcined at 230 °C for 2 h to prepare the ZnO nanoparticles whose mean particle size was 15 nm.

Procedure for preparation of 1,8-dioxo-octahydroxanthenes **3a–r**

To a solution of aromatic aldehyde (1 mmol) and dimedone (2 mmol), in EtOH (10 mL), ZnO nanoparticles (10 mol %) were added. Then, the mixture was stirred in reflux conditions for the appropriate time (Table 2). The progress of the reaction was monitored by TLC. After completion of the reaction, the mixture was filtered to remove the catalyst and the solvent was concentrated in a rotary evaporator. The crude product was purified by recrystallization from EtOH to obtain the pure compound. All the products were known compounds and characterized by comparing melting point and ^{1}H NMR spectra with those reported in the literature.

Spectral data of selected compounds

*3,4,6,7-Tetrahydro-3,3,6,6-tetramethyl-9-phenyl-2H-xanthene-1,8(5H,9H)-dione (**3a**)*

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.28 (m, 2H), 7.24 (m, 2H), 7.13 (m, 1H), 4.80 (s, 1H), 2.51 (s, 4H), 2.20 (q, J = 16. 1 Hz, 4H), 1.15 (s, 6H), 1.03 (s, 6H).

*9-(3-Chlorophenyl)-3,4,6,7-tetrahydro-3,3,6,6-tetramethyl-2H-xanthene-1,8(5H,9H)-dione (**3d**)*

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.27 (s, 1H), 7.19 (t, J = 7.8 Hz, 2H), 7.11 (d, J = 7.7 Hz, 1H), 4.77 (s, 1H), 2.53 (s, 4H), 2.25 (q, J = 16.0 Hz, 4H), 1.14 (s, 6H), 1.05 (s, 6H).

*3,4,6,7-Tetrahydro-3,3,6,6-tetramethyl-9-(3-nitrophenyl)-2H-xanthene-1,8(5H,9H)-dione (**3i**)*

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.09 (s, 1H), 8.01 (d, J = 8.2 Hz, 1H), 7.84 (d, J = 7.7 Hz, 1H), 7.43 (t, J = 7.9 Hz, 1H), 4.88 (s, 1H), 2.55 (s, 4H), 2.29 (q, J = 16.3 Hz, 4H), 1.16 (s, 6H), 1.03 (s, 6H).

*3,4,6,7-Tetrahydro-3,3,6,6-tetramethyl-9-(4-nitrophenyl)-2H-xanthene-1,8(5H,9H)-dione (**3j**)*

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.13 (d, *J* = 8.1 Hz, 2H), 7.51 (d, *J* = 8.1 Hz, 2H), 4.86 (s, 1H), 2.54 (s, 4H), 2.24 (q, *J* = 16.2 Hz, 4H), 1.16 (s, 6H), 1.02 (s, 6H).

*3,4,6,7-Tetrahydro-3,3,6,6-tetramethyl-9-p-tolyl-2H-xanthene-1,8(5H,9H)-dione (**3k**)*

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.20 (d, *J* = 8.0 Hz, 2H), 7.04 (d, *J* = 8.0 Hz, 2H), 4.76 (s, 1H), 2.52 (s, 4H), 2.25 (s, 3H), 2.21 (q, *J* = 16.3 Hz, 4H), 1.11 (s, 6H), 1.03 (s, 6H).

*3,4,6,7-Tetrahydro-9-(4-hydroxy-3-methoxyphenyl)-3,3,6,6-tetramethyl-2H-xanthene-1,8(5H,9H)-dione (**3p**)*

^{1}H NMR (400 MHz, $CDCl_3$): δ = 6.98 (m, 1H), 6.73 (m, 1H), 6.67 (m, 1H), 5.56 (s, 1H), 4.69 (s, 1H), 3.90 (s, 3H), 2.47 (s, 4H), 2.21 (q, *J* = 16.3 Hz, 4H), 1.13 (s, 6H), 1.03 (s, 6H).

*3,4,6,7-Tetrahydro-3,3,6,6-tetramethyl-9-(thiophen-2-yl)-2H-xanthene-1,8(5H,9H)-dione (**3q**)*

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.84–8.00 (m, 2H), 6.81 (m, 1H), 4.94 (s, 1H), 2.56 (s, 4H), 2.28 (q, *J* = 16.0 Hz, 4H), 1.16 (s, 6H), 1.03 (s, 6H).

Results and discussion

Initially, ZnO nanoparticles were prepared by uniform precipitation using urea hydrolysis [36]. The ZnO precursor was slowly deposited from aqueous solution. An anionic surfactant, sodium dodecyl sulfonate (SDS), was added into the solution to block ZnO crystal growth and its agglomeration. Then, ZnO nanoparticles were synthesized by the calcination of the precursor at high temperature. X-ray diffraction (XRD) studies (Fig. 1) confirmed that the synthesized materials were ZnO with a hexagonal wurtzite structure, and all the diffraction peaks agreed with the reported JCPDS data [37]. The results obtained from the XRD pattern and TEM micrograph of the ZnO nanoparticles (Figs. 1, 2) show that the mean particle size was 15 nm.

To optimize the reaction conditions, the reaction between benzaldehyde and dimedone was chosen as a model reaction. When benzaldehyde was treated with dimedone in the presence of ZnO nanoparticles (10 mol%) in ethanol under reflux conditions, the corresponding xanthene (**3a**) was obtained in 94 % yield within 1.5 h. This reaction was carried out in different solvents such as CH_2Cl_2, THF, H_2O, MeCN, and under solvent-free condition, and the best results in terms of reaction time and yield of the desired product **3a**, was obtained when the reaction was

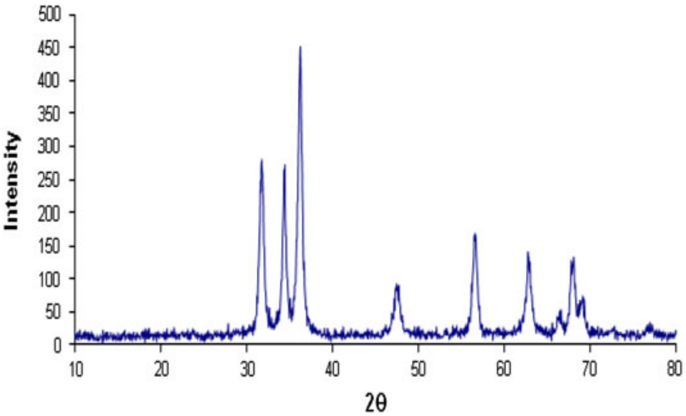
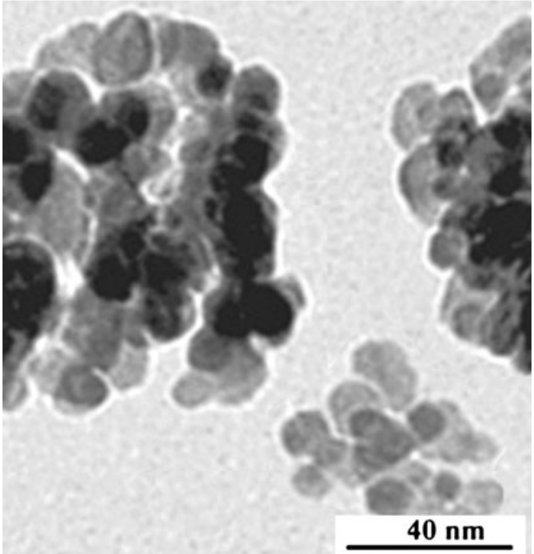


Fig. 1 XRD pattern of the ZnO nanoparticles

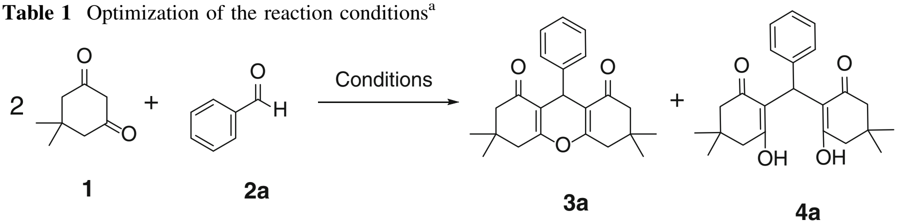
Fig. 2 The TEM image of ZnO nanoparticles



conducted in ethanol (Table 1, entries 1–6). Only 15 % of the desired product (**3a**) was obtained after heating the reaction mixture for 12 h in ethanol under reflux conditions in the absence of ZnO nanoparticles (Table 1, entry 12). ZnO bulk was also employed in the model reaction as a catalyst in ethanol, and product **3a** was formed in 60 % yield (Table 1, entry 13). Decreasing the catalyst loading from 10 to 2 mol% significantly lowered the yield of the reaction (Table 1, entries 6 and 9–11). The best catalyst loading was found in 10 mol%, which gave an excellent yield of **3a** after only 1.5 h. It is worth mentioning that the reaction temperature was optimized to be reflux in ethanol (Table 1, entries 6–8).

To show the generality of this procedure, several aromatic aldehydes were treated with dimedone in the presence of ZnO nanoparticles under optimized conditions

Table 1 Optimization of the reaction conditions[a]



Entry	Amount of catalyst (mol%)	Solvent	Temperature (°C)	Time (h)	Yield (%)[b]	
					3a	**4a**
1	Nano-ZnO (10)	MeCN	Reflux	3	65	–
2	Nano-ZnO (10)	THF	Reflux	3	57	–
3	Nano-ZnO (10)	CH_2Cl_2	Reflux	3	20	–
4	Nano-ZnO (10)	H_2O	Reflux	1.5	68	–
5	Nano-ZnO (10)	Neat	80	1.5	77	–
6	Nano-ZnO (10)	EtOH	Reflux	1.5	94	–
7	Nano-ZnO (10)	EtOH	60	1.5	70	–
8	Nano-ZnO (10)	EtOH	25	1.5	30	–
9	Nano-ZnO (8)	EtOH	Reflux	2	87	–
10	Nano-ZnO (5)	EtOH	Reflux	3	80	–
11	Nano-ZnO (2)	EtOH	Reflux	3	75	–
12	–	EtOH	Reflux	12	15	25
13	ZnO bulk (30)	EtOH	Reflux	6	60	–

[a] Reaction conditions: benzaldehyde (1 mmol) and dimedone (2 mmol) in solvent (10 mL)

[b] Isolated yields

(Table 2). The results indicate that the rate of reaction is dependent on the substituent effect. As a matter of fact, substrates having an electron-withdrawing at the aryl ring benzaldehyde give a higher yield of desired products (**3**) in shorter reaction times (Table 2, entries 2–10) compared with one having an electron-donating substituent (Table 2, entries 11–14). Aromatic aldehydes possessing two groups on the aryl ring such as 3,4-dimethoxybenzaldehyde and 4-hydroxy-3-methoxybenzaldehyde reacted with dimedone under reaction conditions to give the corresponding xanthenes in good yields (Table 2, entries 15–16).

Having these data in hand, we decided to apply this method for heterocyclic benzaldehydes. Thiophene-2-carbaldehyde and picolinaldehyde reacted with dimedone under optimum reaction conditions, and were found to produce the corresponding products **3** in somewhat lower yields than those with aromatic aldehydes (Table 2, entries 17–18).

An interesting feature of this method is that the catalyst can be recovered at the end of the reaction and can be used several times without losing its activity. To recover the catalyst after completion of the reaction, the mixture was filtered and the catalyst was washed with $CHCl_3$. After activating the catalyst at 100 °C, it was used

Table 2 Synthesis of different 9-aryl-1,8-dioxooctahydroxanthene derivatives **3** catalyzed by ZnO nanoparticle

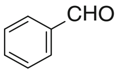
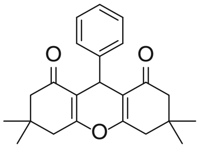
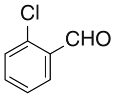
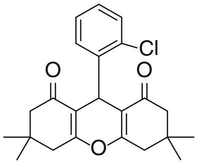
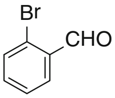
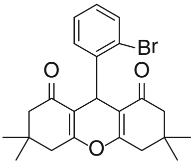
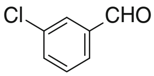
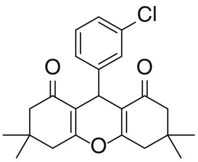
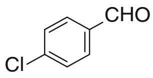
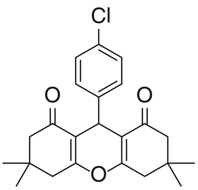
Entry	Aldehyde	Product[a]	Time (h)	Yield (%)[b]	M.p. (°C)	
					Found	Reported [Ref]
1	 **2a**	 **3a**	1.5	94	203–205	204–205 [20]
2	 **2b**	 **3b**	1.2	95	225–226	224–226 [20]
3	 **2c**	 **3c**	1.3	94	225–227	226–229 [10]
4	 **2d**	 **3d**	1	96	184–186	183–186 [20]
5	 **2e**	 **3e**	1	96	227–229	228–230 [20]

Table 2 continued

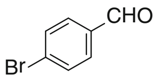
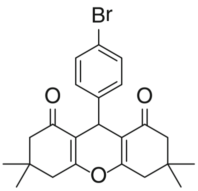
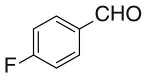
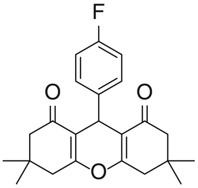
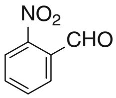
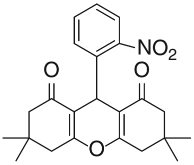
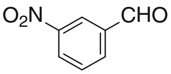
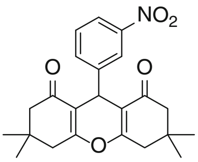
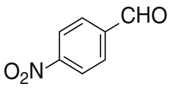
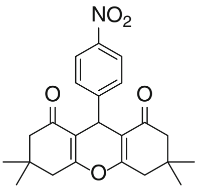
Entry	Aldehyde	Product[a]	Time (h)	Yield (%)[b]	M.p. (°C)	
					Found	Reported [Ref]
6	 **2f**	 **3f**	1	95	232–233	231–233 [10]
7	 **2g**	 **3g**	0.5	98	225–227	226–227 [10]
8	 **2h**	 **3h**	0.7	96	248–250	247–249 [20]
9	 **2i**	 **3i**	0.6	97	167–169	168–170 [20]
10	 **2j**	 **3j**	0.5	97	227–228	226–228 [20]

Table 2 continued

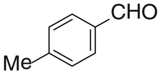
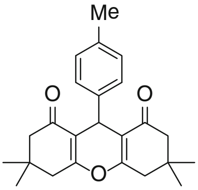
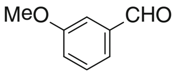
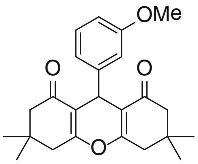
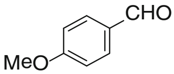
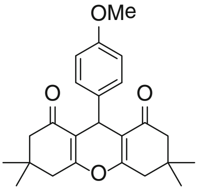
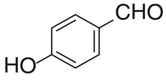
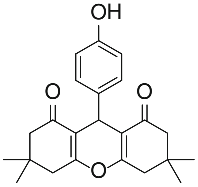
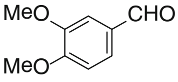
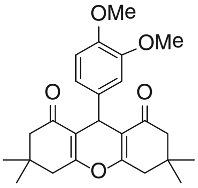
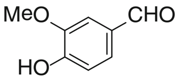
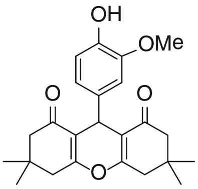
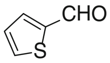
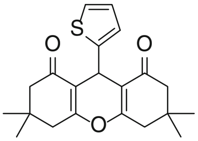
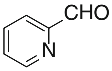
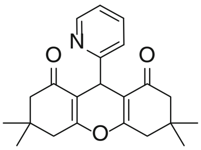
Entry	Aldehyde	Product[a]	Time (h)	Yield (%)[b]	M.p. (°C)	
					Found	Reported [Ref]
11	 **2k**	 **3k**	2	90	215–217	216–218 [15]
12	 **2l**	 **3l**	2.1	92	160–162	161–162 [16]
13	 **2m**	 **3m**	2.1	90	241–243	242–244 [20]
14	 **2n**	 **3n**	2.1	88	247–249	246–248 [20]
15	 **2o**	 **3o**	2.2	83	185–186	184–186 [15]

Table 2 continued

Entry	Aldehyde	Product[a]	Time (h)	Yield (%)[b]	M.p. (°C)	
					Found	Reported [Ref]
16	 **2p**	 **3p**	2.2	85	223–225	224–225 [15]
17	 **2q**	 **3q**	2.3	79	163–165	162–164 [16]
18	 **2r**	 **3r**	2.5	78	203–205	203–204 [16]

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[a] All the products were identified by comparing of melting point and ^{1}H NMR spectra with those of authentic samples reported in the literature

[b] Yields refer to pure isolated products

Table 3 Recycling studies of reaction between benzaldehyde with dimedone in the presence of ZnO nanoparticles to give product **3a**

Entry	Cycle	Time (h)	Yield (%)[a]
1	Fresh	1.5	94
2	1	1.5	92
3	2	1.5	89
4	3	1.5	87

[a] Yields refer to isolated products

for the reaction of benzaldehyde with dimedone. This process was repeated for three cycles and the yield of product **3a** did not significantly change (Table 3). These results clearly show the stability of ZnO nanoparticles in the reaction media as well its recovery and recycling without any appreciable decrease in its activity.

In order to show the merits and drawbacks of this catalyst, some of our results were compared with other catalysts reported in the literature (Table 4). As shown in Table 4, the condensation of benzaldehyde with dimedone catalyzed by ZnO

Table 4 Comparison of the efficiency of ZnO nanoparticles with reported catalysts for the synthesis of **3a**

Entry	Catalyst	Conditions	Time (h)	Yield (%) [Ref]
1	Amberlyst-15	CH_3CN/reflux	5	92 [19]
2	DBSA	H_2O/reflux	6	89 [20]
3	TMSCl	CH_3CN-DMF/reflux	8	84 [18]
4	Dowex-50 W	Neat/100 °C	3	87 [38]
5	$InCl_3{\cdot}4H_2O$	[bmim][BF4]/80 °C	4	87 [10]
6	$InCl_3$	Neat/100 °C	0.6	83 [39]
7	P_2O_5	Neat/100 °C	0.7	80 [39]
8	(L)-proline	DCE//60 °C	6	83 [17]
9	ZnO-CH_3COCl	CH_3CN/reflux	5	86 [15]
10	Silica sulfuric acid	Neat/80 °C	1.5	82 [40]
11	PPA.SiO_2	Neat/140 °C	0.5	93 [14]
12	$NaHSO_4.SiO_2$	CH_3CN/reflux	6.5	90 [13]
13	H_2SO_4	H_2O/80 °C	3	90 [41]
14	[Hmim]TFA	Neat/80 °C	3	85 [42]
15	$BiCl_3$	Neat/80 °C	3	90 [16]
16	ZnO nanoparticles	EtOH/reflux	1.5	94

nanoparticles in EtOH offer the corresponding product **3a** very fast in excellent yield in mild conditions, while other catalysts need harder conditions and longer reaction times for synthesis of xanthene **3a**.

Conclusion

In summary, we have reported an efficient procedure for one-pot synthesis of 1,8-dioxo-octahydroxanthenes using ZnO nanoparticles as a solid acid, reusable, and a non-toxic nanocatalyst. The main advantages of this procedure include high yields, simple experimental procedure, short reaction times, use of various substrates, ease of the work-up, non-use of hazardous organic solvents, and ease of recovering and reusing of the catalyst, which make it a useful, attractive, and green strategy for the preparation of xanthene derivatives.

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References

1. M.M.M. Pinto, M.E. Sousa, M.S.J. Nascimento, Curr. Med. Chem. **12**, 2517–2538 (2005)
2. R.W. Lambert, J.A. Martin, J.H. Merrett, K.E.B. Parkes, G.J. Thomas, PCT Int. Appl. WO 9706178 (1997); Chem. Abstr. **126**, p212377y (1997)
3. H. Takeshiba, T. Jiyoujima, Japan Kokai Tokkyo Koho JP 56005480 (1981)
4. F. Zelefack, D. Guilet, N. Fabre, C. Bayet, S. Chevalley, S. Ngouela, B.N. Lenta, A. Valentin, E. Tsamo, M.G. Dijoux-Franca, J. Nat. Prod. **72**, 954–957 (2009)

5. J.P. Poupelin, G. Saint-Rut, O. Fussard-Blanpin, G. Narcisse, G. Uchida-Ernouf, R. Lakroix, Eur. J. Med. Chem. **13**, 67–71 (1978)
6. R. Giri, J.R. Goodell, C. Xing, A. Benoit, H. Kaur, H. Hiasa, D.M. Ferguson, Bioorg. Med. Chem. **18**, 1456–1463 (2010)
7. M. Ahmad, T.A. King, D.K. Ko, B.H. Cha, J. Lee, J. Phys. D Appl. Phys. **35**, 1473–1476 (2002)
8. R.M. Ion, A. Planner, K. Wiktorowicz, D. Frackowiak, Acta Biochim. Pol. **45**, 833–845 (1998)
9. C.G. Knight, T. Stephens, Biochem. J. **258**, 683–689 (1989)
10. X. Fan, X. Hu, X. Zhang, J. Wang, Can. J. Chem. **83**, 16–20 (2005)
11. X.S. Fan, Y.Z. Li, X.Y. Zhang, X.Y. Hu, J.J. Wang, Chin. Chem. Lett. **16**, 897–899 (2005)
12. T.S. Jin, J.S. Zhang, A.Q. Wang, F.S. Zhang, Chin. J. Org. Chem. **25**, 335–338 (2005)
13. B. Das, P. Thirupathi, K.R. Reddy, B. Ravikanth, L. Nagarapu, Catal. Commun. **8**, 535–538 (2007)
14. S. Kantevari, R. Bantu, L. Nagarapu, J. Mol. Catal. A: Chem. **269**, 53–57 (2007)
15. M.T. Maghsoodlou, S.M. Habibi-Khorassani, Z. Shahkarami, N. Maleki, M. Rostamizadeh, Chin. Chem. Lett. **21**, 686–689 (2010)
16. J.J. Li, X.Y. Tao, Z.H. Zhang, Phosphorus Sulfur Silicon Relat. Elem. **183**, 1672–1678 (2008)
17. B. Das, J. Kashanna, R.A. Kumar, P. Jangili, Synth. Commun. **42**, 2876–2884 (2012)
18. S. Kantevari, R. Bantu, L. Nagarapu, Arkivoc **xvi**, 136–148 (2006)
19. B. Das, P. Thirupathi, I. Mahender, V.S. Reddy, Y.K. Rao, J. Mol. Catal. A: Chem. **247**, 233–239 (2006)
20. T.S. Jin, J.S. Zhang, J.C. Xiao, A.Q. Wang, T.S. Li, Synlett **5**, 866–870 (2004)
21. H.N. Karade, M. Sathe, M.P. Kaushik, Arkivoc **xiii**, 252–258 (2007)
22. M.S. Gudiksen, L.J. Lauhon, J. Wang, D.C. Smith, C.M. Lieber, Nature **415**, 617–620 (2002)
23. D.V. Talapin, J.S. Lee, M.V. Kovalenko, E.V. Shevchenko, Chem. Rev. **110**, 389–458 (2010)
24. G. Liu, Y. Lin, V. Ostatná, J. Wang, Chem. Commun. **27**, 3481–3483 (2005)
25. Y. Cai, H. Ke, J. Dong, Q. Wei, J. Lin, Y. Zhao, L. Song, Y. Hu, F. Huang, W. Gao, H. Fong, Appl. Energy **88**, 2106–2112 (2011)
26. H. Tong, S. Ouyang, Y. Bi, N. Umezawa, M. Oshikiri, J. Ye, Adv. Mater. **24**, 229–251 (2012)
27. D. Astruc, F. Lu, J.R. Aranzaes, Angew. Chem. Int. Ed. **44**, 7852–7872 (2005)
28. V. Polshettiwar, R.S. Varma, Green Chem. **12**, 743–754 (2010)
29. M. Lakshmi Kantam, K.B.S. Kumar, C. Sridhar, Adv. Synth. Catal. **347**, 1212–1214 (2005)
30. M. Hosseini-Sarvari, H. Sharghi, S. Etemad, Helv. Chim. Acta **91**, 715–724 (2008)
31. Z. Mirjafary, H. Saeidian, A. Sadeghi, F.M. Moghaddam, Catal. Commun. **9**, 299–306 (2008)
32. M.Z. Kassaee, F. Movahedi, H. Masrouri, Synlett **8**, 1326–1330 (2009)
33. M. Kiamehr, F.M. Moghaddam, Tetrahedron Lett. **50**, 6723–6727 (2009)
34. R. Hekmatshoar, G.N. Kenary, S. Sadjadi, Y. Beheshtiha, Synth. Commun. **40**, 2007–2013 (2010)
35. S. Paul, P. Bhattacharyya, A.R. Das, Tetrahedron Lett. **52**, 4636–4641 (2011)
36. E. Tang, B. Tian, E. Zheng, C. Fu, G. Cheng, Chem. Eng. Commun. **195**, 479–491 (2008)
37. JCPDS Card No. 36-1451
38. G.I. Shakibaei, P. Mirzaei, A. Bazgir, Appl. Catal. A **325**, 188–192 (2007)
39. G.K. Verma, K. Raghuvanshi, R.K. Verma, P. Dwivedi, M.S. Singh, Tetrahedron **67**, 3698–3704 (2011)
40. F. Zhang, Y.X. Wang, F.L. Yang, H.Y. Zhang, Y.F. Zhao, Synth. Commun. **41**, 347 (2011)
41. R.H. Wang, B. Cui, W.T. Zhang, G.F. Han, Synth. Commun. **40**, 1867–1872 (2010)
42. M. Dabiri, M. Baghbanzadeh, E. Arzroomchilar, Catal. Commun. **9**, 939–942 (2008)