Solvent-Free Synthesis of 1,8-Dioxo-octahydroxanthenes and 14-Aryl-14H-dibenzo[*a,j*]xanthenes using Saccharin Sulfonic Acid as an Efficient and Green Catalyst

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Abstract: Saccharin sulfonic acid (SaSA) is utilized as a highly efficient, green and inexpensive sulfonic acid-containing catalyst for the following one-pot multi-component organic transformations: (i) the condensation of dimedone (2 eq.) with aromatic aldehydes (1 eq.) leading to 1,8-dioxo-octahydroxanthenes, and (ii) the reaction of 2-naphthol (2 eq.) with arylaldehydes (1 eq.) leading to 14-aryl-14H-dibenzo[*a,j*]xanthenes. In these protocols, the title compounds are produced in high to excellent yields and in relatively short reaction times under solvent-free conditions.

Keywords: 1,8-Dioxo-octahydroxanthene, 14-Aryl-14H-dibenzo[*a,j*]xanthene, Saccharin sulfonic acid (SaSA), Sulfonic acid-containing catalyst, One-pot multi-component reaction, Solvent-free.

Introduction

Xanthene derivatives such as 1,8-dioxo-octahydroxanthenes and 14-aryl-14H-dibenzo[*a,j*]xanthenes are of importance as they have various industrial, pharmaceutical and biological applications¹⁻⁶. For example, these compounds have been applied as dyes in laser technology¹, and as pH sensitive fluorescent materials for visualization of biomolecules². Moreover, xanthenes derivatives have been used as antibacterial³, antiviral⁴, antitumor⁵ and anti-inflammatory⁶ agents. The usual method for the synthesis of 1,8-dioxo-octahydroxanthenes involves the one-pot multi-component reaction of dimedone (5,5-dimethyl-1,3-cyclohexanedione) (2 eq.) with aldehydes (1 eq.)⁷⁻¹⁶. Some catalysts have been applied for this reaction, such as silica chloride⁷, H₃PW₁₂O₄₀ supported MCM-41⁸, cyanuric chloride⁹,

1-butyl-3-methylimidazolium trifluoroacetate¹⁰, PPA-SiO₂¹¹, DABCO-bromine¹², TMSCl¹³, silica-supported H₁₄[NaP₅W₃₀O₁₁₀] nanoparticles¹⁴, SbCl₃/SiO₂¹⁵ and ZrO(OTf)₂¹⁶. The best synthetic route towards 14-aryl-14*H*-dibenzo[*a,j*]xanthenes is the one-pot multi-component condensation between 2-naphthol (2 eq.) and aldehydes (1 eq.) using some catalysts, *e. g.* ZrO(OTf)₂¹⁶, selectfluor^{TM} {1-(chloromethyl)-4-fluoro-1,4-diazoniabicyclo[2.2.2]octane bis(tetrafluoroborate)}¹⁷, Sc[N(SO₂C₈F₁₇)₂]₃¹⁸, nano-TiO₂¹⁹, Yb(OTf)₃²⁰, InCl₃²¹, HClO₄-SiO₂²², KAl(SO₄)₂·12H₂O²³, silica sulfuric acid²⁴ and TaCl₅²⁵. Nevertheless, most of the reported methods for the synthesis of the xanthenes derivatives are associated with one or more of the following drawbacks: (i) moderate yields, (ii) long reaction times, (iii) the use of large amount of catalyst, and (iv) the use of expensive, non-available or toxic catalysts, and (v) no agreement with the green chemistry protocols.

In recent years, the use of sulfonic acid-containing catalysts and reagents has gained considerable attention in organic synthesis, due to their unique properties such as enhanced reactivity as well as selectivity, efficiency, straightforward workup, easy availability of their starting materials, eco-friendly reaction conditions and ability to promote a wide range of reactions²⁶⁻³⁹. They are also non-toxic, non-corrosive and inexpensive. Saccharin sulfonic acid (SaSA) is certainly one of the most interesting SO₃H-containing catalysts which has been recently prepared, and used in some organic transformations including chemoselective trimethylsilylation of alcohols³⁶, acetylation of alcohols, phenols and amines³⁷, *N*-Boc protection of amines and formation of *tert*-butyl ethers from alcohols³⁸, and preparation as well as deprotection of 1,1-diacetates³⁹. The structure of saccharin sulfonic acid (SaSA) is shown in Figure 1. Nevertheless, in most of the existing processes in organic synthesis, the use of toxic and volatile organic solvents as reaction media is inevitable, and these are environmentally unacceptable from green chemistry view point. One of the most effective techniques to solve this problem is solvent-free conditions which makes synthesis simpler, saves energy and prevents solvent waste, hazards, and toxicity⁴⁰⁻⁴⁵. Consequently, it is noteworthy that the combination of safe catalysts with the use of solvent-free technique represents a suitable way toward the so-called “ideal synthesis”⁴⁰⁻⁴⁵.

Saccharin sulfonic acid (SaSA)

Figure 1

Multi-component reactions (MCRs) have drawn great interest enjoying an outstanding status in modern organic synthesis and medicinal chemistry because they are one-pot processes bringing together three or more components and show high atom economy and high selectivity⁴⁴⁻⁴⁷. Moreover, MCRs offer the advantage of simplicity and synthetic efficiency over conventional chemical reactions⁴⁴⁻⁴⁷.

Having the above subjects in mind, we report here a new, highly efficient and simple method for the one-pot multi-component synthesis of 1,8-dioxo-octahydroxanthenes from dimedone (2 eq.) and aromatic aldehydes (1 eq.), as well as 14-aryl-14*H*-dibenzo[*a,j*] xanthenes from 2-naphthol (2 eq.) and arylaldehydes (1 eq.) using SaSA as an inexpensive and green SO₃H-containing catalyst in the absence of solvent (Schemes 1 and 2). It is worth noting that this method has none of the above-mentioned drawbacks at all.

Scheme 1

Scheme 2

Experimental

All chemicals were purchased from Merck or Fluka Chemical Companies. All known compounds were identified by comparison of their melting points and spectral data with those reported in the literature. Progress of the reactions was monitored by TLC using silica gel SIL G/UV 254 plates. The ^1H NMR (500 or 300 MHz) and ^{13}C NMR (125 or 75 MHz) were run on a Bruker Avance DPX-250, FT-NMR spectrometer (δ in ppm). Melting points were recorded on a Büchi B-545 apparatus in open capillary tubes.

Procedure for the preparation of saccharin sulfonic acid (SaSA)

A flask (500 mL) charged with saccharin (17.1 g, 0.1 mol) was equipped with a constant pressure dropping funnel containing chlorosulfonic acid (11.65 g, 0.1 mol) and a gas outlet tube which was dipped into water to dissolve the generated HCl gas during the reaction. The flask was put into an ice bath and chlorosulfonic acid was added dropwise over a period of 10 min, and the resulting mixture was stirred slowly for another 10 min. The temperature of the mixture was brought up to the room temperature and was stirred for an additional 30 min. The mixture was triturated with *n*-hexane (10 mL) and then filtered. The solid residue was washed with *n*-hexane (10 mL) and dried under vacuum to give SaSA³⁶⁻³⁹.

General procedure for the synthesis of 1,8-dioxo-octahydroxanthene derivatives

To a mixture of dimedone (0.28 g, 2 mmol) and aldehyde (1 mmol) in a 10 mL round-bottomed flask connected to a reflux condenser, SaSA (0.04 g, 0.15 mmol) was added. The resulting mixture was stirred in an oil-bath (90 °C), and the reaction was monitored by TLC. After completion of the reaction, EtOAc (10 mL) was added to the reaction mixture, stirred for 2 min and filtered. The solvent of the filtrate was evaporated and the resulting solid (crude product) was recrystallized from EtOH (96%) to afford the pure product.

Selected spectral data of 1,8-dioxo-octahydroxanthenes

3,3,6,6-Tetramethyl-9-phenyl-1,8-dioxo-octahydroxanthene (1a)

^1H NMR (CDCl_3 , 500 MHz): δ 0.90 (s, 6H), 1.04 (s, 6H), 2.09 (d, J = 16.1 Hz, 2H), 2.27 (d, J = 16.2 Hz, 2H), 2.53 (d, J = 17.1 Hz, 2H), 2.58 (d, J = 17.7 Hz, 2H), 4.53 (s, 1H), 7.10 (t, J = 7.0 Hz, 1H), 7.18 (d, J = 7.0 Hz, 2H), 7.21 (t, J = 7.20 Hz, 2H); ^{13}C NMR (CDCl_3 , 125 MHz): δ 27.7, 29.6, 32.3, 32.6, 41.3, 51.2, 116.1, 126.8, 128.4, 128.8, 144.5, 162.7, 196.8.

3,3,6,6-Tetramethyl-9-(3-nitro-phenyl)-1,8-dioxo-octahydroxanthene (1f)

¹H NMR (CDCl₃, 500 MHz): δ 0.91 (s, 6H), 1.04 (s, 6H), 2.11 (d, *J* = 16.1 Hz, 2H), 2.29 (d, *J* = 16.1 Hz, 2H), 2.57 (d, *J* = 17.9 Hz, 2H), 2.60 (d, *J* = 17.9 Hz, 2H), 4.65 (s, 1H), 7.56 (t, *J* = 7.7 Hz, 1H), 7.66 (d, *J* = 7.6 Hz, 1H), 8.00 (d, 2H); ¹³C NMR (CDCl₃, 125 MHz): δ 27.2, 29.6, 32.0, 32.7, 50.9, 56.6, 60.7, 106.3, 115.1, 136.8, 140.7, 153.2, 163.9, 197.0.

3,3,6,6-Tetramethyl-9-(4-chloro-phenyl)-1,8-dioxo-octahydroxanthene (1i)

¹H NMR (CDCl₃, 500 MHz): δ 0.90 (s, 6H), 1.04 (s, 6H), 2.09 (d, *J* = 16.1 Hz, 2H), 2.27 (d, *J* = 16.1 Hz, 2H), 2.52 (d, 2H), 2.57 (d, *J* = 17.6 Hz, 2H), 4.51 (s, 1H), 7.19 (d, *J* = 8.3 Hz, 2H), 7.29 (d, *J* = 8.2 Hz, 2H); ¹³C NMR (CDCl₃, 125 MHz): δ 27.7, 29.7, 31.8, 32.6, 41.3, 51.1, 115.6, 128.6, 130.2, 132.4, 143.1, 162.8, 196.7.

General procedure for the synthesis of 14-aryl-14H-dibenzo[a,j]xanthene derivatives

A mixture of 2-naphthol (0.288 g, 2 mmol), aldehyde (1 mmol) and SaSA (0.04 g, 0.15 mmol) in a 10 mL round-bottomed flask connected to a reflux condenser, was stirred in an oil-bath (120 °C). After completion of the reaction, as monitored by TLC, EtOAc (10 mL) was added to the reaction mixture, stirred for 2 min and filtered. Then, the EtOAc from the filtrate was evaporated, and the resulting solid (crude product) was recrystallized from *iso*-propanol/CHCl₃ (4/1) to afford the pure product.

Selected spectral data of 14-aryl-14H-dibenzo[a,j]xanthenes**14-(3-Nitrophenyl)-14H-dibenzo[a,j]xanthene (2f)**

¹H NMR (DMSO-d₆, 300 MHz): δ 6.91 (s, 1H), 7.11-7.25 (m, 1H), 7.43-7.48 (m, 2H), 7.55-7.71 (m, 4H), 7.88-8.03 (m, 7H), 8.66 (d, *J* = 8.4 Hz, 2H); ¹³C NMR (DMSO-d₆, 75 MHz): δ 36.7, 116.6, 118.2, 123.6, 124.2, 125.2, 127.6, 129.1, 129.5, 130.0, 131.10, 131.19, 146.3, 148.4, 153.1.

14-(3-Bromophenyl)-14H-dibenzo[a,j]xanthenes (2h)

¹H NMR (DMSO-d₆, 500 MHz): δ 6.63 (s, 1H), 6.90-7.02 (m, 2H), 7.25 (d, *J* = 12.9 Hz, 2H), 7.37-7.49 (m, 5H), 7.56-7.61 (m, 2H), 7.85-7.89 (m, 4H), 8.52 (d, *J* = 14.0 Hz, 1H); ¹³C NMR (DMSO-d₆, 125 MHz): δ 34.8, 116.9, 118.2, 123.3, 124.9, 127.4, 128.5, 128.8, 129.1, 129.8, 130.2, 130.3, 130.9, 131.4, 132.0, 143.2, 148.6.

14-(4-Chlorophenyl)-14H-dibenzo[a,j]xanthene (2i)

¹H NMR (DMSO, 500 MHz): δ 6.76 (s, 1H), 7.18 (d, *J* = 11.4 Hz, 2H), 7.46-7.94 (m, 12H), 8.66 (d, *J* = 12.7 Hz, 2H); ¹³C NMR (DMSO-d₆, 125 MHz): δ 36.3, 117.4, 118.1, 123.4, 123.7, 125.0, 127.4, 128.8, 129.1, 129.6, 130.1, 131.1, 131.2, 144.8, 148.4.

Results and Discussion

As above mentioned, more recently, SaSA has been introduced as an efficient SO₃H-containing catalyst in organic synthesis³⁶⁻³⁹. In addition, xanthenes derivatives have various industrial and pharmaceutical applications¹⁻⁶, and the reported methods for the preparation of these compounds have several drawbacks. These subjects encouraged us to examine applicability of SaSA in the synthesis of two important types of xanthene derivatives including 1,8-dioxo-octahydroxanthenes and 14-aryl-14H-dibenzo[a,j]xanthenes, and introduced new efficient procedures for the preparation of these compounds. For this purpose, at first, the one-pot multi-component reaction of dimedone (2 mmol) with 4-methoxybenzaldehyde (1 mmol) was studied in the presence of different amounts of SaSA at range of 60-100 °C under solvent-free conditions to provide compound **1b**. The respective results are summarized in Table 1. As Table 1 indicates, 15 mol% of the catalyst was

sufficient to perform the reaction efficiently at 90 °C. Moreover, increment the amount of SaSA, the temperature and the reaction time did not improve the yield.

Table 1. The reaction of dimedone with 4-methoxybenzaldehyde using different molar ratios of SaSA at range of 60-100 °C.

Catalyst Amount, mol%	Time, min	Temperature, °C	Yield ^a , %
-	150	90	36
5	120	90	65
10	100	90	86
15	40	90	93
20	40	90	92
15	75	80	87
15	90	70	76
15	100	60	64
15	40	100	93

^aIsolated yield.

To recognize efficiency and scope of the method for the synthesis of 1,8-dioxo-octahydroxanthenes, dimedone was reacted with different aromatic aldehydes under the optimal reaction conditions (Table 2). As it is shown in Table 2, all aldehydes including benzaldehyde and aromatic aldehydes possessing electron-donating substituents, electron-withdrawing substituents or halogens on their aromatic rings afforded the desired 1,8-dioxo-octahydroxanthene derivatives in high to excellent yields and in short reaction times. Thus, the protocol was efficient and general.

Table 2. The synthesis of 1,8-dioxo-octahydroxanthenes from dimedone and aromatic aldehyde using SaSA.

Aldehyde	Product	Time, min	Yield ^a , %	M.p. °C (Lit.)
	 (1a)	25	94	199-201 (203-205) ¹⁴
	 (1b)	40	93	243-245 (248-250) ¹⁴
	 (1c)	45	90	216-218 (219-221) ¹⁴

	 (1d)	45	92	209-211 (208-210) ¹⁰
	 (1e)	10	97	225-227 (228-230) ¹⁴
	 (1f)	10	97	164-166 (165-166) ¹⁵
	 (1g)	10	94	252-254 (252-254) ¹⁰
	 (1h)	25	93	242-244 (240-242) ¹³
	 (1i)	20	97	229-231 (231-233) ¹⁴
	 (1j)	20	93	185-157 (183-185) ¹⁵

^aIsolated yield.

After the successful application of SaSA in the preparation of 1,8-dioxo-octahydroxanthenes, we checked its applicability to catalyze the preparation of 14-aryl-14*H*-dibenzo[*a,j*]xanthenes, by studying the multi-component condensation of 2-naphthol (2 mmol) with 4-methoxybenzaldehyde (1 mmol) using different molar ratios of SaSA at range of 90-130 °C in the absence of solvent to provide compound **2b**. The results are displayed in Table 3. As it can be seen in Table 3, higher yield and shorter reaction time were obtained when the reaction was carried out in the presence of 15 mol% of the catalyst at 120 °C.

Table 3. Effect of amount of SaSA and temperature on the condensation of 2-naphthol with 4-methoxybenzaldehyde.

Catalyst Amount, mol%	Time, min	Temperature, °C	Yield ^a , %
-	240	120	Trace
5	100	120	43
10	70	120	78
15	50	120	94
20	50	120	92
15	50	130	88
15	60	110	86
15	80	100	73
15	110	90	60

^aIsolated yield.

In another study, the efficiency and generality of the method were evaluated by the reaction of 2-naphthol with arylaldehydes bearing electron-donating substituents, electron-withdrawing substituents or halogens on their aromatic rings (Table 4). As Table 4 indicates, the method was general and efficient; all reactions were performed successfully to furnish the corresponding 14-aryl-14*H*-dibenzo[*a,j*]xanthenes in high to excellent yields and in relatively short reaction times.

Table 4. The SaSA-catalyzed condensation of 2-naphthol with arylaldehydes leading to 14-aryl-14*H*-dibenzo[*a,j*]xanthenes.

Aldehyde	Product	Time, min	Yield ^a , %	M.p. °C (Lit.)
	 (2a)	70	94	186-188 (184-185) ¹⁹
	 (2b)	50	94	201-203 (203-205) ²³
	 (2c)	80	95	226-228 (227-229) ¹⁹
	 (2d)	60	87	135-137 (133-134) ¹⁹

		100	96	312-314 (311-312) ¹⁷
		90	94	216-218 (213) ¹⁷
		90	91	296-298 (297-298) ²³
		80	89	186-188 (190-191) ¹⁹
		100	96	284-286 (289-290) ²³
		100	93	207-209 (209-211) ¹⁹

^aIsolated yield.

Conclusion

In summary, we have developed new protocols for the synthesis of 1,8-dioxo-octahydroxanthenes and 14-aryl-14*H*-dibenzo[*a,j*]xanthenes using SaSA under solvent-free conditions. The advantages of these methods are efficiency, generality, high yield, relatively short reaction time, cleaner reaction profile, simplicity, ease of preparation of the catalyst, ease of product isolation, and compliance with the green chemistry protocols.

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