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# Tamarind water catalyzed improved synthesis of 2,2′-arylmethylene bis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one) derivatives: A green approach

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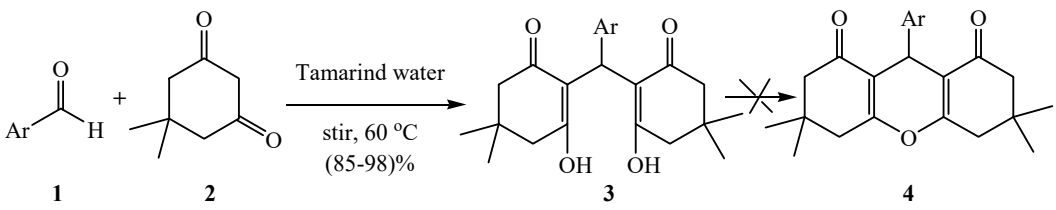
The reaction of 5,5-dimethyl-1,3-cyclohexanedione (dimedone) with various aromatic aldehydes catalyzed by tamarind water produces 2,2′-arylmethylenebis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one) derivatives in 85-98% yield under water bath heating at 60°C. Xanthene derivatives as cyclized products have not been obtained by this procedure. This method provides several advantages such as environmental friendliness, high yields and simple work-up procedure. The product is purified by simple filtration followed by crystallization with ethanol and drying processes.

**Keywords**: Tamarind water, catalysis, condensation reaction, green synthesis, 2,2′-arylmethylenebis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one)

2,2′-arylmethylenebis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one) compounds are biologically important, which can be evaluated as tyrosinase inhibitors[1]. They are also very useful synthetic intermediates and can serve as versatile precursors for the synthesis of various xanthenes and acridinedione derivatives that display biological and therapeutical function such as antiviral[2], antibacterial[3], antioxidant[4] and also action against disorders like asthma and inflammatory processes[5] besides they are also used in laser technology[6].

A good number of synthetic methods have been reported in the literature for the synthesis of 2,2′-arylmethylenebis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one) compounds which involve the usage of different types of catalysts such as $KF/Al_2O_3$ (Ref 7), urea[8], silica-diphenic acid[9], iodine[10], $HClO_4.SiO_2$ (Ref 11), zirconium oxychloride/ sodium amide[12], $FeCl_3.6H_2O$/TMSCl/[bmim][$BF_4$][13], cetyltrimethyl ammonium bromide (CTMAB)[14], sodium dodecyl sulphate (SDS)[15] and ethyldiamine diacetate (EDDA)[16]. However, in spite of their potential utility, some of the above mentioned methods suffer from one or more drawbacks such as long reaction times, poor yield of the products, harsh reaction conditioned and use of expensive and/or toxic catalysts/solvents. Due to the biological importance of 2,2′-arylmethylenebis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one) compounds, there is a strong demand for the development of mild and highly efficient method for their synthesis. In this regard, we have synthesized the title compounds by the reaction between various aromatic aldehydes and dimedone using tamarind water as natural acid catalysts (Scheme I).

Tamarind (*Tamarindus indica*) (Figure 1) has long been one of the most popular of the non-citrus tropical and subtropical fruits, largely because of its attractive flavour and refreshing sugar-acid balance. The composition of tamarind varies with geographical, cultural and seasonal harvesting and processing. It contains plant acid (16-18%) composed mainly of tartaric acid (up to *ca*. 18%) with minor amount of ascorbic acid. Other constituents include sugar (20-40%), flavonoids, polyphenolics, fat, vitamin, minerals (Ca, K, P, *etc*.) and tartarates[17,18]. Tamarind water is acidic in nature due to presence of tartaric acid mainly along with ascorbic acid, thus it may work as an acid catalyst for the condensation reaction between for aldehydes and dimedone. Since, there are no examples of the use of tamarind water as a catalyst for the preparation of 2,2′-arylmethylenebis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one), from the reaction between aldehydes and dimedone herein we report the use of tamarind water as a catalyst cum solvent for the synthesis of the title compounds.




Scheme I — Tamarind water promoted synthesis of 2,2′-arylmethylenebis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one)

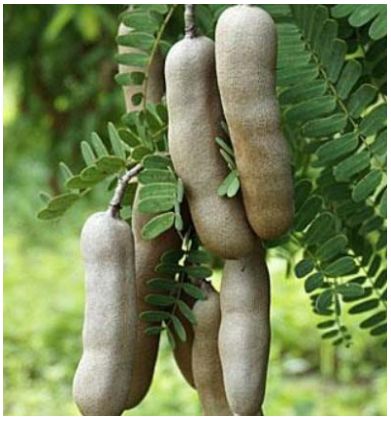


Figure 1 — Photograph of green tamarind fruit

## Results and Discussion

In continuation of our research work on the application of edible fruit juice: a cheap and ecofriendly material as catalyst for the development of new synthetic methodologies[19-25], here we are pleased to report a simple and facile synthesis of 2, 2′-arylmethylenebis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one) derivatives by treatment of various aromatic aldehydes (**1**) and 5,5-dimethyl-cyclohexane-1,3-dione (dimedone) (**2**) catalyzed by tamarind water under heating at 60ºC in water bath in 85-98% yields (Scheme I, Table I).

The reaction was optimized by varying amount of catalyst. We choose the reaction between benzaldehyde (**1a**) and dimedone (**2**) as the model reaction. From the Table II (entry 5), it was found that the best result was obtained by using 5 mL of tamarind water (*p*H=3.0) under reflux in water bath at 60ºC (conversion rate 98% of the product **3a**) over the room temperature (RT) stirring method (conversion rate 30% of the product **3a**) in short reaction time. Various 2,2′-arylmethylenebis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one) (**3a-o**) were synthesized with good to excellent yields (85-98%) using this methodology in shorter reaction time (2-3 h). We found that condensation of aromatic aldehydes and dimedone gave only 2,2′-arylmethylenebis (3-hydroxy-5,5-dimethyl-2-cyclo-hexene-1-one) **3** as lone product, even in 1:1 (aldehyde: dimedone) experiments. Xanthene as a cyclized product **4** was not obtained. A wide range of aromatic aldehydes

Table I — Synthesis of 2,2-arylmethylene bis(3-hydroxy-5, 5-dimethyl-2-cyclohexene-1-one)[a] catalyzed by tamarind water[b]

| Compd | Ar | Reaction time (h) | Yield[c] (%) | m.p. (ºC) Obsd. | m.p. (ºC) Lit. (Ref.) |
|---|---|---|---|---|---|
| **3a** | $C_6H_5$ | 2.5 | 90 | 190-192 | 189-190 (8) |
| **3b** | 4-$CH_3$-$C_6H_4$ | 3.0 | 95 | 142-144 | 142-143 (8) |
| **3c** | 4-$OCH_3$-$C_6H_4$ | 3.0 | 85 | 138-140 | 140-141 (8) |
| **3d** | 3,4-($OCH_2O$)-$C_6H_3$ | 3.0 | 85 | 184-186 | 187-189 (7) |
| **3e** | 4-OH-$C_6H_5$ | 3.0 | 85 | 200-202 | 201-203 (7) |
| **3f** | 4-OH-3-$CH_3O$-$C_6H_3$ | 3.0 | 90 | 194-196 | 194-195 (8) |
| **3g** | 2-$NO_2$-$C_6H_4$ | 2.0 | 98 | 188-190 | 188-189 (8) |
| **3h** | 3-$NO_2$-$C_6H_4$ | 2.0 | 92 | 192-194 | 190-191 (8) |
| **3i** | 4-$NO_2$-$C_6H_4$ | 2.0 | 98 | 194-196 | 195-196 (7) |
| **3j** | 3-Cl-$C_6H_4$ | 2.0 | 96 | 186-188 | 188-190 (7) |
| **3k** | 4-Cl-$C_6H_4$ | 2.0 | 93 | 146-148 | 145-147 (7) |
| **3l** | 3-Br-$C_6H_4$ | 2.0 | 98 | 190-192 | - |
| **3m** | 4-Br-$C_6H_4$ | 2.0 | 95 | 154-156 | 152-154 (9) |
| **3n** | Thiophene | 2.5 | 85 | 160-162 | - |
| **3o** | Furan | 2.5 | 85 | 140-142 | 141-143 (9) |

[a]All the products were characterized by $^1$H NMR, some of them by $^{13}$C NMR & Mass spectral data
[b]Tamarind water was taken 5 mL in each reaction having *p*H 3.0
[c]Isolated yields based upon starting material

Table II — Model reaction of dimedone (2 mmol) and benzaldehyde (1 mmol) using tamarind water as catalyst

| Entry | Amount of water (mL) | Amount of tamarind water[b] (mL) | Reaction time (h) | Conversion rate at RT (%) | Conversion rate[a] at 60°C (%) |
|---|---|---|---|---|---|
| 1 | 5 | 0 | 2 | 0 | 5 |
| 2 | 5 | 0 | 3.0 | 0 | 10 |
| 3 | 0 | 3 | 1.0 | 5 | 30 |
| 4 | 0 | 5 | 1.0 | 10 | 15 |
| 5 | 0 | 5 | 2.5 | 30 | 98 |
| 6 | 0 | 10 | 3.0 | 30 | 98 |

[a]% of conversion of the reaction was measured by TLC with respect to aldehyde
[b]*p*H of the tamarind water was 3.0

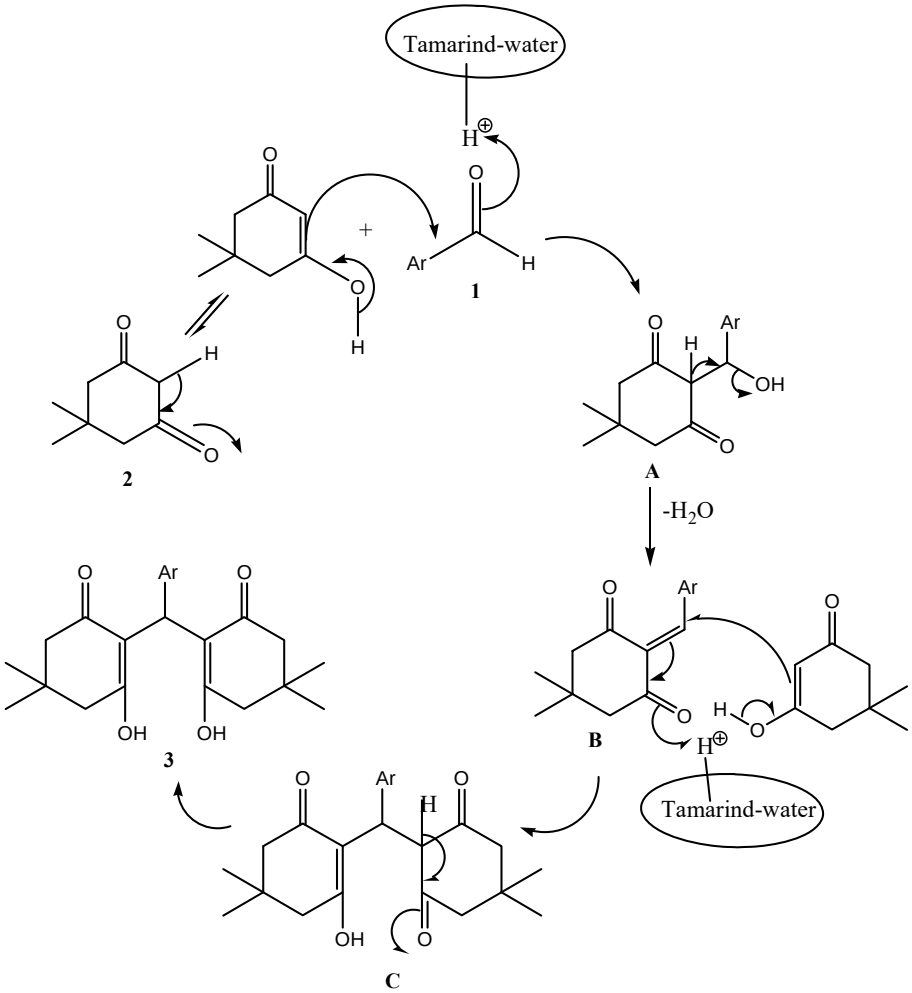
were subjected with dimedone to prove the general applicability of our present procedure which is summarized in Table I. The presence of electron-donating groups (such as methoxy, hydroxyl) and electron-withdrawing groups (such as nitro, halide) in the aromatic ring of the aldehydes did not affect the rate of conversion under the present experimental conditions.

$^{1}$H NMR spectra of compounds **3a**-**o** showed the resonated signals at the range of 1.06-1.26 ppm for –$CH_3$ aliphatic protons with singlet signal, resonated signals at 5.40-5.59 ppm singlet for CH proton and two enolic hydroxyl proton (C=CH-OH) resonated at two different positions in the range of 11.50 to 11.98 ppm.

$^{13}$C NMR spectra of the compound **3b** only recorded and showed mainly the resonated signals at δ 190.38 ppm for **C**=O, δ at 189.37 for =**C**-OH, in addition δ at 115.69 ppm for C=C, with a signal at δ 126-135 ppm for aromatic carbons and aliphatic carbons $CH_3$, $CH_2$, CH resonated at the range of at δ 20-47 ppm.

Mass spectra of compound **3f** and **3n** showed that a molecular ion peak with a base peak at *m/z* 415.2161 and at *m/z* 357.1631 respectively attributed to our target products.

A plausible mechanism for the reaction between aldehydes (1 mmol) and dimedone (2 mmol) in presence of tamarind water as a mild and efficient catalyst has been proposed (Scheme II). In the first step of the proposed mechanism, aldehyde molecule (**1**) was protonated and activated by the catalyst and undergoes Knoevenagel condensation with dimedone (**2**) to form intermediate product **A,** followed by dehydration and form another intermediate product **B** which was further activated by the catalyst, so that it undergo Michael reaction with another molecule of dimedone to form final desired product **3**.




Scheme II — Plausible mechanism for the tamarind water catalyzed synthesis of 2,2′-arylmethylene bis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one)

## Experimental Section

All reactions were run in dried glassware. Reagents were purchased from Spectrochem or SRL or Sigma-Aldrich and were used without further purification. Melting points were determined on a Kofler block and uncorrected. $^1$H NMR spectrum was recorded in $CDCl_3$ on Bruker AV-300 (300 MHz) and Bruker AV-400 (400 MHz) spectrometers using TMS as an internal standard. Mass spectra were acquired on a QTOF Micro Mass spectrometer. Analytical samples were dried *in vacuo* at RT. Thin layer chromatography was carried out on silica gel G for TLC made of SRL Pvt. Ltd.

## Preparation of tamarind water

The green tamarind fruit were purchased from the local market. The upper shell of the green tamarind and its inner grain were removed. The hard green material (50 g) was boiled with water (100 mL), cooled and it was centrifuged using micro centrifuge (REMI RM-12C). The clear portion of the aqueous extract of the tamarind generally called "tamarind water" (*p*H=3.0) was used as catalyst for the reactions.

## General procedure for the synthesis of 2, 2′-arylmethylenebis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one) derivative

In a 50 mL round-bottom flax, aromatic aldehyde (1 mmol) and 5,5-dimethyl-1,3-cyclohexanedione (dimedone) (2mmol) were stirred in the presence of tamarind water (5 mL, *p*H=3.0) for 5 min at RT in a water bath; the temperature was then raised to 60ºC and maintained for an appropriate time (monitored by TLC). After completion of the reaction, the mixture was diluted with water (5 mL), stirred for 5 min, and the resulting solid product was collected by simple filtration. The crude product was then purified by recrystallization from ethanol. All the products were characterized by $^1$H NMR spectral data and comparison with the reported melting points and some of them by $^{13}$C NMR and mass spectral data. Compounds **3a-k**, **3m** and **3o** are known however, compounds **3l** and **3n** are not known in the literature.

## Spectral data of representative compounds

**2,2′-Phenylmethylenebis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one), 3a**: White crystal; m.p. 190-192ºC; $^1$H NMR (300 MHz, $CDCl_3$) ppm: 1.10 (s, 6H, $CH_3$), 1.23 (s, 6H, $CH_3$), 2.28-2.53 (m, 8H, $CH_2$), 5.54 (s, 1H, CH), 7.09 (d, *J* = 7.5 Hz, 2H, Ph-H), 7.16 (t, *J* = 6.9 Hz, 1H, Ph-H), 7.25 (d, *J* = 7.5 Hz, 2H, Ph-H), 11.52 (br. s, 1H, OH), 11.91 (s, 1H, OH).

**2,2′-(4-Methylphenyl)methylene bis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one), 3b**: White crystal, m.p. 142-144ºC; $^1$H NMR (300 MHz, $CDCl_3$) ppm: 1.09 (s, 6H, $CH_3$), 1.22 (s, 6H,$CH_3$), 2.29 (s, 3H, $CH_3$), 2.33-2.53 (m, 8H, $CH_2$), 5.49 (s, 1H, CH), 6.97 (d, *J* = 7.8 Hz, 2H, Ph-H), 7.07 (d, *J* = 8.1 Hz, 2H, Ph-H), 11.58 (br. s, 1H, OH), 11.91 (s, 1H, OH); $^{13}$C NMR (75 MHz, $CDCl_3$): δ 20.87 ($CH_3$), 27.34 ($2CH_3$), 29.65 ($2CH_3$), 31.38 ($2CMe_2$), 32.36 (CH), 46.40 ($2CH_2$), 47.01 ($2CH_2$), 115.69 (2C=C), 126.62 (2C), 128.92 (2C), 134.85, 135.24, 189.37 (2C=C-OH), 190.38 (2C=O).

**2,2′-(4-Methoxyphenyl)methylene bis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one), 3c**: White crystal, m.p. 138-140ºC; $^1$H NMR (300 MHz, $CDCl_3$) ppm: 1.09 (s, 6H, $CH_3$), 1.22 (s, 6H,$CH_3$), 2.27-2.48 (m, 8H, $CH_2$), 3.77 (s, 3H, $OCH_3$), 5.48 (s, 1H, CH), 6.81 (d, *J* = 9.0 Hz, 2H, Ph-H), 7.00 (d, *J* = 8.1 Hz, 2H, Ph-H), 11.57 (br. s, 1H, OH), 11.91 (s, 1H, OH).

**2,2′-(3,4-Methylenedioxyphenyl)methylene bis (3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one), 3d**: White crystal, m.p. 184-186ºC; $^1$H NMR (300 MHz, $CDCl_3$) ppm: 1.09 (s, 6H, $CH_3$), 1.21 (s, 6H,$CH_3$), 2.17-2.48 (m, 8H, $CH_2$), 5.45 (s, 1H, CH), 5.91 (s, 2H, -$OCH_2$O-), 6.54 (d, *J* = 9.0 Hz, 1H, Ph-H), 6.56 (s, 1H, Ph-H), 6.70 (d, *J* = 8.1 Hz, 1H, Ph-H), 11.54 (br. s, 1H, OH), 11.94 (s, 1H, OH).

**2,2′-(4-Hydroxyphenyl)methylene bis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one), 3e**: White crystal, m.p. 200-202ºC; $^1$H NMR (300 MHz, $CDCl_3$) ppm: 1.09 (s, 6H, $CH_3$), 1.21 (s, 6H,$CH_3$), 2.33-2.42 (m, 8H, $CH_2$), 2.53 (s, 1H, OH), 5.47 (s, 1H, CH), 6.72 (d, *J* = 8.7 Hz, 2H, Ph-H), 6.93 (d, *J* = 8.1 Hz, 2H, Ph-H), 11.52 (br. s, 1H, OH), 11.89 (s, 1H, OH).

**2,2′-(4-Hydroxy-3-methoxyphenyl)methylene bis (3-hydroxy-5,5-dimethyl-2-cyclohex-ene-1-one), 3f**: White crystal, m.p. 194-196ºC; $^1$H NMR (300 MHz, $CDCl_3$) ppm: 1.11 (s, 6H, $CH_3$), 1.23 (s, 6H, $CH_3$), 2.28-2.48 (m, 8H, $CH_2$), 3.76 (s, 3H, $OCH_3$), 5.49 (s, 2H, CH & OH), 6.58 (d, *J* = 8.4 Hz, 1H, Ph-H), 6.61 (s, 1H, Ph-H), 6.81 (d, *J* = 8.4 Hz, 1H, Ph-H), 11.60 (br. s, 1H, OH), 11.98 (s, 1H, OH); QTOF MS ($ES^+$): *m/z* Calcd for $C_{24}H_{31}O_6$ $[M+H]^+$ 415.2115, Found $[M+H]^+$ 415.2161.

**2,2′-(2-Nitrophenyl)methylene bis(3-hydroxy-5, 5-dimethyl-2-cyclohexene-1-one), 3g**: yellow crystal, m.p. 188-190ºC; $^1$H NMR (300 MHz, $CDCl_3$) ppm: 1.12 (s, 6H, $CH_3$), 1.14 (s, 6H,$CH_3$), 2.17-2.33 (m, 8H, $CH_2$), 5.02 (s, 1H, CH), 7.27-7.49 (m, 4H, Ph-H), 11.41 (br. s, 1H, OH), 11.59 (s, 1H, OH).

**2,2′-(3-Nitrophenyl)methylene bis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one), 3h**: White crystal, m.p. 192-194ºC; $^1$H NMR (300 MHz, $CDCl_3$) ppm: 1.12 (s, 6H, $CH_3$), 1.27 (s, 6H,$CH_3$), 2.30-2.53 (m, 8H, $CH_2$), 5.54 (s, 1H, CH), 7.42-7.47 (m, 2H, Ph-H), 8.00 (s, 1H, Ph-H), 8.04 (d, *J* = 7.8 Hz, 1H, Ph-H), 11.65 (br. s, 1H, OH), 11.86 (s, 1H, OH).

**2,2′-(4-Nitrophenyl)methylene bis(3-hydroxy-5, 5-dimethyl-2-cyclohexene-1-one), 3i**: White crystal, m.p. 194-196ºC; $^1$H NMR (300 MHz, $CDCl_3$) ppm: 1.11 (s, 6H, $CH_3$), 1.23 (s, 6H,$CH_3$), 2.30-2.52 (m, 8H, $CH_2$), 5.54 (s, 1H, CH), 7.25 (d, *J* = 8.7 Hz, 2H, Ph-H), 8.14 (d, *J* = 8.7 Hz, 2H, Ph-H), 11.52 (br. s, 1H, OH), 11.80 (s, 1H, OH).

**2,2′-(3-Chlorophenyl)methylene bis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one), 3j**: White crystal, m.p. 186-188ºC; $^1$H NMR (300 MHz, $CDCl_3$) ppm: 1.10 (s, 6H, $CH_3$), 1.23 (s, 6H,$CH_3$), 2.13-2.50 (m, 8H, $CH_2$), 5.48 (s, 1H, CH), 6.96 (d, *J* = 6.9 Hz, 1H, Ph-H), 7.06 (s, 1H, Ph-H), 7.13-7.22 (m, 2H, Ph-H), 11.52 (br. s, 1H, OH), 11.90 (s, 1H, OH).

**2,2′-(4-Chlorophenyl)methylene bis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one), 3k**: White crystal, m.p. 146-148ºC; $^1$H NMR (300 MHz, $CDCl_3$) ppm: 1.10 (s, 6H, $CH_3$), 1.21 (s, 6H,$CH_3$), 2.27-2.49 (m, 8H, $CH_2$), 5.47 (s, 1H, CH), 7.01 (d, *J* = 8.1 Hz, 2H, Ph-H), 7.23 (d, *J* = 8.7 Hz, 2H, Ph-H), 11.58 (br. s, 1H, OH), 11.87 (s, 1H, OH).

**2,2′-(3-Bromophenyl)methylene bis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one), 3l**: White crystal, m.p. 190-192ºC; $^1$H NMR (300 MHz, $CDCl_3$) ppm: 1.10 (s, 6H, $CH_3$), 1.23 (s, 6H,$CH_3$), 2.28-2.61 (m, 8H, $CH_2$), 5.48 (s, 1H, CH), 7.01 (d, *J* = 7.8 Hz, 1H, Ph-H), 7.13 (t, 1H, Ph-H), 7.22 (s, 1H, Ph-H), 7.30 (d, *J* = 7.8 Hz, 1H, Ph-H), 11.61 (br. s, 1H, OH), 11.90 (s, 1H, OH).

**2,2′-(4-Bromophenyl)methylene bis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one), 3m**: White crystal, m.p. 182-184ºC; $^1$H NMR (300 MHz, $CDCl_3$) ppm: 1.10 (s, 6H, $CH_3$), 1.21 (s, 6H,$CH_3$), 2.27-2.49 (m, 8H, $CH_2$), 5.44 (s, 1H, CH), 6.95 (d, *J* = 8.1 Hz, 2H, Ph-H), 7.38 (d, *J* = 8.7 Hz, 2H, Ph-H), 11.62 (br. s, 1H, OH), 11.87 (s, 1H, OH).

**2,2′-(Thiophenyl)methylene bis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one), 3n**: White crystal, m.p. 160-162ºC; $^1$H NMR (400 MHz, $CDCl_3$) ppm: 1.06 (s, 6H, $CH_3$), 1.21 (s, 6H,$CH_3$), 2.28-2.40 (m, 8H, $CH_2$), 5.62 (s, 1H, CH), 6.63 (d, *J* = 3.2 Hz, 1H), 6.86-6.88 (m, 1H), 7.10 (d, *J* = 5.2 Hz, 1H), 11.77 (br. s, 1H, OH), 12.30 (s, 1H, OH); QTOF MS ($ES^+$): *m/z* Calcd for $C_{21}H_{27}O_4S$ $[M+H]^+$ 357.1630, Found $[M+H]^+$ 357.1631.

**2,2′-(Furyl)methylene bis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one), 3o**: White crystal, m.p. 140-142ºC; $^1$H NMR (400 MHz, $CDCl_3$) ppm: 1.16 (s, 6H, $CH_3$), 1.25 (s, 6H,$CH_3$), 2.21-2.41 (m, 8H, $CH_2$), 5.29-5.41 (m, 2H), 5.92 (s, 1H, CH), 6.27-6.31 (m, 1H), 11.60 (br. s, 1H, OH), 12.14 (s, 1H, OH); MS (ESI): *m/z* Calcd for $C_{21}H_{27}O_5$ $[M+H]^+$ 359.1858, Found $[M+H]^+$ 359.1857.

## Conclusions

The current work presents a powerful method for the preparation of 2,2′-arylmethylenebis(3-hydroxy-5,5-dimethyl-2-cyclohexene-1-one) derivatives using tamarind water as catalyst cum solvent. The general and efficient procedure offers several advantages including no usage of any organic solvent or costly reagent, high yield of the product, usage of very cheap and readily available catalyst precursors (tamarind fruit) and the facile separation of the products by simple filtration after washing with water followed by crystallisation with ethanol. All of these points make this process as a very useful and practical alternative in the synthesis of these compounds even in large scale preparation. The compounds **3l** and **3n** are new compounds and may have potential biological activities.

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