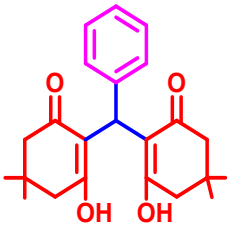
Electronic Supplementary Information (ESI)

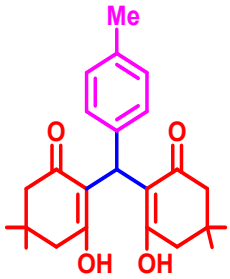
S1: Spectral Data of Representative Products of Tetraketone Derivatives

1. 2,2'-(phenylmethylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)



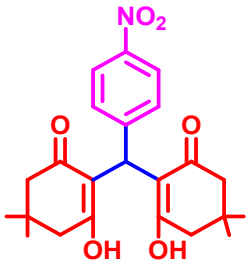
Colorless crystals; ^{1}H NMR (400 MHz, $CDCl_3$): δ 11.91 (brs, 1H), 7.24-7.28 (m, 2H), 7.14-7.18 (m, 1H), 7.08-7.10 (m, 2H), 5.53 (s, 1H), 2.28- 2.48 (m, 8H), 1.23 (s, 6H), 1.09 (s, 6H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ 190.59, 189.51, 138.13, 128.31, 126.85, 125.94, 115.67, 47.12, 46.51, 32.81, 31.49, 29.76, 27.47 ppm. HRMS (ES) Calcd: 368.3664. Found: 369.3722 $[M + H]^+$ and 370.3805 $[MH + 2]^+$.

2. 2,2'-(p-tolylmethylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)



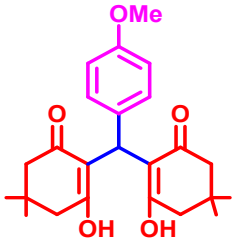
Light yellow solid; ^{1}H NMR (400 MHz, $CDCl_3$): δ 11.93 (brs, 1H), 7.07 (d, 2H), 6.97 (d, 2H), 5.50 (s, 1H), 2.37-2.51 (m, 8H), 2.28 (s, 3H), 1.22 (s, 6H), 1.09 (s, 6H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ 190.55, 189.53, 135.35, 134.97, 129.04, 126.74, 115.80, 54.19, 47.13, 46.50, 32.47, 31.49, 29.75, 28.33, 27.45, 20.99 ppm. HRMS (ES) Calcd: 382.2144. Found: 383.2246 $[M + H]^+$ and 384.2334 $[MH + 2]^+$.

3. 2,2'-((4-nitrophenyl)methylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)



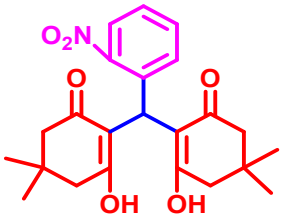
White solid; ^{1}H NMR (400 MHz, $CDCl_3$): δ 11.77 (brs, 1H), 8.10 (d, 2H), 7.22 (d, 2H), 5.52 (s, 1H), 2.29-2.49 (m, 8H), 1.21 (s, 6H), 1.09 (s, 6H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ 191.03, 189.67, 146.65, 146.18, 127.72, 123.57, 114.97, 54.17, 47.06, 33.31, 31.54, 29.56, 27.53 ppm. HRMS (ES) Calcd: 413.1838. Found: 414.1925 $[M + H]^+$ and 415.2004 $[MH + 2]^+$.

4. 2,2'-((4-methoxyphenyl)methylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)



White solid; ^{1}H NMR (400 MHz, $CDCl_3$): δ 11.91 (brs, 1H), 6.98 (d, 2H), 6.79 (d, 2H), 5.47 (s, 1H), 3.75 (s, 3H), 2.27-2.46 (m, 8H), 1.21 (s, 6H), 1.08 (s, 6H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ 190.51, 189.47, 157.66, 129.90, 127.88, 115.86, 113.72, 55.28, 47.14, 46.50, 32.10, 31.47, 29.76, 27.45 ppm. HRMS (ES) Calcd: 398.2093. Found: 399.2184 $[M + H]^+$ and 400.2245 $[MH + 2]^+$.

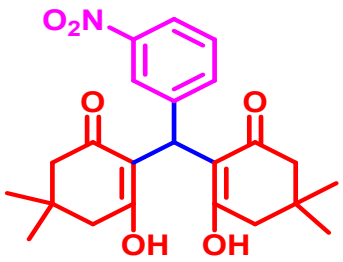
5. 2,2'-((2-nitrophenyl)methylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)



Yellow solid; ^{1}H NMR (400 MHz, $CDCl_3$): δ 11.59 (brs, 1H), 7.53 (d, 1H), 7.45 (t, 1H), 7.30 (t, 1H), 7.23 (d, 1H), 6.02 (s, 1H), 2.16-2.50 (m, 8H), 1.13 (s, 6H), 1.00 (s, 6H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ 190.99, 189.45, 149.79, 132.21,

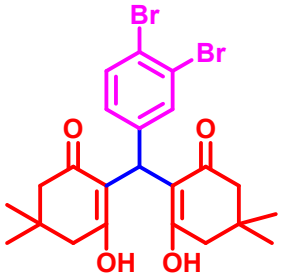
131.44, 129.67, 127.24, 124.40, 114.72, 46.93, 46.35, 31.97, 30.11, 28.62, 28.23 ppm. HRMS (ES) Calcd: 413.3705. Found: 414.3778 $[M + H]^+$ and 415.3788 $[MH + 2]^+$.

6. 2,2'-((3-nitrophenyl)methylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)



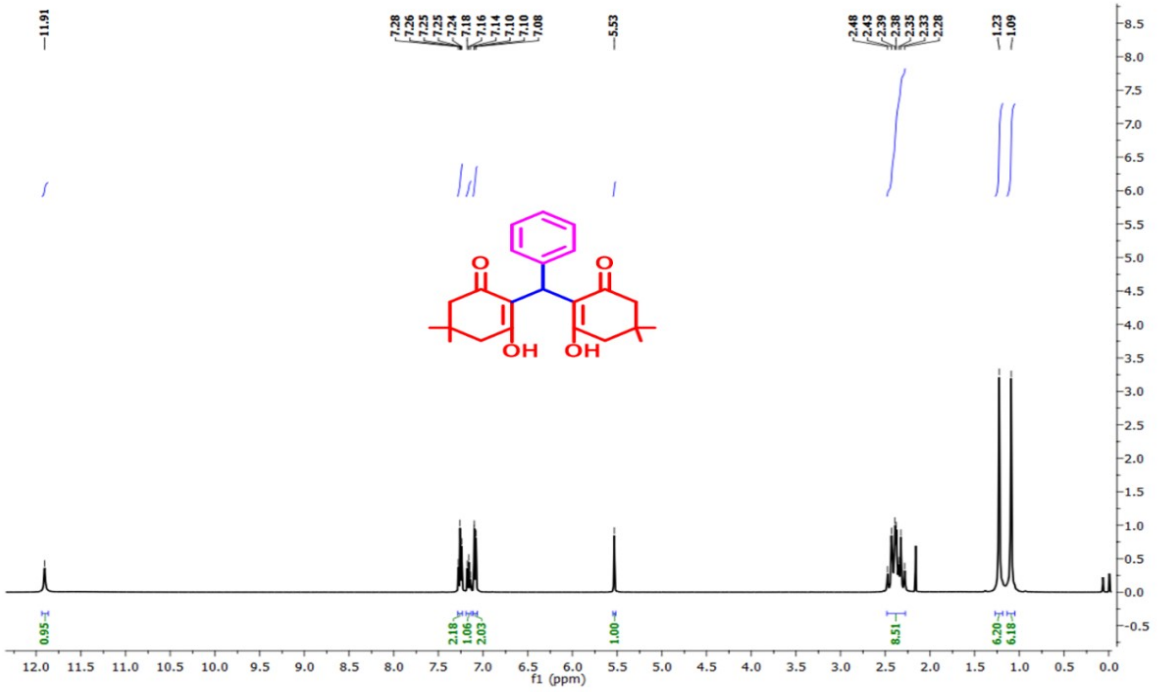
Yellow solid; ^{1}H NMR (400 MHz, $CDCl_3$): δ 11.84 (brs, 1H), 8.00 (d, 1H), 7.97 (s, 1H), 7.37-7.43 (m, 2H), 5.51 (s, 1H), 2.28-2.49 (m, 8H), 1.24 (s, 6H), 1.09 (s, 6H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ 191.19, 189.68, 148.44, 140.79, 132.99, 129.18, 122.27, 121.08, 114.81, 47.04, 46.47, 32.92, 31.49, 31.01, 29.75, 27.32 ppm. HRMS (ES) Calcd: 413.2006. Found: 414.2079 $[M + H]^+$ and 415.1933 $[MH + 2]^+$.

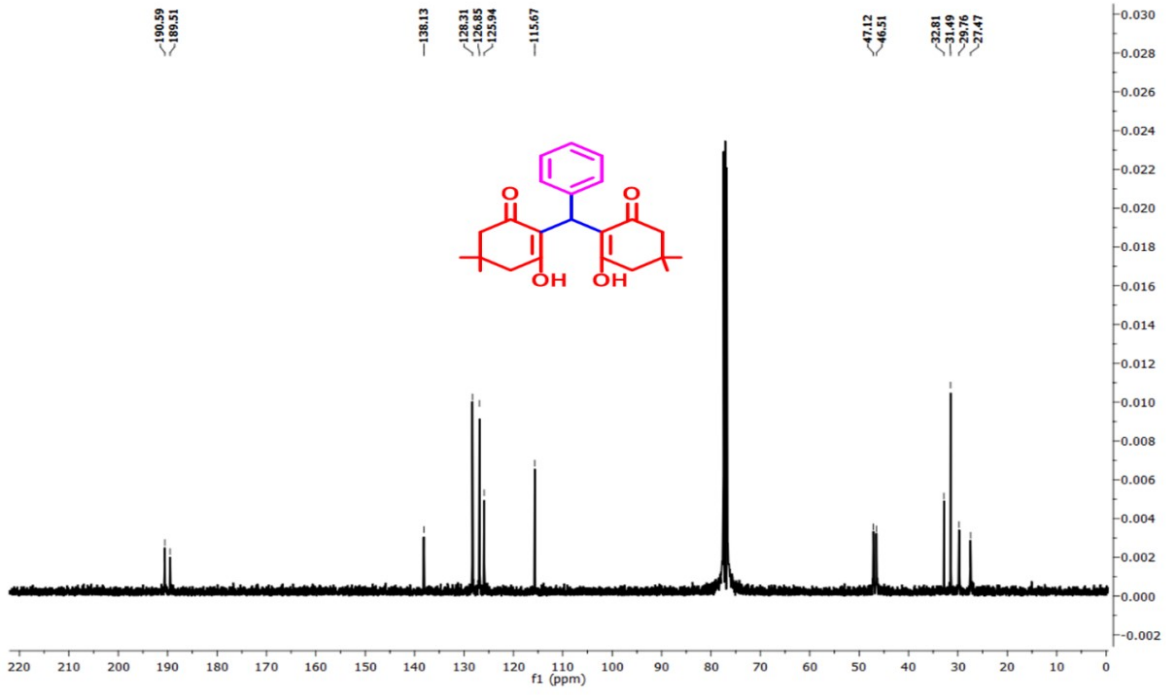
7. 2,2'-((3,4-dibromophenyl)methylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)



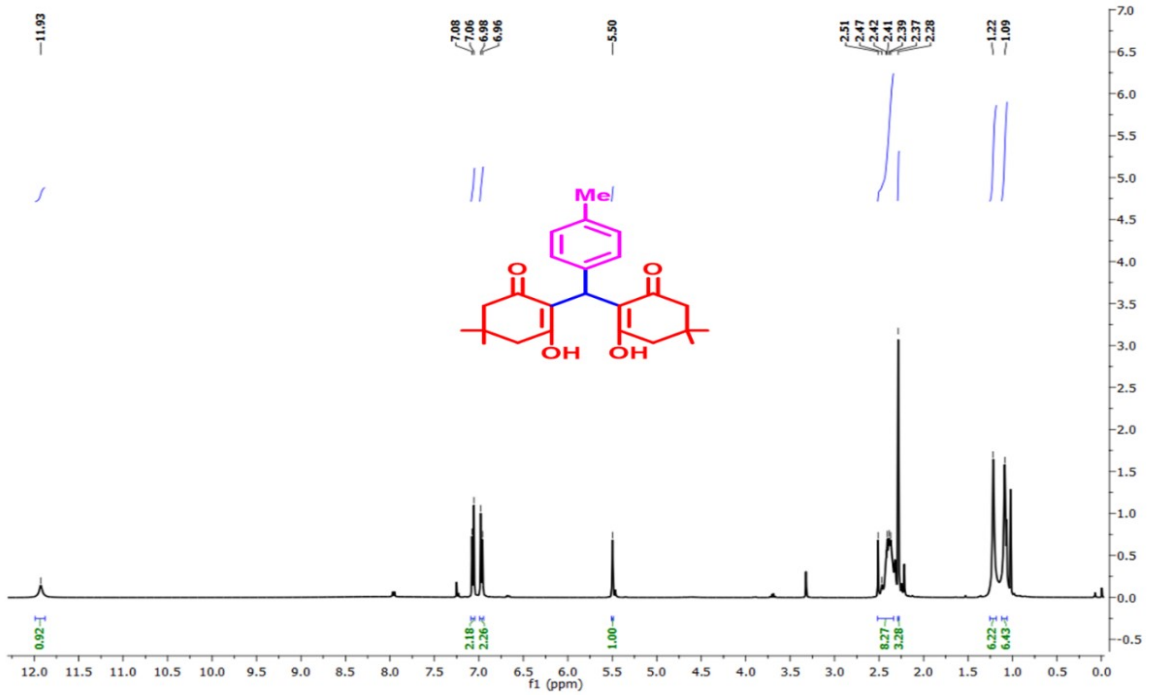
White solid; ^{1}H NMR (400 MHz, $CDCl_3$): δ 11.97 (brs, 1H), 6.77 (d, 1H), 6.59-6.63 (m, 2H), 5.49 (s, 1H), 2.33-2.41 (m, 8H), 1.22 (s, 6H), 1.10 (s, 6H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ 190.50, 189.44, 148.70, 147.07, 130.49, 118.93, 115.84, 110.91, 110.47, 55.90, 55.72, 47.15, 46.46, 32.37, 31.30, 29.99, 27.13 ppm. HRMS (ES) Calcd: 524.0198. Found: 525.0176 $[M + H]^+$ and 526.0252 $[MH + 2]^+$.

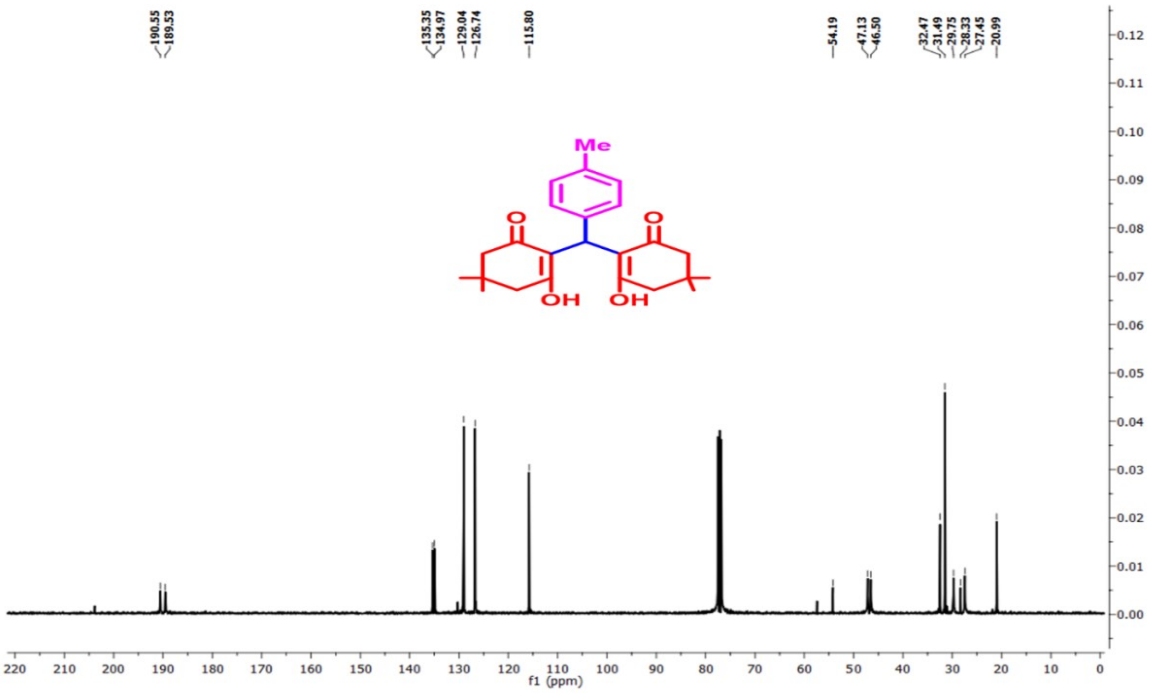
1. ^{1}H and ^{13}C spectra of 2,2'-(phenylmethylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)



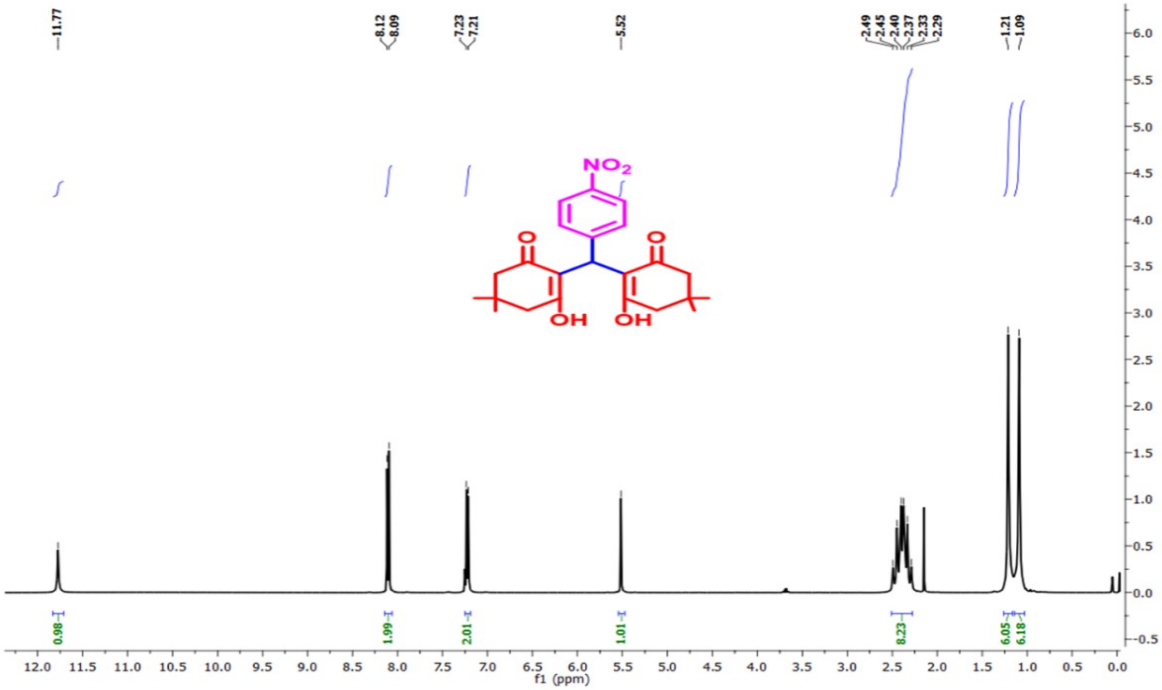


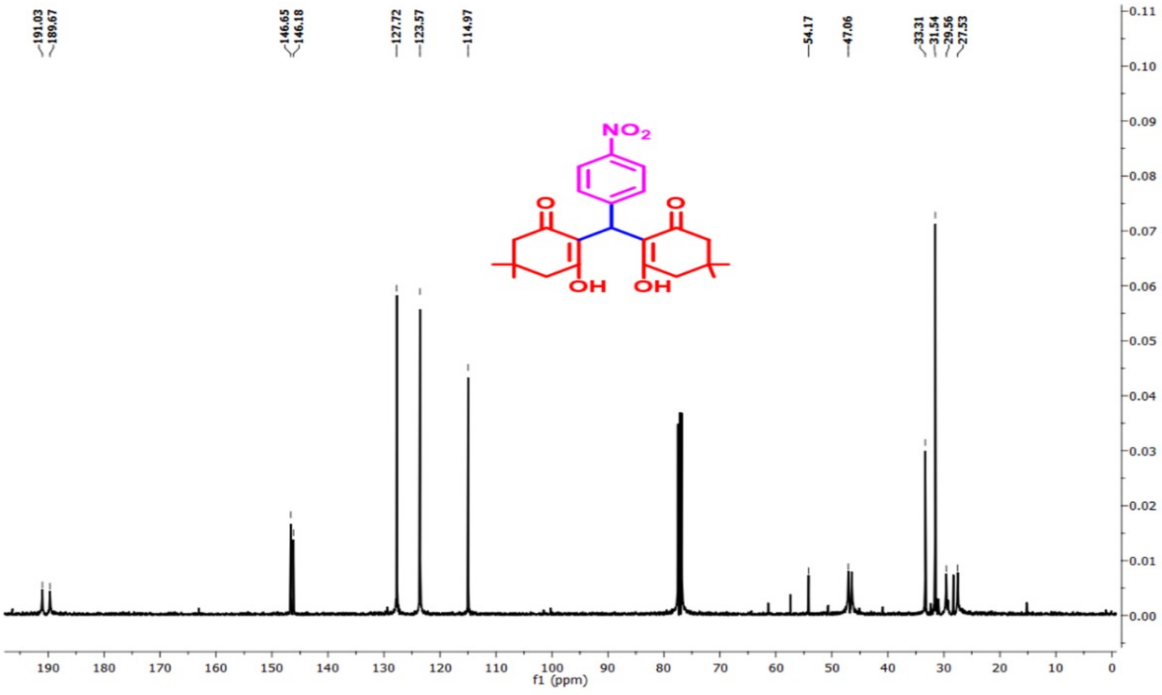
2. ^{1}H and ^{13}C spectra of 2,2'-(p-tolylmethylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)



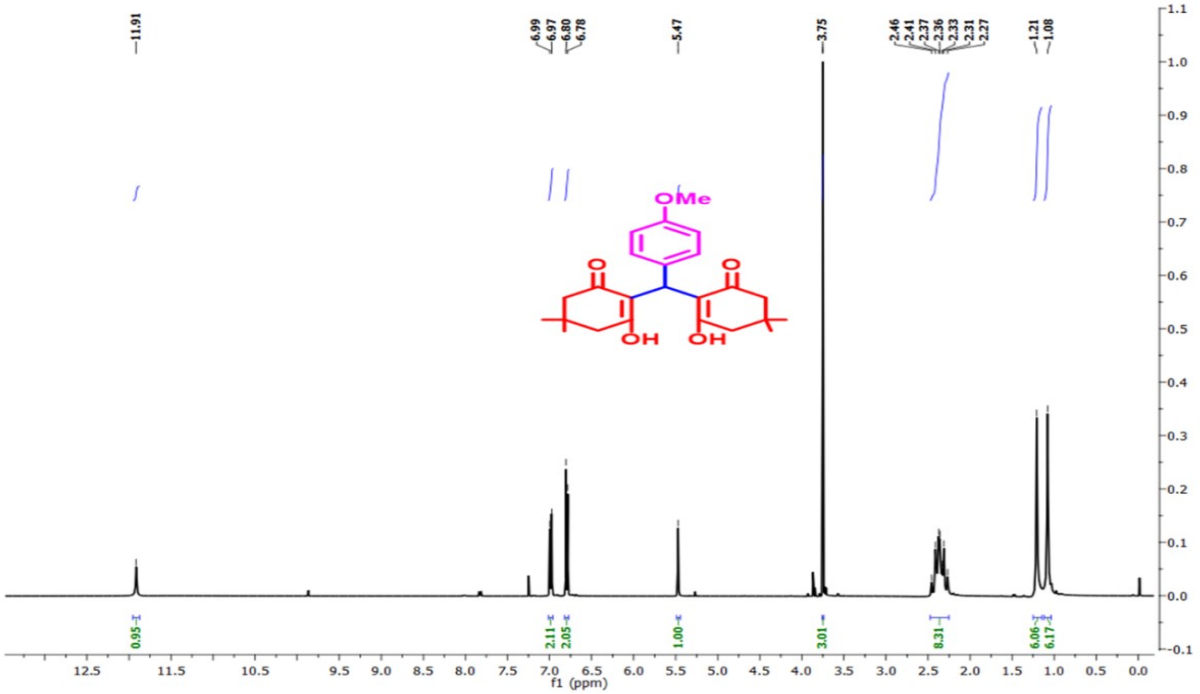


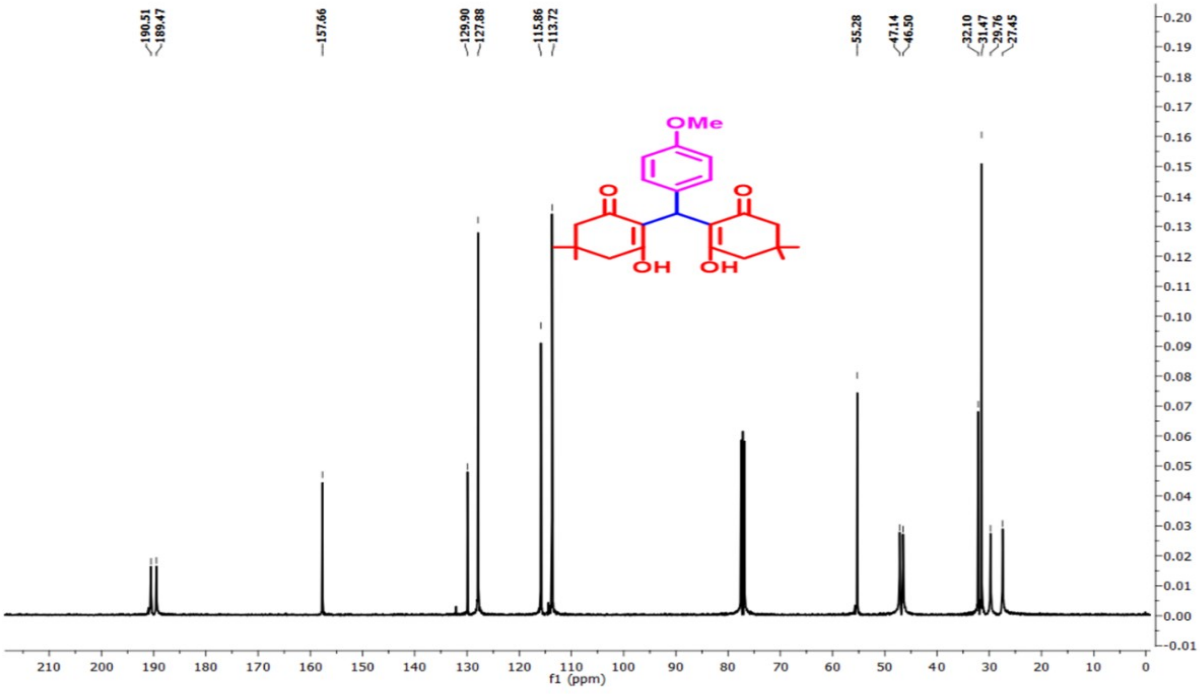
3. ^{1}H and ^{13}C spectra of 2,2'-((4-nitrophenyl)methylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)



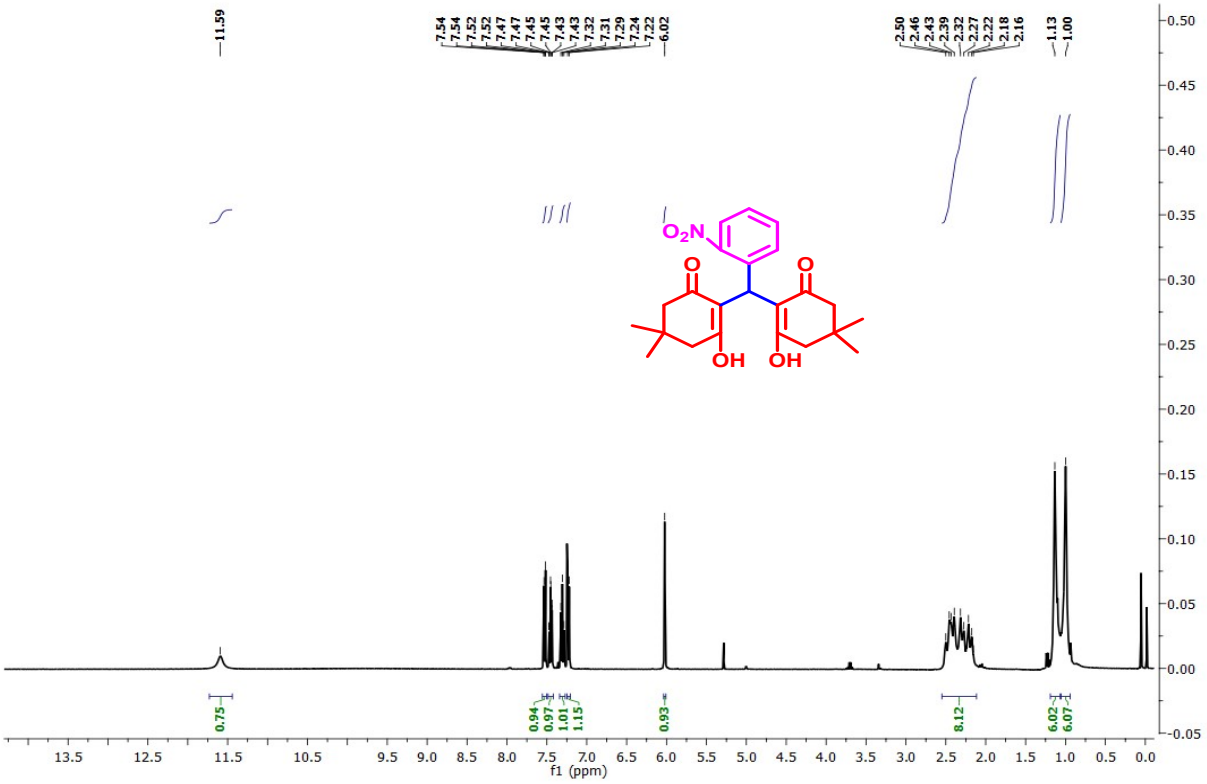


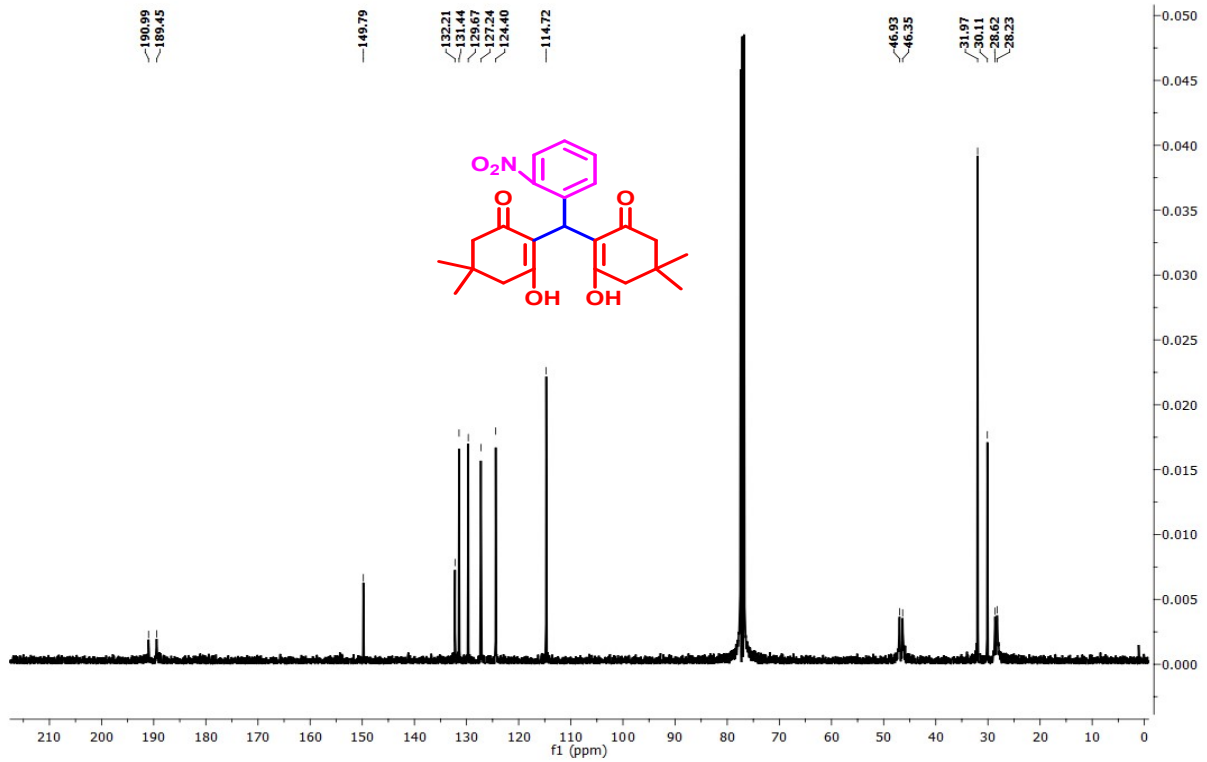
4. ^{1}H and ^{13}C spectra of 2,2'-((4-methoxyphenyl)methylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)



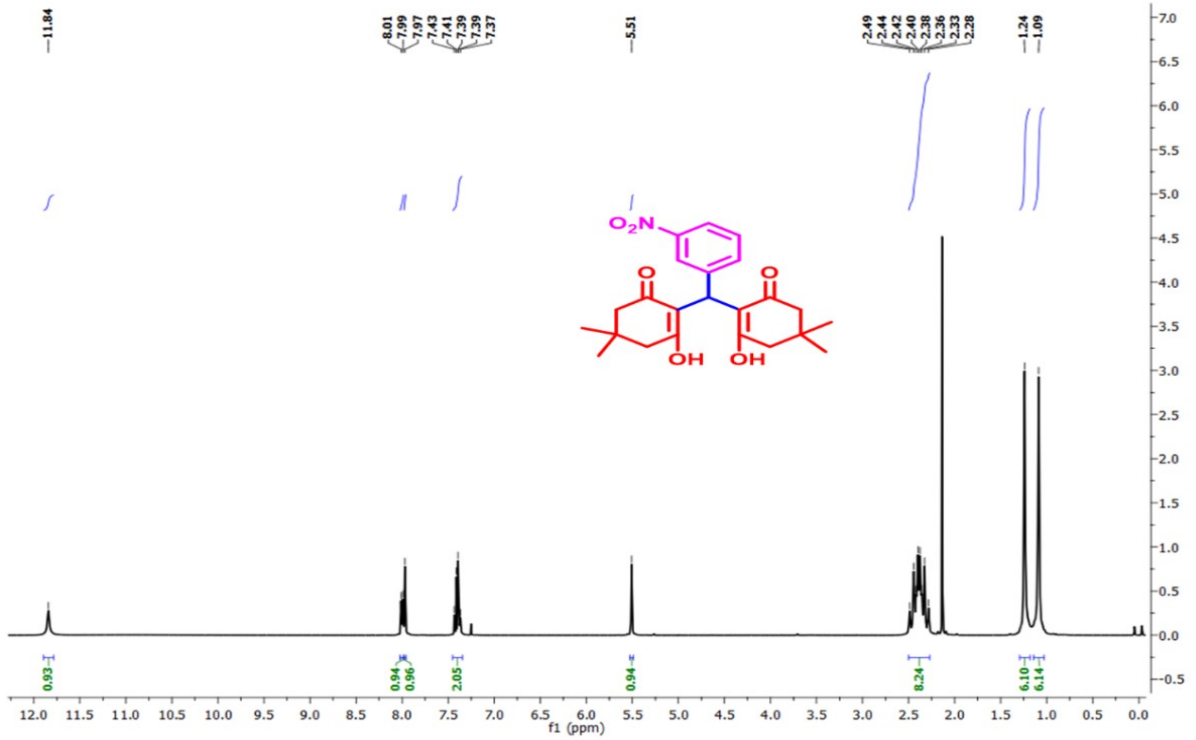


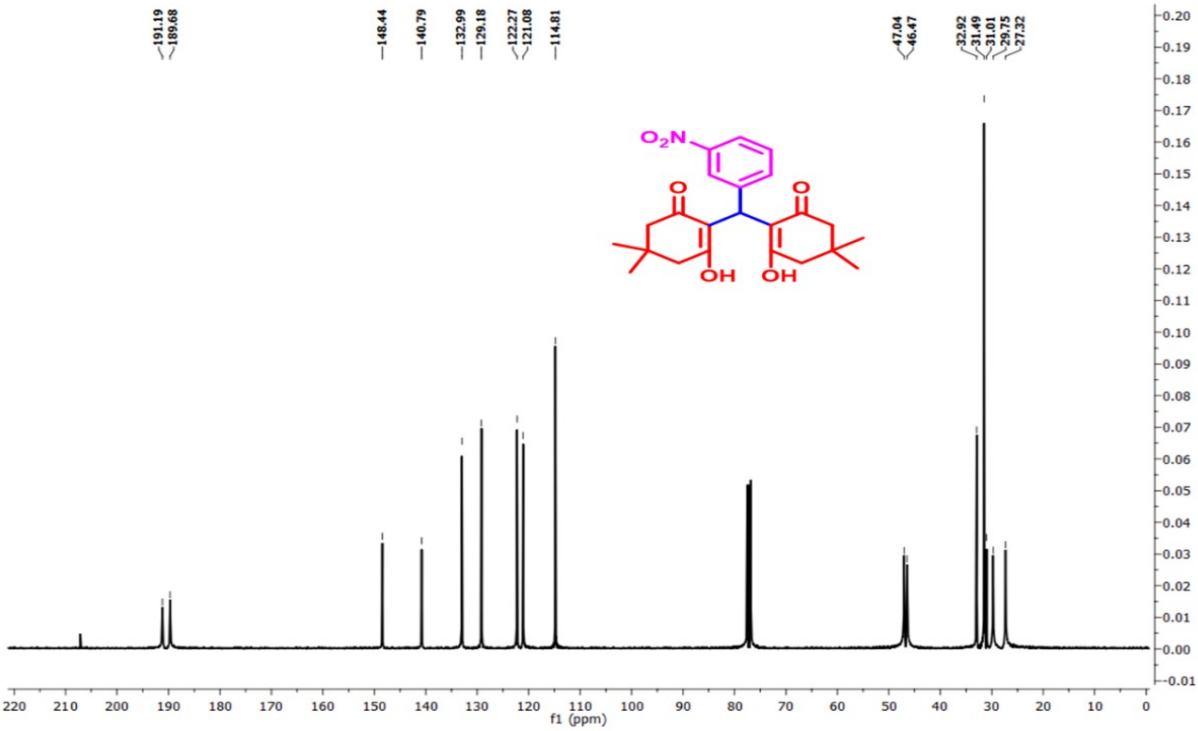
5. ^{1}H and ^{13}C spectra of 2,2'-((2-nitrophenyl)methylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)



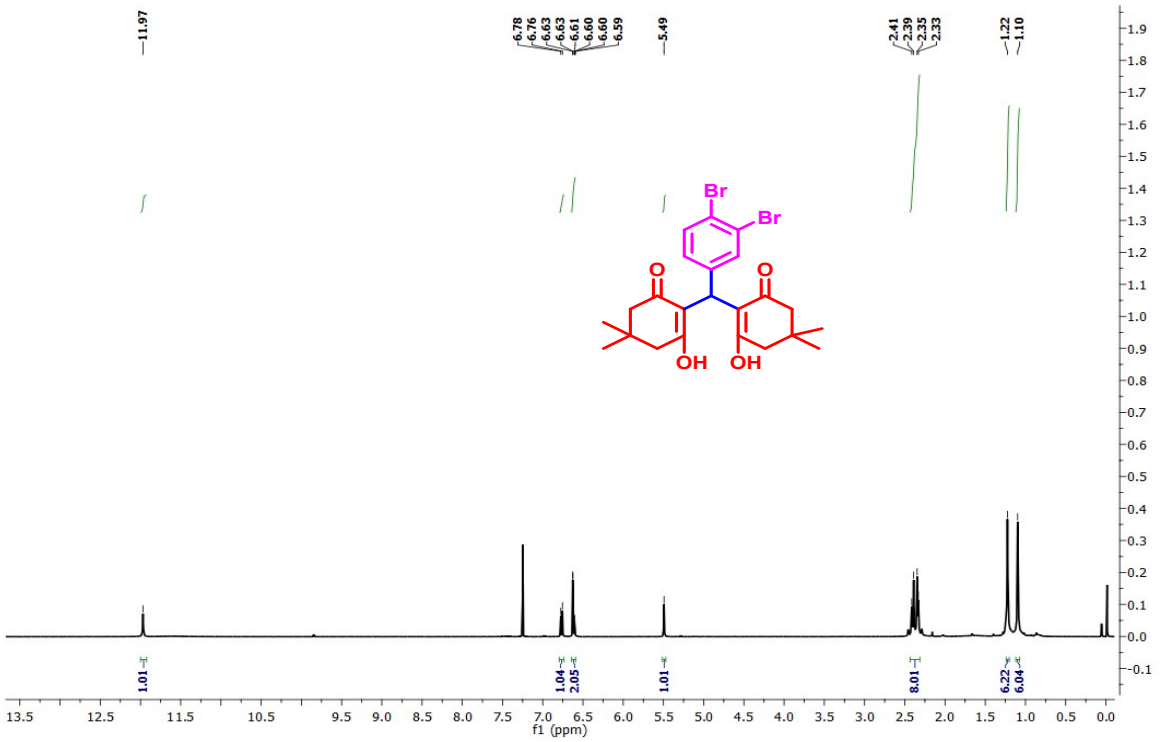


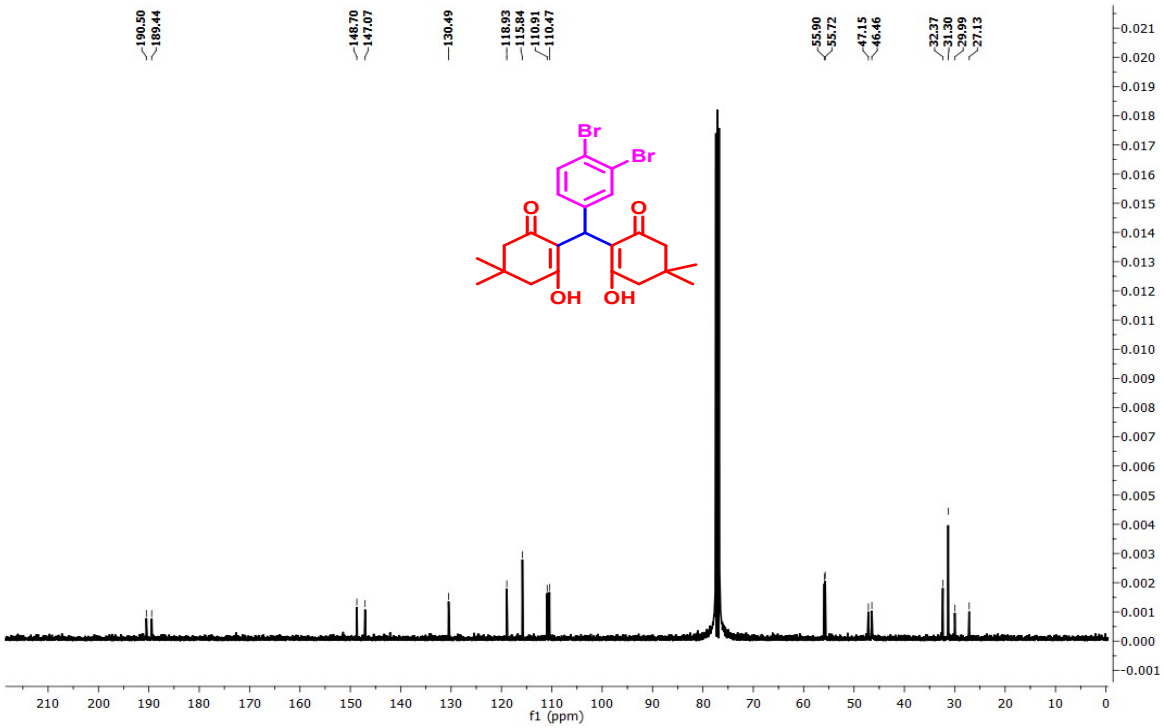
6. ^{1}H and ^{13}C spectra of 2,2'-((3-nitrophenyl)methylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)





7. ^{1}H and ^{13}C spectra of 2,2'-((3,4-dibromophenyl)methylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)





S2: Single Crystal X-ray Diffraction Analysis of 2,2'-(phenylmethylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)

Compound (**2,2'-(phenylmethylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)**) was dissolved in methyl alcohol and single crystals (appropriate for X-ray diffraction studies) were grown by allowing slow evaporation of the solvent at room temperature. The X-ray diffraction data was collected on X'calibur CCD diffractometer employing a graphite monochromatized Mo/Kα radiation (λ = 0.71073 Å) at a temperature of 293 K. The crystal data was analyzed using CrysAlis pro software available with the diffractometer. Further, least square refinement, following introduction of anisotropic displacement parameters, yielded the R values mentioned in the **Table S1**. The structure was solved by direct methods using SHELXT-2018 and refined by the full-matrix least-squares method on F^2 (SHELXL-2018/3). All calculations were carried out using the OLEX2 package of the crystallographic programs and the program Mercury (4.2.0) was used for molecular graphics. The selected bond lengths, bond angles, *etc.* are given in **Table S1**.

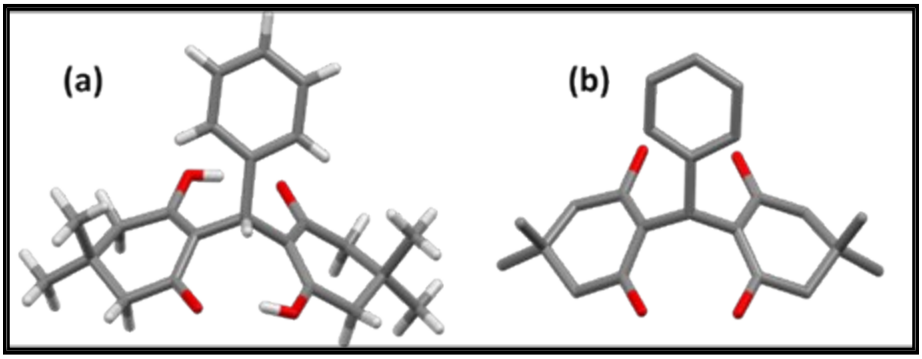


Figure S1: Molecular structures (**a & b**) of **2,2'-(phenylmethylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)** H atoms are omitted in **b** for clarity. C = gray, O = red.

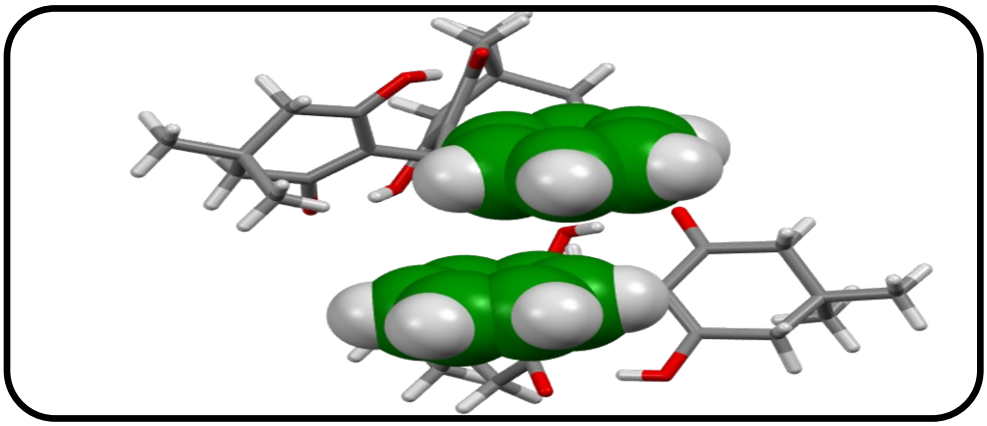


Figure S2: Double decker π-stacked arrangement of two phenolic ring between the adjacent molecules.

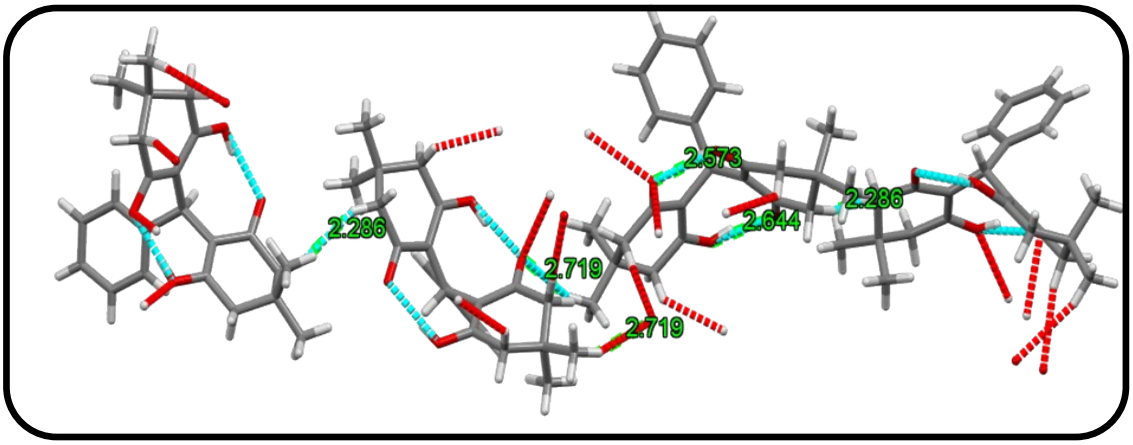


Figure S3: 1D arrangement of **2,2'-(phenylmethylene)bis(3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)** *via* intra- and intermolecular non-covalent interactions within the molecule and between the adjacent molecules.

Table S1: Crystal data and structure refinement for compound

Identification Name	**2,2'-(phenylmethylene)bis (3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)**
Empirical formula	$C_{23}H_{28}O_4$
Formula weight	368.45
Temperature/K	100
Crystal system	Monoclinic
Space group	P 21/n
a/Å	20.9275(4)
b/Å	20.9275(4)
c/Å	36.9289(11)
α/°	90
β/°	90
γ/°	90
Volume/Å^3	16173.4(8)
Z	450
ρ_{calc}g/cm^3	1.211
μ/mm^{-1}	0.106
F(000)	6336

Identification Name	**2,2'-(phenylmethylene)bis (3-hydroxy-5,5-dimethylcyclohex-2-en-1-one)**
Radiation	Mo/K_α (λ = 0.71073)
2Θ range for data collection/°	3.2550 to 28.3542
Index ranges	$-10 \leq h \leq 10$, $-10 \leq k \leq 10$, $-20 \leq l \leq 20$
No of Reflections measured	10572
Independent reflections	6374
Goodness-of-fit on F^2	1.06
R [$F^2 > 2\sigma(F^2)$], *wR*(all data)	0.0781, 0.1813