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An efficient and facile synthesis of 3-aryl-1,5-pentanedione derivatives under solvent-free conditions

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Abstract An efficient synthesis of 3-aryl-1,5-pentanedione derivatives from the reaction of aromatic aldehydes and 1,2-diphenylethanone has been reported. Although 3-aryl-1,5-pentanedione is a frequently used synthetic intermediate, few mature methods have been reported for the synthesis of this kind of derivative. In our research, this reaction could be carried out smoothly promoted by NaOH under solvent-free conditions. Because of the low cost, and readily available reagent, NaOH is an efficient catalyst for the synthesis of these dione compounds. The other advantages of this procurement are high yields, simple operation, atomic economy, mild reaction conditions, and environmental friendliness. The d-D structure has been additional confirmed by X-ray diffraction analysis.

Keywords 1,5-Pentanedione · 1,2-Diphenylethanone · Solvent-free · Green synthesis · Environmental friendliness

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Introduction

The open-chain 1,5-pentanedione compounds are important intermediates in organic synthesis chemistry, which always involves synthesis of piperidine and pyran compounds [1–3]. Under investigation, it was found that few in the literature have reported the synthesis of these 1,5-pentanedione derivatives.

Green chemistry techniques become a major challenge for chemists that aim to conserve resources and minimize economic burden [4, 5]. In recent years, many new methods and techniques have been developed for green synthesis chemistry, such as ionic liquid synthesis [6–8], the ultrasonic technique [9–11], the microwave irradiation technique [12, 13], and solvent-free organic synthesis [4, 14–17]. Because of its simple operation, ease of control, and convenient disposition, solvent-free synthesis is the most efficient approach for application in many classic reactions, for example, the Grignard reaction [18], the Reformatsky reaction [19, 20], aldol condensations [21–24], Dieckmann condensations [25], the phenol coupling reaction [26–28], the reduction reaction [29–31], peptide bond formation [32], and so on.

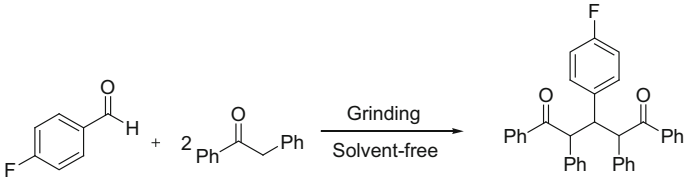
In this article, we report an efficient and facile process for preparation of open-chain 1,5-pentanedione derivatives under solvent-free conditions.

Results and discussion

Before our research, we investigated the reported methods for the synthesis of this kind of compound. Beyond our expectations, few papers were found to synthesize similar 3-aryl-1,5-pentanedione derivatives. It could be definitely confirmed that Lantaño [33] had reported synthesizing the similar 1,5-pentanedione using the strong base (MeONa) as a catalyst. This reaction condition was harsh because of anhydrous EtOH and the long reaction time (72 h). Bagavant [34] also reported a process to prepare similar products in MeONa/MeOH after about 20 h. These approaches both needed a strong base (MeONa), a long reaction time, and organic solvent. So, it is necessary to find an efficient and facile approach for synthesis of open-chain 1,5-pentanedione derivatives. Because of the advantages of solvent-free synthesis, we planned to carry out our research under solvent-free conditions.

In order to implement our research plan, 4-fluorobenzaldehyde **1a** (1 mmol) and 1,2-diphenylethanone **2** (2 mmol) were chosen as standard substrates for optimizing the reaction conditions, and various parameters were studied, such as the catalysts, temperature, and reaction time (Table 1) by grinding starting materials in a mortar. As shown in Table 1, when acid catalysts such as $B(OH)_3$, $ZnCl_2$, *P*-TSA (*p*-toluenesulfonic acid), NH_2SO_3H, or a weak base such as K_2CO_3, Na_2CO_3 were used in model reactions, the products were not found or they had very low yields (Table 1, Entry 1–6). However, when strong alkali (NaOH) was used in this operation, the reaction could be carried out smoothly with a good yield (Table 1, Entry 7). As the amount of NaOH was 0.1 g, the reaction time was about 15 min, and room temperature (r.t.), it gave the best yield (Table 1, Entry 9). Higher temperatures did not improve the yields (Table 1, Entry 11, 12). Similar, longer reaction times were no help for the yields (Table 1, Entry 10, 11). Different amounts of NaOH were also studied in

Table 1 Optimization of the reaction of 4-fluorobenzaldehyde and 1,2-diphenylethanone



1a **2** **3a**

Entry	Conditions				Yield (%)[a]
	Catalyst	Catalyst amount (g)	Temperature (°C)	Time (min)	
1	$B(OH)_3$	0.2	r.t.	10	None
2	$ZnCl_2$	0.2	r.t.	10	None
3	P-TSA	0.2	r.t.	10	None
4	NH_2SO_3H	0.2	r.t.	10	None
5	K_2CO_3	0.2	55	10	Trace
6	Na_2CO_3	0.2	55	10	Trace
7	NaOH	0.2	r.t.	10	61
8	NaOH	0.1	r.t.	10	89
9	NaOH	0.1	r.t.	15	92
10	NaOH	0.1	r.t.	30	91
11	NaOH	0.1	50	30	90[b]
12	NaOH	0.1	70	30	91[b]
13	NaOH	0.3	r.t.	15	50
14	NaOH	0.4	r.t.	15	45
15	KOH	0.1	r.t.	15	87
16	KOH	0.2	r.t.	15	58
17	NaOH (EtOH)	0.1	r.t.	60	None
18	NaOH (EtOH)	0.1	70	60	None

General reaction condition: the reactions were carried out using 4-fluorobenzaldehyde **1a** (1 mmol) and 1,2-diphenylethanone (2 mmol) and several kinds of catalysts

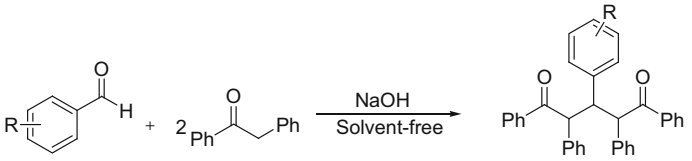
[a] Isolated yields

[b] After grinding, the mixture was transferred into a flask and heated at 70 °C for the reaction

this reaction, it showed the yields were gradually reduced with the increase loading of NaOH (Table 1, Entry 7, 8, 14, 15). It could be explained that more NaOH could turn the reagents into solid more quickly, which hindered the reaction from going to completion. The other stronger base, KOH, could also promote the reaction, but the effect was the same as NaOH. The reaction was also checked in EtOH, but no product was found (Table 1, Entry 17, 18). So, solvent-free, NaOH (0.1 g), room temperature, and 15 min were the preferred reaction conditions.

Under the optimized conditions, different aromatic aldehydes with different substituted groups were reacted with 1,2-diphenylethanone under a solvent-free condition to expand the scope of the reaction, and all reactions could be carried out smoothly, and corresponding products could be obtained with high yields

Table 2 Synthesis of 3-aryl-1,5-pentanedione derivatives under a solvent-free condition



Entry	R	Product	Time (min)	Yield (%)[a]
1	4-F	**3a**	15	92
2	3-Cl	**3b**	20	85
3	4-Cl	**3c**	10	93
4	3,4-Cl_2	**3d**	10	90
5	3-Br	**3e**	15	88
6	4-Br	**3f**	10	87
7	4-CH_3	**3g**	20	91
8	4-CH_3O	**3h**	15	92
10	3,4-OCH_2O	**3j**	10	93

General reaction condition: different aromatic aldehydes **1** (1 mmol), 1,2-diphenylethanone **2** (2 mmol) and 0.1 g NaOH as catalyst under a solvent-free condition at r.t

[a] Isolated yields

(85–93 %). The results are summarized in Table 2. As shown in Table 2, the aldehydes bearing either electron-withdrawing or electron-donating groups perform well in this reaction. Therefore, we could conclude the electronic nature of the substituted groups has no significant effect on this reaction.

The structures of the products derivatives were characterized by their IR, ^{1}H NMR, ^{13}C NMR, and HRMS spectra. We take **3a** as an example to analyze their structure. In its IR, the wave numbers at 1675 and 1596 cm^{-1} are the signals of carbonyl (C=O). In its ^{1}H NMR, The triplet signal at $\delta = 4.18$ (1H, $J = 10.4$ Hz) ppm is due to the tertiary hydrogen of C-3. The doublet signals at 5.51 (1H, $J = 9.6$ Hz) and 5.60 (1H, $J = 10.8$ Hz) are the two protons of C-2 and C-4. The products have four stereoisomers: two meso forms (**A**, **B**) and one pair of enantiomers(**C**, **D**) (Fig. 1). However, the structures must not be the two mesomers **A** or **B**. Because the mesomers **A** or **B** have planes of symmetry, leading to the same chemical environment of C-2 and C-4, their attached hydrogen atoms will have identical chemical shifts in NMR spectra. So the structure may be C or D. Fortunately, we obtained the crystal structure of the product **3d**. With the help of X-diffraction, we could confirm the product was the structure of the **C** (see in Fig. 2). The absolute configuration of the product is "2R" and "4R." We did not find its enantiomer **D** (2S, 4S). This condition may be a result of the characteristic of a solvent-free reaction. This analysis results can be found in [33].

The phenyl protons are distributed from 6.78 to 7.83 ppm. In its ^{13}C NMR, the chemical shifts of the 35 carbon atoms are at 49.5, 55.1, 58.8, 113.8, 114.0, 126.2,

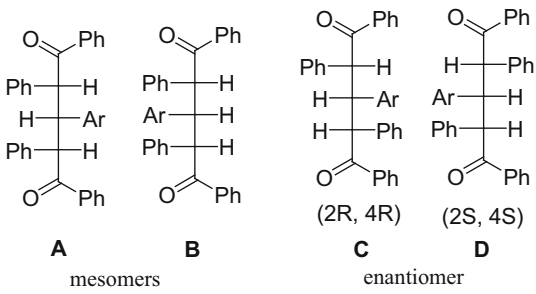


Fig. 1 Stereoisomers of products

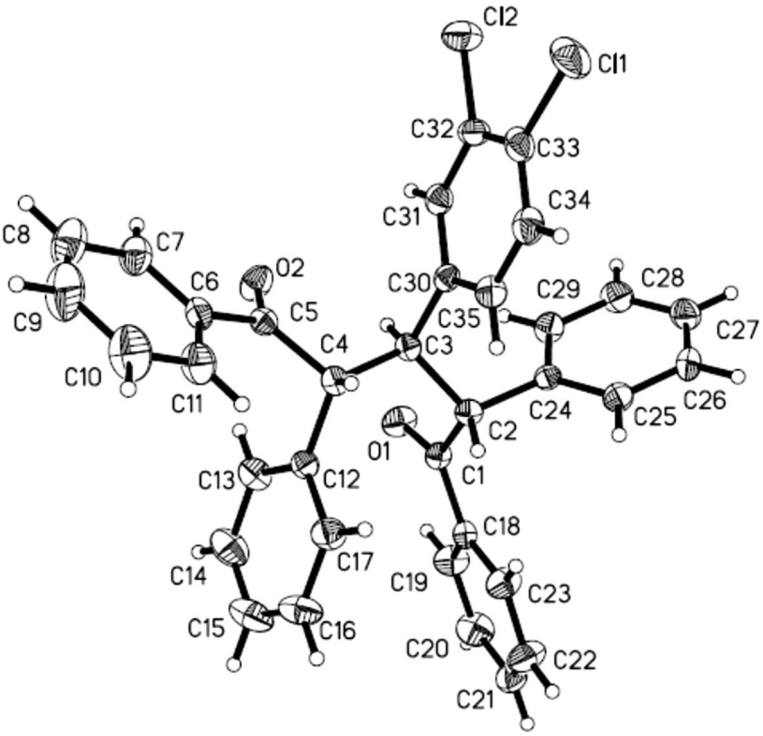


Fig. 2 The crystal-structure of **3d**

127.3, 127.8, 128.0, 128.1, 128.6, 129.6, 130.5, 132.5, 133.1, 135.5, 136.2, 137.7, 138.2, 158.8, 161.2, 197.2, and 198.2, respectively. In the HR mass spectrum, the calculated and found values of m/z for $C_{35}H_{27}F$ $[M + H]^+$: 467.2170, and 467.2155. The structure of **3d** was also confirmed by X-ray diffraction analysis; the crystal structure **3d** is shown in Fig. 2.

Conclusions

In summary, we have reported an efficient and convenient method for preparation of 3-aryl-1,5-pentanedione derivatives by grinding aromatic aldehydes and 1,2-diphenylethanone under solvent-free conditions. As this uses inexpensive and easily available NaOH as catalyst, this was an attractive approach to give open-chain 1,5-pentanedione derivatives. Furthermore, a solvent-free condition have other advantages, such as good yields, lower cost, reduced environmental impact, and simple operation.

Experimental

All reactions were performed at room temperature under air atmosphere on an agate mortar. The melting points were metered on an XT-5 micro melting point apparatus and were uncorrected. IR spectra were obtained from a FT Bruker Tensor 27 spectrometer using KBr as background. ^{1}H NMR (400 MHz) and ^{13}C NMR (100 MHz) spectra were recorded under solution in DMSO-d_6 or $CDCl_3$ using a Bruker NMR spectrometer with TMS as an internal standard. HRMS were measured with a Bruker microTOF-Q134 instrument. All reagents were purchased commercially and used without further purification. X-ray diffractions were recorded on a Siemens P4 or Simart-1000 diffractometer.

General procedure for the synthesis of 3-aryl-1,5-pentanedione

The mixture of aromatic aldehydes 1 (1 mmol), 1,2-diphenylethanone 2 (2 mmol) and 0.1 g NaOH was added into a mortar. Then, the mixture was ground at room temperature about 10–20 min and the solid was formed. The solid was pour into water and soaked for 30 min, and the products were floated on the water. Then, filtered, and then washed twice with 10 mL of water thoroughly. The separated solid was dried and recrystallized from 95 % ethanol.

*3-(4-Fluorophenyl)-1,2,4,5-tetraphenylpentane-1,5-dione (**3a**)*

m.p. 210–212 °C; IR (KBr, v, cm^{-1}): 1675, 1596, 1579, 1509, 1491, 1448, 1264, 1227, 1177, 1078, 1003, 979, 825, 758, 693, 548 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 4.84 (1H, t, J = 10.4 Hz, CH), 5.51 (1H, d, J = 9.6 Hz, CH), 5.60 (1H, d, J = 10.8 Hz, CH), 6.78 (2H, t, J = 8.8 Hz, ArH), 6.85–6.92 (4H, m, ArH), 6.99 (2H, t, J = 7.2 Hz, ArH), 7.12 (2H, d, J = 7.6 Hz, ArH), 7.31 (2H, t, J = 7.6 Hz, ArH), 7.37 (3H, t, J = 7.6 Hz, ArH), 7.43–7.50 (5H, m, ArH), 7.66 (2H, d, J = 7.2 Hz, ArH), 7.83 (2H, d, J = 7.2 Hz, ArH);^{13}C NMR (100 MHz, DMSO-d_6) (δ, ppm):49.5, 55.1, 58.8, 113.8, 114.0, 126.2, 127.3, 127.8, 128.0, 128.1, 128.6, 129.6, 130.5, 132.5, 133.1, 135.5, 136.2, 137.7, 138.2, 158.8, 161.2, 197.2, 198.2; HRMS m/z calculated for $C_{35}H_{27}F$ $[M + H]^+$: 467.2170, found: 467.2155.

*3-(3-Chlorophenyl)-1,2,4,5-tetraphenylpentane-1,5-dione (**3b**)*

m.p. 218–219 °C; IR (KBr, *v*, cm^{-1}): 1672, 1596, 1571, 1491, 1448, 1265, 1193, 1174, 1079, 1003, 975, 785, 693, 629, 542, 516 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$) (δ, ppm): 4.84 (1H, dd, $J = 2.4$ Hz, $J = 7.6$ Hz, CH), 5.06 (1H, d, $J = 7.6$ Hz, CH), 5.19 (1H, d, $J = 10.0$ Hz, CH), 6.89 (2H, d, $J = 5.2$ Hz, ArH), 6.96–6.99 (1H, m, ArH), 7.01–7.06 (5H, m, ArH), 7.12 (3H, t, $J = 7.6$ Hz, ArH), 7.21–7.27 (5H, m, ArH), 7.37 (4H, t, $J = 7.2$ Hz, ArH), 7. 60 (2H, d, $J = 7.6$ Hz, ArH), 7.72 (2H, d, $J = 7.2$ Hz, ArH); ^{13}C NMR (100 MHz, DMSO-d_6) (δ, ppm):49.9, 54.8, 58.4, 125.6, 126.4, 127.3, 127.9, 128.1, 128.7, 129.6, 130.4, 132.0, 132.6, 133.2, 135.3, 135.5, 136.1, 137.4, 144.6, 196.9, 198.0; HRMS m/z calculated for $C_{35}H_{27}Cl$ $[M + Na]^+$: 505.1694, found: 505.1701.

*3-(4-Chlorophenyl)-1,2,4,5-tetraphenylpentane-1,5-dione (**3c**)*

m.p. 229–230 °C; IR (KBr, *v*, cm^{-1}): 1672, 1597, 1579, 1492, 1447, 1267, 1177, 984, 823, 754, 701, 689, 609, 545 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) (δ, ppm):4.84 (1H, t, $J = 10.4$ Hz, $J = 10.0$ Hz, CH), 5.52 (1H, d, $J = 9.6$ Hz, CH), 5.60 (1H, d, $J = 10.8$ Hz, CH), 6.84–6.91 (4H, m, ArH), 7.01 (4H, dd, $J = 2.4$ Hz, $J = 7.6$ Hz, ArH), 7.13 (2H, d, $J = 7.6$ Hz, ArH), 7.31 (2H, t, $J = 7.6$ Hz, ArH), 7.37 (2H, t, $J = 7.6$ Hz, ArH), 7.42–7.48 (6H, m, ArH), 7.66 (2H, d, $J = 7.6$ Hz, ArH), 7.83 (2H, d, $J = 7.2$ Hz, ArH); ^{13}C NMR (100 MHz, DMSO-d_6) (δ, ppm): 49.5, 54.8, 58.6, 126.3, 127.2, 127.9, 128.0, 128.7, 129.6, 130.0, 130.4, 132.6, 133.1, 135.3, 136.0, 137.5, 141.2, 197.0, 198.0; HRMS m/z calculated for $C_{35}H_{27}Cl[M + Na]^+$: 505.1694, found: 505.1703.

*3-(3,4-Dichlorophenyl)-1,2,4,5-tetraphenylpentane-1,5-dione (**3d**)*

m.p. 230–231 °C; IR (KBr, *v*, cm^{-1}): 1672, 1596, 1579, 1492, 1472, 1448, 1399, 1296, 1267, 1194, 1175, 1128, 1028, 1003, 984, 828, 754, 693, 623, 526 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) (δ, ppm):4.82 (1H, t, $J = 10.0$ Hz,CH), 5.55 (1H, d, $J = 9.6$ Hz, CH), 5.64 (1H, d, $J = 10.4$ Hz, CH), 6.84–6.93 (4H, m, ArH), 7.03 (2H, t, $J = 7.2$ Hz, ArH), 7.15 (2H, d, $J = 7.2$ Hz, ArH), 7.23 (1H, d, $J = 8.0$ Hz, ArH), 7.32 (3H, t, $J = 7.6$ Hz, ArH), 7.37–7.44 (5H, m,ArH), 7.47 (2H, t, $J = 7.2$ Hz, ArH), 7.67 (2H, d, $J = 7.6$ Hz, ArH), 7.85 (2H, d, $J = 7.2$ Hz, ArH); ^{13}C NMR (100 MHz, DMSO-d_6) (δ, ppm): 49.5, 54.8, 58.7, 126.3, 127.2, 127.9, 128.1, 128.7, 129.6, 130.0, 130.4, 132.6, 133.1, 135.3, 136.0, 137.5, 141.2, 197.0, 198.0; HRMS m/z calculated for $C_{35}H_{26}Cl_2$ $[M + H]^+$: 517.1484, found: 517.1492.

*3-(3-Bromophenyl)-1,2,4,5-tetraphenylpentane-1,5-dione (**3e**)*

m.p. 215–216 °C; IR (KBr, *v*, cm^{-1}): 1673, 1596, 1566, 1491, 1448, 1265, 1193, 1175, 1072, 975, 784, 698, 629, 542, 515 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) (δ, ppm):4.80 (1H, t, $J = 10.0$ Hz,CH), 5.53 (1H, d, $J = 9.6$ Hz, CH), 5.63 (1H, d, $J = 10.8$ Hz, CH), 6.83–6.93 (5H, m, ArH), 7.00 (3H, t, $J = 7.6$ Hz, ArH), 7.12 (2H, d, $J = 7.6$ Hz, ArH), 7.31 (3H, t, $J = 7.6$ Hz, ArH), 7.38 (3H, t, $J = 8.0$ Hz,

ArH), 7.43–7.51 (4H, m,ArH), 7.67 (2H, d, $J = 7.6$ Hz, ArH), 7.84 (2H, d, $J = 7.6$ Hz, ArH); ^{13}C NMR (100 MHz, DMSO-d_6) (δ, ppm): 49.5, 54.8, 58.4, 126.4, 127.3, 128.0, 128.5, 129.2, 129.6, 130.4, 132.6, 133.2, 135.3, 135.5, 136.1, 137.4, 144.8, 196.9, 198.0; HRMS m/z calculated for $C_{35}H_{27}Br$ $[M + H]^+$: 527.1369, found: 527.1364.

*3-(4-Bromophenyl)-1,2,4,5-tetraphenylpentane-1,5-dione (**3f**)*

m.p. 240–242 °C; IR (KBr, v, cm^{-1}): 1680, 1596, 1579, 1490, 1446, 1267, 1178, 1076, 1008, 983, 819, 697, 681, 607, 543 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 4.82 (1H, t, $J = 10.0$ Hz, CH), 5.52 (1H, d, $J = 9.6$ Hz, CH), 5.60 (1H, d, $J = 10.8$ Hz, CH), 6.84–6.91 (4H, m, ArH), 6.99 (2H, t, $J = 7.6$ Hz, ArH), 7.14 (4H, dd, $J = 4.4$ Hz, $J = 8.8$ Hz, ArH), 7.31 (3H, t, $J = 7.6$ Hz, ArH), 7.37 (3H, t, $J = 7.6$ Hz,ArH), 7.42–7.50 (4H, m, ArH), 7.66 (2H, d, $J = 7.6$ Hz, ArH), 7.83 (2H, d, $J = 7.6$ Hz, ArH); ^{13}C NMR (100 MHz, DMSO-d_6) (δ, ppm): 50.5, 55.7, 58.4, 120.2, 127.2, 127.9, 128.2, 128.4, 128.5, 129.0, 130.2, 130.3, 130.6, 132.0, 132.7, 132.9, 136.4, 136.6, 136.7, 137.2, 139.5, 199.7, 199.8; HRMS m/z calculated for $C_{35}H_{27}Br$ $[M + Na]^+$: 549.1188, found: 549.1186.

*1,2,4,5-Tetraphenyl-3-(p-tolyl)pentane-1,5-dione (**3g**)*

m.p. 235–236 °C; IR (KBr, v, cm^{-1}): 1681, 1597, 1580, 1513, 1492, 1447, 1267, 1179, 1034, 1002, 982, 813, 755, 696, 616, 548 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$) (δ, ppm): 2.11 (3H, s, CH_3), 4.81 (1H, dd, $J = 3.6$ Hz, $J = 7.2$ Hz, CH), 5.02 (1H, d, $J = 7.2$ Hz, CH), 5.20 (1H, d, $J = 10.4$ Hz, CH), 6.78 (2H, d, $J = 7.6$ Hz, ArH), 6.94 (2H, d, $J = 8.0$ Hz, ArH), 7.01–7.08 (7H, m, ArH), 7.14 (2H, t, $J = 7.2$ Hz, ArH), 7.24 (3H, t, $J = 7.6$ Hz, ArH), 7.36 (2H, t, $J = 8.8$ Hz, ArH), 7.42 (2H, d, $J = 7.6$ Hz, ArH), 7.60 (2H, d, $J = 7.6$ Hz, ArH), 7.71 (2H, d, $J = 7.6$ Hz, ArH); ^{13}C NMR (100 MHz, DMSO-d_6) (δ, ppm): 54.9, 55.3, 55.4, 56.4, 110.6, 111.9, 113.2, 119.0, 120.1, 122.3, 126.3, 127.3, 127.9, 128.2, 128.6, 129.0, 129.2, 129.6, 131.0, 131.5, 134.1, 137.2, 137.7, 139.2, 147.2, 148.8, 152.2, 162.2, 165.0, 165.9, 171.3;HRMS m/z calculated for $C_{36}H_{30}$ $[M + H]^+$: 463.2420, found: 463.2413.

*3-(4-Methoxyphenyl)-1,2,4,5-tetraphenylpentane-1,5-dione (**3h**)*

m.p. 237–238 °C; IR (KBr, v, cm^{-1}): 1670, 1597, 1580, 1513, 1492, 1449, 1342, 1303, 1265, 1177, 1111, 1035, 1004, 984, 826, 755, 697, 613, 547 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$) (δ, ppm): 3.64 (3H, s, OCH_3), 4.79 (1H, dd, $J = 3.6$ Hz, $J = 7.2$ Hz, CH), 5.01 (1H, d, $J = 7.2$ Hz, CH), 5.18 (1H, d, $J = 10.4$ Hz, CH), 6.52 (2H, d, $J = 8.8$ Hz, ArH), 6.97 (2H, d, $J = 8.4$ Hz, ArH), 7.00–7.09 (6H, m, ArH), 7.15 (2H, t, $J = 7.2$ Hz, ArH), 7.22–7.26 (4H, m, ArH), 7.36 (2H, t, $J = 7.2$ Hz, ArH), 7.42 (2H, d, $J = 7.6$ Hz,ArH), 7.60 (2H, d, $J = 7.6$ Hz, ArH), 7.71 (2H, d, $J = 7.2$ Hz, ArH); ^{13}C NMR (100 MHz, DMSO-d_6) (δ, ppm): 49.3, 54.5, 55.2, 59.0, 112.6, 126.1, 127.2, 127.8, 128.0, 128.6, 129.7, 130.5, 132.5, 133.0, 133.9, 135.6, 136.2, 137.9, 156.7, 197.5, 198.1;HRMS m/z calculated for $C_{36}H_{30}O$ $[M + H]^+$: 479.2369, found: 479.2378.

*1,2,4,5-Tetraphenyl-3-(4-(pyridin-4-yl)phenyl)pentane-1,5-dione (**3i**)*

m.p. 220–222 °C; IR (KBr, *v*, cm^{-1}): 1672, 1597, 1578, 1559, 1491, 1449, 1267, 1173, 985, 762, 693, 606, 556, 540 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) (δ, ppm):4.83 (1H, t, $J = 10.4$ Hz, CH), 5.56 (1H, d, $J = 9.6$ Hz, CH), 5.66 (1H, d, $J = 10.8$ Hz, CH), 6.87–6.93 (4H, m, ArH), 6.99 (2H, t, $J = 7.6$ Hz, ArH), 7.15 (2H, d, $J = 7.6$ Hz, ArH), 7.30–7.40 (6H, m, ArH), 7.43–7.49 (4H, m, ArH), 7.67 (2H, d, $J = 7.6$ Hz, ArH), 7.84 (2H, d, $J = 7.6$ Hz, ArH), 8.14 (2H, d, $J = 5.2$ Hz, ArH); ^{13}C NMR (100 MHz, DMSO-d_6) (δ, ppm): 49.3, 54.5, 55.2, 59.0, 112.6, 126.1, 127.2, 127.8, 128.0, 128.6, 129.7, 130.5, 132.5, 133.0, 133.9, 135.6, 136.2, 137.9, 156.7, 197.5, 198.1; HRMS m/z calculated for $C_{34}H_{27}N$ $[M + H]^+$: 450.2216, found: 450.2209.

*3-(Benzo[d][1,3]dioxol-5-yl)-1,2,4,5-tetraphenylpentane-1,5-dione (**3j**)*

m.p. 231–233 °C; IR (KBr, *v*, cm^{-1}): 1669, 1557, 1519, 1505, 1443, 1370, 1316, 1240, 1198, 1039, 811, 769, 699 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) (δ, ppm): 4.24 (1H, d, $J = 13.6$ Hz, CH), 4.45 (1H, d, $J = 13.6$ Hz, CH), 5.30 (1H, s, CH), 6.03 (2H, s, OCH_2O), 6.84–6.91 (6H, m, ArH), 7.06–7.11 (5H, m, ArH), 7.24–7.27 (12H, m, ArH); ^{13}C NMR (100 MHz, DMSO-d_6) (δ, ppm): 49.8, 55.2, 58.5, 100.3, 101.2, 107.4, 108.4, 111.1, 120.6, 126.4, 127.2, 127.8, 128.0, 128.5, 129.0, 129.7, 130.6, 132.5, 133.0, 133.6, 135.5, 136.5, 137.3, 144.6, 146.3, 147.0, 147.7, 173.4, 197.3, 198.0; HRMS m/z calculated for $C_{36}H_{28}O_2$ $[M + Na]^+$: 515.1987, found: 515.1992.

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