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One-Pot Synthesis of Unsymmetrical Benzils by Oxidative Coupling Using Selenium Dioxide and *p*-Toluenesulfonic Acid Monohydrate

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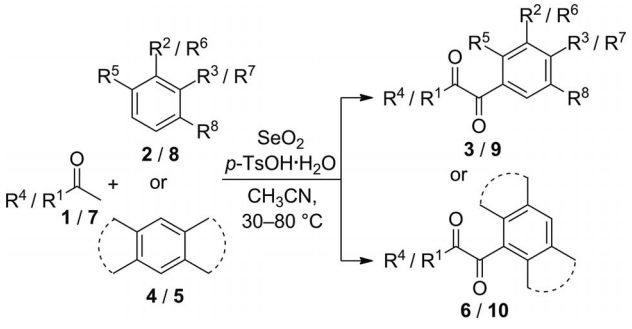
The oxidative coupling of the α-carbon atom of aromatic ketones with unactivated arenes in the presence of selenium dioxide and *p*-toluenesulfonic acid monohydrate is described. A number of unsymmetrical benzils have been prepared in good yields (38–75 %) with high regioselectivity. The generality and functional tolerance of this new protocol is demonstrated. The mechanistic pathway for the oxidative coupling reaction is also described. The reaction displays superiority in terms of minimization of steps with the C–C bond formation promoted by SeO_2 and pTsOH·H_2O. This method is advantageous as the reactants used as the solvent can be recovered.

Introduction

Synthetic studies of symmetrical, unsymmetrical, and heteroaryl 1,2-diketones have long been of interest as they are precursors for important ligands such as dioximes and dithiolenes in the field of discotic metallomesogens or polymeric mesogens and for the synthesis of heterocyclic compounds.[1,2] Some benzils exhibit the potential for biological activities, which includes the inhibition of mammalian carboxylesterases.[3] A number of synthetic pathways for the preparation of aryl/heteroaryl 1,2-diketones involve the oxidation of precursors such as olefins with selenium dioxide[4,5] or potassium permanganate,[6,7] methylene ketones by selenium dioxide,[8,9] bismuth nitrate–copper(II) acetate,[10] or oxygen in the presence of Fe^{III}–ethylenediaminetetraacetic acid.[11]

Recently, we conducted a literature survey of the synthetic utility of SeO_2 oxidation processes in synthetic chemistry, in particular, the use of SeO_2 in the presence of acids to promote new C–C bond formation to synthesize unsymmetrical benzils. Surprisingly, except for the work by Fuson et al., which relates to the intramolecular condensation of 2-biphenylyl-glyoxal to give phenanthrenequinone and the synthesis of mixed benzoins by the oxidation of acetophenones to glyoxals with a modification of Riley and Gray's method,[12,13] there was no report of one-pot oxidative coupling with SeO_2 for the synthesis of 1,2-diketones from acetophenones. As part of our continued efforts to develop new synthetic methodologies,[14] we carried out the reaction of acetophenone with SeO_2 in the presence of pTsOH·H_2O in benzene, which resulted in the formation of benzil **3a**. In this approach, an intermolecular C–C bond is formed between the α-carbon atom and the unactivated arene by the initial oxidation of the corresponding methyl aryl ketone with SeO_2 in the presence of pTsOH·H_2O. The overall attractiveness of preparing benzils in such a straightforward manner has motivated us to carry out a detailed study of this method. Furthermore, the advantage of this method also lies in the fact that the excess arenes used as reactants can be recovered, purified, and reused. Although the initial oxidation of aryl methyl ketones to glyoxal is well known,[15,16] we wish to report the first one-pot synthesis of benzils from the oxidative coupling of aryl methyl ketones and unactivated arenes using SeO_2 and pTsOH·H_2O (Scheme 1).



Scheme 1. Synthesis of unsymmetrical benzils by the oxidative coupling of aryl/heteroaryl methyl ketones with arenes.

This methodology proved to be versatile as hitherto unknown benzils were easily prepared. Of particular interest

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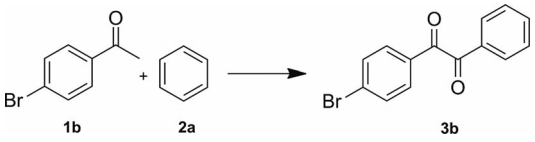
Table 1. Optimization of reaction conditions and screening of various oxidants and acids for the synthesis of **3b**.



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

Entry	Reagents [mmol] and conditions[a]	Yield [%][b]
1	SeO_2 (5.0), *p*TsOH·H_2O (2.5), 40 °C, 24 h	17
2	SeO_2 (5.0), *p*TsOH·H_2O (2.5), 40 °C, 4 h then at 80 °C, 12 h	60
3	SeO_2 (2.5), *p*TsOH·H_2O (2.5), 23–100 °C, 48 h	23
4	SeO_2 (5.0), 23–100 °C, 48 h	–
5	*p*TsOH·H_2O (5.0), 23–100 °C, 48 h	–
6	SeO_2 (5.0), CH_3COOH (2.5), 23–100 °C, 24 h	–
7	SeO2 (5.2), $AlCl_3$(2.5), 23–100 °C, 24 h	–
8	$KMnO_4$ (2.5), SeO_2 (2.5), *p*TsOH·H_2O (2.5), 23–100 °C, 48 h	5
9	$KMnO_4$ (5.0), SeO_2 (2.5) *p*TsOH·H_2O (2.5), 23–100 °C, 48 h	5

[a] Substrate **1b** (2.5 mmol), **2a** (8 mL). [b] Isolated yields.

is the coupling of acetophenones with polynuclear hydrocarbons, which results in the formation of a new series of unsymmetrical benzils.

Results and Discussion

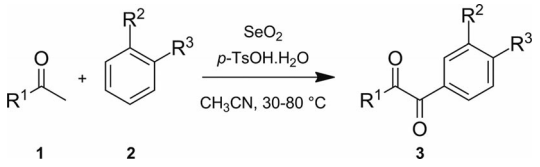
In a preliminary experiment, when a mixture of 1-(4-bromophenyl)ethanone (**1b**) (2.5 mmol), SeO_2 (5.0 mmol) and *p*TsOH·H_2O (2.5 mmol) in benzene (**2a**, 8 mL) was heated at 40 °C for 24 h, **3b** was obtained in very low yield. However, when the same reaction mixture was heated initially at 40 °C for 4 h and then at 80 °C for 12 h the yield significantly increased to 60% (Table 1, Entries 1–2). The formation of **3b** was confirmed by ^1H and ^{13}C NMR (two distinct carbonyl carbon signals) and IR spectroscopy.

In a control experiment, the use of either SeO_2 or *p*TsOH·H_2O alone failed to give the desired product (Table 1, Entries 4–5). When SeO_2 and *p*TsOH·H_2O were used in stoichiometric amounts (1 equiv. of each), **3b** was isolated in only 23% yield after 48 h, which showed that the optimum amount of SeO_2 is 2.0 equiv. (Table 1, Entry 3). The use of other acids instead of *p*TsOH·H_2O, e.g. acetic acid or a Lewis acid such as $AlCl_3$ in the presence of *p*TsOH·H_2O, under varying reaction conditions either gave no reaction or resulted in the formation of intractable products (Table 1, Entries 6–7).

To establish the unique properties of SeO_2 that affects the transformation, we carried out the reaction of acetophenones with various oxidants, such as potassium dichromate, vanadium pentoxide, and manganese dioxide, in the presence of *p*TsOH·H_2O and found that in all cases the desired product was not formed. However, potassium permanganate (1 equiv.) in the presence of SeO_2 (1 equiv.) and *p*TsOH·H_2O (1 equiv.) gave **3b** in 5% yield (Table 1, Entry 8). When the amount of potassium permanganate was increased (2 equiv.) in the presence of SeO_2 (1 equiv.) and *p*TsOH·H_2O (1 equiv.), no significant increase in the yield of the product was observed even with prolonged reaction times (Table 1, Entry 9).

Similarly, the oxidative coupling of **1a** and **1c** with **2a** under the optimized conditions proceeded smoothly to give **3a** and **3c** in 59 and 60% yields, respectively (Table 2, Entries 1 and 3). The generality of the method was further established by the reaction between substituted acetophenones, which contain electron withdrawing or donating groups, with arenes other than benzene and extended arenes. Irrespective of the nature of the substituents on the ring, all the reactions proceeded fairly well to afford unsymmetrical benzils in moderate to good yields (Table 2, Entries 4–9). The substituted acetophenones with a nitro or *N*-acetyl group in either *meta* or *para* positions appear to offer cleaner reactions and higher yields of the products without affecting the *N*-acetyl group (Table 2, Entries 3 and 6–8). Notably, the substrates **2b** and **2c** effectively provided **3d**–**h**

Table 2. Synthesis of substituted benzils by the oxidative coupling of substituted acetophenones **1** with arenes **2**.[a]



1 **2** **3**

Entry	Substrate **1** (R^1)	Substrate **2** (R^2, R^3)	Product **3**	Yield [%][b]
1	C_6H_5 (**1a**)	$R^2 = R^3 = H$ (**2a**)	**3a**[c]	59
2	4-BrC_6H_4 (**1b**)	**2a**	**3b**	58
3	4-$NO_2C_6H_4$ (**1c**)	**2a**	**3c**[c]	60
4	4-ClC_6H_4 (**1d**)	$R^2 = H$, $R^3 = CH_3$ (**2b**)	**3d**[c]	60[d]
5	**1b**	**2b**	**3e**	61[d]
6	**1c**	**2b**	**3f**	62[d]
7	3-$NO_2C_6H_4$ (**1e**)	$R^2 = R^3 = CH_3$ (**2c**)	**3g**	62
8	3-$NHCOCH_3C_6H_4$ (**1f**)	**2b**	**3h**	67
9	**1f**	$R^2 = H$, $R^3 = OCH_3$ (**2d**)	**3i**	70

[a] Reaction conditions: (i) **1** (2.5 mmol), SeO_2 (5.0 mmol), *p*TsOH·H_2O (2.5 mmol), **2a**–**2c** (6–8 mL), 30→80 °C, 16–20 h. (ii) **1** (2.5 mmol), SeO_2 (5.0 mmol), *p*TsOH·H_2O (2.5 mmol), **2d** (2.5 mmol), CH_3CN (6 mL), 35 °C, 14–16 h. [b] Isolated yields. [c] Commercially available. [d] Combined yield of the *ortho* and *para* isomers.

at 35 °C without the need to raise the temperature (Table 2, Entries 4–8). The excess arene substrates used in all the experiments were recovered and reused. In the case of substrate **2d**, acetonitrile was used as the solvent when it was found that stoichiometric amounts (1 equiv.) of the reactants at 35 °C were sufficient to furnish the corresponding 1,2-diketone (**3i**) in 70% yield (Table 2, Entry 9).

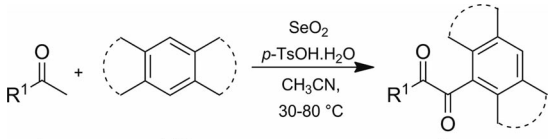
A striking feature of the reaction is the high regioselectivity, which is evident from the fact that only one regioisomer, **3g** and **3i**, was obtained when *ortho*-xylene (**2c**) and anisole (**2d**) were treated with **1e** and **1f**, respectively. The electrophilic substitution appears to be sensitive to steric hindrance. However, ^{1}H NMR spectroscopy revealed that toluene did show the formation of trace amounts of the *ortho* isomer with the *para* isomer predominating in a ratio of about 0.5:9.5 (Table 2, Entries 4–6).

Encouraged by these results, the scope of this oxidative coupling reaction was extended to the reaction of polynuclear hydrocarbons with aryl methyl ketones that bear a wide range of substituents (Table 3). Significantly, naphthalene (**4**) and anthracene (**5**) gave better yields of the unsymmetrical benzils **6** when reacted with acetophenones that contain electron-withdrawing substituents, such as chloro, bromo, and nitro groups, on the ring at 35 °C with acetonitrile as the solvent, compared to benzene and its derivatives. For example, **6d** was obtained in 70% yield when **4** was treated with **1c** (Table 3, Entry 4). Similarly, substituted acetophenones **1d**, **1b**, and **1c** reacted with **5** to give **6k**, **6l**, and **6p** in 68, 62, and 66% overall yields, respectively (Table 3, Entries 11, 12, and 16). The same acetophenones reacted with benzene and its derivatives to give **3b**–**f** in 58, 60, 60, 61, and 62% overall yields, respectively (Table 2, Entries 2–6). This may be attributed to the more reactive nature of the polynuclear hydrocarbons. As summarized in Table 3, of the two polynuclear hydrocarbons, **4** gave slightly better yields of products than **5** when reacted with aryl methyl ketones that bear substituents, such as hydroxy, nitro, and *N*-acetyl groups, on the *ortho*, *meta* or *para* positions. For example, **1h**, **1i**, **1c**, and **1f** reacted with **4** to give **6b**–**e** in 69–75% yield. Reaction of the same aryl methyl ketones with **5**, however, provided **6m**–**n** and **6p**–**q** in 50–57 and 66–73% yields, respectively (Table 3, Entries 2–5, 13–14, and 16–17). Similarly, methyl and methoxy substituted acetophenones afforded moderate to good yields when subjected to the same reaction conditions with **4** and **5** (Table 3, Entries 1, 6–9, and 18–19).

Only the products substituted at C1 and C9 were isolated from reactions with **4** and **5**, respectively (Table 3). The presence of other isomers was not observed. Disappointingly, the reactions of chlorobenzene, bromobenzene, and nitrobenzene with various acetophenones with this protocol did not provide any of the isomers, even under different reaction conditions. This is thought to be due to the ring deactivating effect of the halide and the nitro group.

The broad scope of this procedure was further demonstrated by the reactions of fused aromatic methyl or heteroaryl methyl ketones with arenes to give the corresponding 1,2-diketones (Table 4). The same reaction trend was

Table 3. Synthesis of unsymmetrical benzils **6** by the oxidative coupling of substituted acetophenones **1** with **4** and **5**.[a]

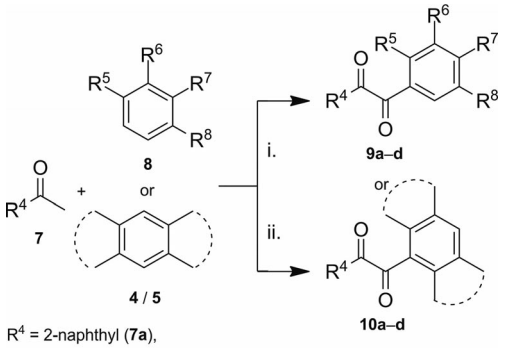


1 **4 / 5** **6a–s**

Entry	Substrate **1** (R^1)	Substrate **4** or **5**	Product **6**	Yield [%][b]
1	3-CH_3O-C_6H_4 (**1g**)	**4**	**6a**	65
2	2-OH-C_6H_4 (**1h**)	**4**	**6b**	69
3	4-OH-C_6H_4 (**1i**)	**4**	**6c**	67
4	**1c**	**4**	**6d**	70
5	**1f**	**4**	**6e**	75
6	2,4-$(CH_3)_2$-C_6H_3 (**1j**)	**4**	**6f**	62
7	2-CH_3-C_6H_4 (**1k**)	**5**	**6g**	60
8	4-CH_3-C_6H_4 (**1l**)	**5**	**6h**	57
9	**1g**	**5**	**6i**	55
10	2-Cl-C_6H_4 (**1m**)	**5**	**6j**	63
11	**1d**	**5**	**6k**	68
12	**1b**	**5**	**6l**	62
13	**1h**	**5**	**6m**	50
14	**1i**	**5**	**6n**	57
15	**1e**	**5**	**6o**	64
16	**1c**	**5**	**6p**	66
17	**1f**	**5**	**6q**	73
18	**1j**	**5**	**6r**	58
19	2,4,6-$(CH_3)_3$-C_6H_2 (**1n**)	**5**	**6s**	53

[a] Reaction conditions: (i) **1** (2.5 mmol), SeO_2 (5.0 mmol), *p*TsOH·H_2O (2.5 mmol), **4**/**5** (2.5 mmol), CH_3CN (6 mL), 35 °C, 12–16 h. [b] Isolated yields.

Table 4. Synthesis of unsymmetrical benzils by the oxidative coupling of extended/hetero aryl ketones **7** with arenes **8**, **4**, and **5**.[a]

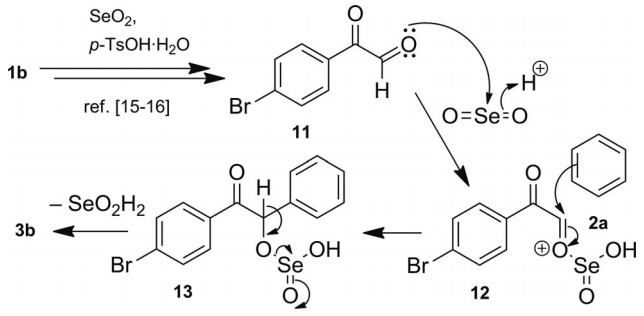


Entry	Substrate **7** (R^4)	Substrate **8** (R^5, R^6, R^7, R^8), **4** or **5**	Product **9**/**10**	Yield [%][b]
1	2-naphthyl (**7a**)	**2a** (**8a**)	**9a**	59
2	**7a**	**2b** (**8b**)	**9b**	51[c]
3	**7a**	R^6 = R^7 = H, R^5 = R^8 = CH_3 (**8c**)	**9c**	61
4	**7a**	**2d** (**8d**)	**9d**	70
5	**7a**	**4**	**10a**	69
6	2-furanyl (**7b**)	**4**	**10b**	38
7	2-thiophenyl (**7c**)	**4**	**10c**	57
8	**7c**	**5**	**10d**	46

[a] The reaction conditions are same those used for the reactions in Tables 2 and 3. For substrate **8c**, reaction conditions (i) were used. [b] Isolated yields. [c] Combined yield of the *ortho* and *para* isomers.

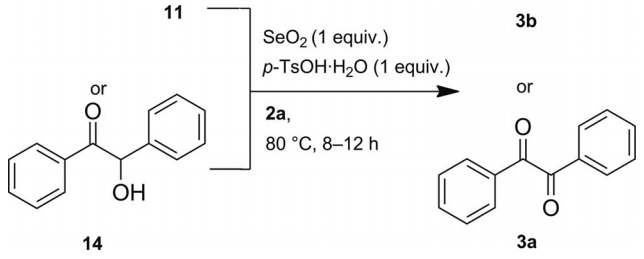
observed when the method was applied to the oxidative coupling of 1-(naphthalen-2-yl)ethanone (**7a**), 1-(furan-2-yl)ethanone (**7b**), and 1-(thiophen-2-yl)ethanone (**7c**) with arenes **4**, **5**, and **8**, which gave consistently moderate to good yields of **9** and **10** (Table 4, Entries 1–8).

A plausible mechanism is shown in Scheme 2. The oxidation of **1b** to glyoxal (**11**)[15] (isolated, m.p. 50–51 °C; lit. 51–52 °C)[13] by SeO_2 is followed by the preferential formation of an O–Se bond through the carbonyl oxygen atom of the aldehyde group in the presence of pTsOH·H_2O to give the intermediate **12**. The activating effect of the keto group and the formation of the O–Se bond generate a strong electrophilic center at the aldehyde carbon atom of **12**, which is highly susceptible to attack from electron-rich arenes to give the selenite intermediate **13**.[16,17] Oxidative decomposition of **13** led to **3b**.



Scheme 2. Proposed mechanism for the one-pot synthesis of unsymmetrical benzils.

The proposed mechanism was supported by the reactions of 2-(4-bromophenyl)-2-oxoacetaldehyde (**11**) and 2-hydroxy-1,2-diphenylethanone (**14**) with **2a** to give **3a** and **3b** in good yields (Scheme 3). The concerted reaction of glyoxals and arenes with SeO_2 as the active oxidant appears to be the major mechanistic pathway for these reactions.



Scheme 3. Reactions of **11** and **14** with **2a**.

Conclusions

We have reported a direct and efficient protocol for the preparation of unsymmetrical and heteroaryl 1,2-diketones through oxidative coupling between the α-carbon atom of the aromatic ketone with unactivated arenes in the presence of SeO_2 and pTsOH·H_2O. The mechanistic pathway for the oxidative coupling reaction has been described. The reaction displays superiority in terms of minimization of steps with a C–C bond formation promoted by SeO_2 and pTsOH·H_2O. The method is regioselective and is also advantageous as the reactants used as solvent can be recovered.

Experimental Section

General: All reactions were carried out in oven-dried glassware at 30–80 °C. The following reagents were purchased and used without purification: p-toluene sulfonic acid (MERCK), acetophenone, 1-(4-chlorophenyl)ethanone, 1-(4-bromophenyl)ethanone, 1-(3-nitrophenyl)ethanone, 1-(4-nitrophenyl)ethanone, *N*-(3-acetylphenyl)-acetamide, 1-(3-methoxyphenyl)ethanone, 1-(2-hydroxyphenyl)ethanone, 1-(4-hydroxyphenyl)ethanone, 1-(2,4-dimethylphenyl)ethanone, 1-(naphthalen-2-yl)ethanone, 1-(thiophen-2-yl)ethanone, 1-(furan-2-yl)ethanone, naphthalene, anthracene (Aldrich), and anisole (Aldrich). The following reagents were purified before use: SeO_2 (Aldrich), benzene, toluene, o-xylene, and p-xylene.

^{1}H NMR and ^{13}C NMR analyses were recorded with a 400 MHz spectrometer and are reported in parts per million from tetramethylsilane with the solvent resonance as the internal standard. Data are reported as follows: chemical shift, multiplicity (s = singlet, d = doublet, dd = double doublet, t = triplet, m = multiplet and br. = broad). Mass spectra were performed with a GC–MS, LC–MS, and mass spectrometer equipped with ESI and APCI mass detector. IR analyses were carried out with an FT-FIR spectrometer. Melting points were recorded with the open capillary tube method. Elemental analyses were carried out with a CHN analyzer.

General Experimental Procedure for the Preparation of Aryl/Heteroaryl Ethane-1,2-dione. Method A: To a stirring mixture of acetophenone (2.5 mmol) and resublimed SeO_2 (5.0 mmol) in benzene (8 mL) was added pTsOH·H_2O (2.5 mmol) in one portion at 30 °C. The reaction mixture was allowed to stir at 40 °C for 4 h and then gradually heated to 80 °C. The reaction mixture was stirred at 80 °C for 16–20 h. The reaction mixture was concentrated under reduced pressure at 40 °C, and the residue was diluted with ethyl acetate and filtered through celite. The celite was washed with ethyl acetate (3 × 5 mL). The combined filtrate was washed with saturated aqueous sodium hydrogen carbonate solution, water (10 mL), and brine (10 mL). The organic layer was separated, dried with anhydrous Na_2SO_4 and concentrated. The benzil was purified by column chromatography on silica gel (100–200 mesh) using ethyl acetate and hexane as eluent.

Method B: The procedure is the same as Method A except for reactants such as toluene, o-xylene, and p-xylene the reactions were performed at 35 °C for 12–16 h and for reactants such as anisole, naphthalene, and anthracene the reactions were performed at 35 °C for 12–16 h in acetonitrile (6 mL).

Benzil (3a): Prepared from acetophenone (300.4 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), pTsOH·H_2O (475.6 mg, 2.5 mmol), and benzene (8 mL) over 20 h following Method A. Eluent: ethyl acetate 3% in hexane; yellow solid (310.1 mg, 59%). The ^{1}H NMR spectrum matched that of a commercial sample.

1-(4-Bromophenyl)-2-phenylethane-1,2-dione (3b): Prepared from 1-(4-bromophenyl)ethanone (497.6 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), pTsOH·H_2O (475.6 mg, 2.5 mmol), and benzene (8 mL) over 20 h following Method A. Eluent: ethyl acetate 5% in hexane; light yellow solid (419.2 mg, 58%); m.p. 71–73 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.90–7.89 (m, 2 H), 7.79–7.77 (m, 2 H), 7.63–7.59 (m, 3 H), 7.48–7.44 (m, 2 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.9, 193.3, 135.1, 132.7, 132.5, 131.7, 131.3, 130.5,

130.0, 129.1 ppm. IR (KBr film): ṽ = 3089, 3064, 2959, 2924, 2852, 1669, 1582, 1569, 1483, 1450, 1399, 1321, 1301, 1261, 1211, 1174, 1071, 1011 cm^{-1}. $C_{14}H_9BrO_2$ (289.13): calcd. C 58.16, H 3.14; found C 58.23, H 3.09.

1-(4-Nitrophenyl)-2-phenylethane-1,2-dione (3c): Prepared from 1-(4-nitrophenyl)ethanone (412.9 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and benzene (8 mL) over 20 h following Method A. Eluent: ethyl acetate 15% in hexane; yellow solid (382.8 mg, 60%); m.p. 140–142 °C. The ^1H NMR spectrum matched that of a commercial sample. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.37 (m, 2 H), 8.18 (m, 2 H), 8.00–7.99 (m, 2 H), 7.74–7.70 (m, 1 H), 7.58–7.54 (m, 2 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 192.9, 192.1, 151.1, 137.3, 135.5, 132.4, 131.0, 130.1, 129.2, 124.1 ppm. IR (KBr film): ṽ = 3108, 3082, 2925, 2853, 1676, 1663, 1604, 1596, 1528, 1451, 1410, 1348, 1324, 1207, 1182, 1173 cm^{-1}. $C_{14}H_9NO_4$ (255.23): calcd. C 65.88, H 3.55, N 5.49; found C 65.97, H 3.50, N 5.50.

1-(4-Chlorophenyl)-2-(*p*-tolyl)ethane-1,2-dione (3d): Prepared from 1-(4-chlorophenyl)ethanone (386.5 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and toluene (6 mL) over 15 h following Method B. Eluent: ethyl acetate 10% in hexane; yellowish solid (388.1 mg, 60%); m.p. 121–123 °C. The ^1H NMR spectrum matched that of a commercial sample. ^1H NMR (400 MHz, $CDCl_3$): δ = 7.92–7.91 (m, 2 H), 7.87–7.85 (m, 2 H), 7.50–7.84 (m, 2 H), 7.33–7.31 (m, 2 H), 2.44 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.7, 193.3, 146.5, 141.5, 131.4, 131.2, 130.3, 130.1, 129.8, 129.4, 22.0 ppm. IR (KBr film): ṽ = 3093, 3066, 2924, 2854, 1663, 1605, 1587, 1573, 1486, 1457, 1402, 1319, 1215, 1185, 1174 cm^{-1}. GC–MS: *m*/*z* (%) = 258 (8) $[M]^+$, 139 (72), 119 (100), 111 (69), 91 (98). $C_{15}H_{11}ClO_2$ (258.70): calcd. C 69.64, H 4.29; found C 69.57, H 4.31.

1-(4-Bromophenyl)-2-(*p*-tolyl)ethane-1,2-dione (3e): Prepared from 1-(4-bromophenyl)ethanone (497.6 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and toluene (6 mL) over 16 h following Method B. Eluent: ethyl acetate 8% in hexane; yellowish solid (462.3 mg, 61%); m.p. 92–94 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 7.88–7.68 (m, 6 H), 7.33–7.31 (m, 2 H), 2.45 (s, 3 H) ppm.^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.6, 193.5, 146.5, 134.0, 132.4, 131.8, 131.2, 130.4, 130.1, 129.8, 22.0 ppm. IR (KBr film): ṽ = 3090, 3061, 3031, 2925, 2854, 1665, 1605, 1584, 1483, 1458, 1400, 1319, 1310, 1206, 1174, 1069 cm^{-1}. MS (ES^+): *m*/*z* = 325.0 $[M + Na]^+$. $C_{15}H_{11}BrO_2$ (303.15): calcd. C 59.43, H 3.66; found C 59.57, H 3.63.

1-(4-Nitrophenyl)-2-(*p*-tolyl)ethane-1,2-dione (3f): Prepared from 1-(4-nitrophenyl)ethanone (412.9 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and toluene (6 mL) over 16 h following Method B. Eluent: ethyl acetate 15% in hexane; light yellow solid (417.4 mg, 62%); m.p. 145–147 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.37–8.16 (m, 4 H), 7.89–7.88 (m, 2 H), 7.36–7.34 (m, 2 H), 2.47 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 192.6, 192.3, 147.0, 137.4, 134.4, 132.8, 131.0, 130.2, 130.0, 124.1, 22.0 ppm. IR (KBr film): ṽ = 3106, 3080, 3052, 2924, 2854, 1677, 1661, 1603, 1570, 1525, 1411, 1348, 1323, 1207, 1184, 1174 cm^{-1}. GC–MS: *m*/*z* (%) = 269 (15) $[M]^+$, 239 (19), 150 (27), 119 (100), 104 (26), 91 (94). $C_{15}H_{11}NO_4$ (269.26): calcd. C 66.91, H 4.12, N 5.20; found C 66.86, H 4.22, N 5.26.

1-(3,4-Dimethylphenyl)-2-(3-nitrophenyl)ethane-1,2-dione (3g): Prepared from 1-(3-nitrophenyl)ethanone (412.9 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and *o*-xylene (6 mL) over 16 h following Method B. Eluent: ethyl acetate 20% in hexane; yellow solid (439.2 mg, 62%); m.p. 105–107 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.81 (s, 1 H), 8.50 (dd, *J* = 1.6, *J* = 7.6 Hz, 1 H), 8.31 (d, *J* = 7.6 Hz, 1 H), 7.77–7.72 (m, 3 H) 7.30 (d, *J* = 8.0 Hz, 1 H), 2.37 (s, 3 H), 2.34 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 192.2, 191.3, 148.1, 145.4, 137.4, 135.3, 134.8, 134.0, 130.4, 130.0, 129.8, 128.2, 127.5, 124.1, 19.9, 19.3 ppm. IR (KBr film): ṽ = 3082, 2951, 2920, 2870, 1682, 1655, 1607, 1578, 1569, 1534, 1437, 1350, 1279, 1232, 1208, 1173, 1135, 1090 cm^{-1}. LC–MS: *m*/*z* = 284.0 $[M + H]^+$. $C_{16}H_{13}NO_4$ (283.28): calcd. C 67.84, H 4.63, N 4.94; found C 67.79, H 4.68, N 4.96.

***N*-[3-{2-Oxo-2-(*p*-tolyl)acetyl}phenyl]acetamide (3h):** Prepared from *N*-(3-acetylphenyl)acetamide (443.0 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and toluene (6 mL) over 16 h following Method B. Eluent: ethyl acetate 25% in hexane; light yellow solid (471.2 mg, 67%); m.p. 117–119 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 7.99 (d, *J* = 7.6 Hz, 1 H), 7.93–7.86 (m, 2 H), 7.76–7.74 (m, 2 H), 7.56 (d, *J* = 7.6 Hz, 1 H), 7.38–7.34 (m, 1 H), 7.22 (m, 2 H), 2.35 (s, 3 H), 2.06 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.5, 193.2, 167.9, 145.5, 137.9, 133.0, 132.1, 131.6, 129.0, 128.8 125.4, 124.7, 119.2, 23.4, 21.0 ppm. IR (KBr film): ṽ = 3287, 3200, 3145, 3119, 3103, 2925, 2857, 2828, 1685, 1665, 1606, 1594, 1571, 1488, 1441, 1372, 1330, 1308, 1290, 1270, 1246, 1180, 1159 cm^{-1}. GC–MS: *m*/*z* (%) = 281 (9) $[M]^+$, 239 (12), 162 (60), 134 (22), 119 (100), 91 (98). $C_{17}H_{15}NO_3$ (281.31): calcd. C 72.58, H 5.37, N 4.98; found C 72.81, H 5.29, N 4.89.

***N*-[3-{2-(4-Methoxyphenyl)-2-oxoacetyl}phenyl]acetamide (3i):** Prepared from *N*-(3-acetylphenyl)acetamide (443.0 mg, 2.5 mmol), anisole (270.4 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 14 h following Method B. Eluent: ethyl acetate 30% in hexane; yellowish solid (520.3 mg, 70%); m.p. 130–132 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.00–8.82 (m, 5 H), 7.58 (d, *J* = 7.2 Hz, 1 H), 7.37 (t, *J* = 7.6 Hz, 1 H), 6.89–6.87 (m, 2 H), 3.81 (s, 3 H), 2.07 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 194.6, 193.1, 169.0, 165.2, 138.8, 133.6, 132.4, 129.8, 126.4, 125.8, 125.7, 120.4, 114.5, 55.7, 24.5 ppm. IR (KBr film): ṽ = 3289, 3199, 3145, 3103, 2924, 2851, 1684, 1668, 1654, 1599, 1571, 1509, 1487, 1441, 1425, 1310, 1261, 1246, 1177, 1159, 1021 cm^{-1}. MS (ES^+): *m*/*z* = 298.0 $[M + H]^+$, 320.0 $[M + Na]^+$. $C_{17}H_{15}NO_4$ (297.31): calcd. C 68.68, H 5.09, N 4.71; found C 68.56, H 5.13, N 4.79.

1-(3-Methoxyphenyl)-2-(naphthalen-1-yl)ethane-1,2-dione (6a): Prepared from 1-(3-methoxyphenyl)ethanone (375.4 mg, 2.5 mmol), naphthalene (320.4 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 15 h following Method B. Eluent: ethyl acetate 15% in hexane; yellow solid (471.7 mg, 65%); m.p. 93–95 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 9.30 (d, *J* = 8.8 Hz, 1 H), 8.12 (d, *J* = 8.0 Hz, 1 H), 7.95–7.89 (m, 2 H), 7.75 (t, *J* = 7.6 Hz, 1 H), 7.65–7.60 (m, 2 H),7.55–7.47 (m, 2 H), 7.40 (t, *J* = 8.0 Hz, 1 H), 7.20 (dd, *J* = 2.4, *J* = 8.4 Hz, 1 H), 3.86 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 197.0, 194.5, 160.1, 135.9, 135.0, 134.7, 134.1, 131.0,130.1, 129.5, 128.8, 128.7, 127.1, 126.0, 124.4, 123.3, 121.8, 113.0, 55.6 ppm. IR (KBr film): ṽ = 3070, 3012, 2964, 2940, 2922, 2837, 1672, 1655, 1594, 1572, 1509, 1485, 1465, 1431, 1339, 1299, 1268, 1245, 1211, 1172, 1161, 1104 cm^{-1}. GC–MS: *m*/*z* (%) = 290 (22) $[M]^+$, 155 (100), 135 (86), 127 (98), 107 (39), 101 (20), 92 (58). $C_{19}H_{14}O_3$ (290.32): calcd. C 78.61, H 4.86; found C 78.51, H 4.89.

1-(2-Hydroxyphenyl)-2-(naphthalen-1-yl)ethane-1,2-dione (6b): Prepared from 1-(2-hydroxyphenyl)ethanone (340.4 mg, 2.5 mmol), naphthalene (320.4 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 15 h following Method B. Eluent: ethyl acetate 30% in hexane; yellow solid (476.6 mg, 69%); m.p. 120–122 °C. ^1H NMR (400 MHz,

$CDCl_3$): δ = 11.48 (s, 1 H), 9.32 (d, *J* = 8.4 Hz, 1 H), 8.16 (d, *J* = 8.0 Hz, 1 H), 7.97–7.93 (m, 2 H), 7.77 (t, *J* = 7.6 Hz, 1 H), 7.67–7.50 (m, 4 H), 7.11 (d, *J* = 8.4 Hz, 1 H), 6.89 (t, *J* = 7.6 Hz, 1 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 199.4, 194.4, 163.4, 137.9, 136.4, 135.5, 134.1, 132.5, 131.0, 129.7, 128.9, 128.1, 127.3, 125.9, 124.4, 119.8, 118.4, 117.1 ppm. IR (KBr film): ṽ = 3241, 3174, 3087, 3062, 3010, 2957, 2924, 2852, 1660, 1631, 1591, 1575, 1509, 1486, 1454, 1397, 1309, 1251, 1230, 1182, 1156, 1100, 1084 cm^{-1}. $C_{18}H_{12}O_3$ (276.29): calcd. C 78.25, H 4.38; found C 78.33, H 4.42.

1-(4-Hydroxyphenyl)-2-(naphthalen-1-yl)ethane-1,2-dione (6c): Prepared from 1-(4-hydroxyphenyl)ethanone (340.4 mg, 2.5 mmol), naphthalene (320.4 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 15 h following Method B. Eluent: ethyl acetate 35% in hexane; yellow solid (462.8 mg, 67%); m.p. 115–117 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.28 (d, *J* = 8.0 Hz, 1 H), 8.12 (d, *J* = 8.0 Hz, 1 H), 7.95–7.89 (m, 4 H), 7.74 (t, *J* = 8.0 Hz, 1 H), 7.63 (t, *J* = 7.6 Hz, 1 H), 7.49 (t, *J* = 7.6 Hz, 1 H), 6.89–6.87 (m, 2 H), 5.31 (s, 1 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 197.6, 193.5, 162.0, 136.0, 135.1, 134.1, 132.8, 130.9, 129.4, 128.8, 128.7, 127.1, 126.2, 125.9, 124.4, 116.0 ppm. IR (KBr film): ṽ = 3280, 3056, 2962, 2821, 2681, 1663, 1645, 1598, 1566, 1510, 1462, 1437, 1397, 1371, 1317, 1296, 1264, 1225, 1185, 1165, 1148, 1108, 1086 cm^{-1}. $C_{18}H_{12}O_3$ (276.29): calcd. C 78.25, H 4.38; found C 78.29, H 4.46.

1-(Naphthalen-1-yl)-2-(4-nitrophenyl)ethane-1,2-dione (6d): Prepared from 1-(4-nitrophenyl)ethanone (412.9 mg, 2.5 mmol), naphthalene (320.4 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 15 h following Method B. Eluent: ethyl acetate 30% in hexane; yellow solid (534.3 mg, 70%); m.p. 150–152 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.20 (d, *J* = 8.4 Hz, 1 H), 8.31–8.29 (m, 2 H), 8.15–8.10 (m, 3 H), 7.91 (d, *J* = 8.4 Hz, 1 H), 7.82 (d, *J* = 7.2 Hz, 1 H), 7.71 (t, *J* = 8.0 Hz, 1 H), 7.60 (t, *J* = 7.6 Hz, 1 H), 7.46 (t, *J* = 7.6 Hz, 1 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.4, 192.2, 151.1, 137.7, 136.7, 135.3, 134.1, 131.0, 130.9, 129.8, 129.0, 128.0, 127.4, 125.8, 124.4, 124.2 ppm. IR (KBr film): ṽ = 3108, 3087, 3075, 3050, 2920, 2853, 1674, 1651, 1602, 1572, 1526, 1511, 1466, 1436, 1399, 1371, 1345, 1322, 1307, 1262, 1220, 1179, 1110 cm^{-1}. GC–MS: *m*/*z* (%) = 305 (6) [M]$^+$, 155 (98), 150 (100), 127 (99), 92 (44). $C_{18}H_{11}NO_4$ (305.29): calcd. C 70.82, H 3.63, N 4.59; found C 70.67, H 3.72, N 4.64.

***N*-[3-{2-(Naphthalen-1-yl)-2-oxoacetyl}phenyl]acetamide (6e):** Prepared from *N*-(3-acetylphenyl)acetamide (443.0 mg, 2.5 mmol), naphthalene (320.4 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 15 h following Method B. Eluent: ethyl acetate 25% in hexane; yellow solid (595.0 mg, 75%); m.p. 145–147 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.27 (d, *J* = 8.8 Hz, 1 H), 8.13–8.07 (m, 2 H), 7.94–7.87 (m, 3 H), 7.75–7.60 (m, 4 H), 7.50–7.43 (m, 2 H), 2.09 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 196.9, 194.2, 168.7, 138.8, 136.1, 135.2, 134.1, 133.9, 130.9, 129.9, 129.5, 128.8, 128.4, 127.2, 126.3, 125.9, 125.8, 124.4, 120.4, 24.5 ppm. IR (KBr film): ṽ = 3292, 3251, 3186, 3135, 3090, 2926, 2856, 1678, 1665, 1600, 1558, 1509, 1487, 1412, 1371, 1326, 1304, 1269, 1236, 1202, 1176, 1162, 1109 cm^{-1}. MS (ES$^+$): *m*/*z* = 317.9 [M + H]$^+$, 340 [M + Na]$^+$. $C_{20}H_{15}NO_3$ (317.34): calcd. C 75.70, H 4.76, N 4.41; found C 75.76, H 4.68, N 4.39.

1-(2,4-Dimethylphenyl)-2-(naphthalen-1-yl)ethane-1,2-dione (6f): Prepared from 1-(2,4-dimethylphenyl)ethanone (370.5 mg, 2.5 mmol), naphthalene (320.4 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 15 h following Method B. Eluent: ethyl acetate 10% in hexane; yellow solid (446.9 mg, 62%); m.p. 87–89 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.30 (d, *J* = 8.8 Hz, 1 H), 8.11 (d, *J* = 8.0 Hz, 1 H), 7.95–7.92 (m, 2 H), 7.73 (t, *J* = 7.6 Hz, 1 H), 7.65–7.60 (m, 2 H), 7.49 (t, *J* = 7.6 Hz, 1 H), 7.17 (s, 1 H), 7.06 (d, *J* = 8.0 Hz, 1 H), 2.73 (s, 3 H), 2.38 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 197.6, 196.3, 144.8, 141.9, 135.6, 134.8, 134.1, 133.6, 133.5, 131.1, 129.4, 129.3, 128.9, 128.7, 127.0, 126.8, 126.0, 124.4, 22.0, 21.7 ppm. IR (KBr film): ṽ = 3089, 3028, 2963, 2924, 2854, 1663, 1611, 1571, 1508, 1451, 1436, 1378, 1314, 1221, 1214, 1181, 1148, 1100 cm^{-1}. GC–MS: *m*/*z* (%) = 288 (25) [M]$^+$, 155 (90), 133 (100), 127 (98), 105 (76), 89 (36). $C_{20}H_{16}O_2$ (288.35): calcd. C 83.31, H 5.59; found C 83.23, H 5.61.

1-(Anthracen-9-yl)-2-(*o*-tolyl)ethane-1,2-dione (6g): Prepared from 1-(*o*-tolyl)ethanone (335.5 mg, 2.5 mmol), anthracene (445.6 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 14 h following Method B. Eluent: ethyl acetate 5% in hexane; yellow solid (486.6 mg, 60%); m.p. 128–130 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.55 (s, 1 H), 8.20 (d, *J* = 7.6 Hz, 1 H), 8.03–7.97 (m, 4 H), 7.51–7.31 (m, 7 H), 2.54 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 197.3, 192.5, 141.2, 132.5, 131.8, 131.7, 130.8, 129.9, 129.9, 129.4, 129.1, 128.1, 126.8, 124.9, 124.5, 123.4, 20.9 ppm. IR (KBr film): ṽ = 3053, 3029, 2979, 2929, 1660, 1600, 1556, 1486, 1452, 1382, 1305, 1249, 1179, 1100 cm^{-1}. MS (ES$^+$): *m*/*z* = 347.0 [M + Na]$^+$. $C_{23}H_{16}O_2$ (324.38): calcd. C 85.16, H 4.97; found C 85.09, H 5.11.

1-(Anthracen-9-yl)-2-(*p*-tolyl)ethane-1,2-dione (6h): Prepared from 1-(*p*-tolyl)ethanone (335.4 mg, 2.5 mmol), anthracene (445.6 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 15 h following Method B. Eluent: ethyl acetate 5% in hexane; light yellow solid (462.3 mg, 57%); m.p. 138–140 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.55 (s, 1 H), 8.16–8.14 (m, 2 H), 7.99–7.95 (m, 4 H), 7.44–7.34 (m, 6 H), 2.44 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 198.5, 191.0, 146.1, 134.1, 131.8, 131.0, 130.9, 130.0, 130.0, 129.1, 127.8, 127.2, 125.6, 124.5, 22.0 ppm. IR (KBr film): ṽ = 3045, 2923, 2850, 1672, 1661, 1601, 1445, 1411, 1333, 1301, 1285, 1259, 1160, 1112 cm^{-1}. MS (ES$^+$): *m*/*z* = 347.1 [M + Na]$^+$. $C_{23}H_{16}O_2$ (324.38): calcd. C 85.16, H 4.97; found C 85.20, H 4.90.

1-(Anthracen-9-yl)-2-(3-methoxyphenyl)ethane-1,2-dione (6i): Prepared from 1-(3-methoxyphenyl)ethanone (375.4 mg, 2.5 mmol), anthracene (445.6 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 14 h following Method B. Eluent: ethyl acetate 5% in hexane; light yellow solid (468.1 mg, 55%); m.p. 123–125 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.52 (s, 1 H), 7.98–7.94 (m, 4 H), 7.85 (d, *J* = 7.6 Hz, 1 H), 7.68–7.67 (m, 1 H), 7.44–7.38 (m, 5 H), 7.18 (dd, *J* = 2.0, *J* = 8.4 Hz, 1 H), 3.77 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 198.1, 191.2, 160.1, 134.1, 132.0, 131.0, 130.2, 130.1, 129.2, 128.0, 127.2, 125.6, 124.4, 123.7, 121.8, 113.9, 55.5 ppm. IR (KBr film): ṽ = 3073, 3012, 2971, 2933, 2832, 1679, 1665, 1593, 1482, 1462, 1452, 1422, 1332, 1274, 1262, 1229, 1156, 1111 cm^{-1}. MS (ES$^+$): *m*/*z* = 363.0 [M + Na]$^+$. $C_{23}H_{16}O_3$ (340.38): calcd. C 81.16, H 4.74; found C 81.29, H 4.89.

1-(Anthracen-9-yl)-2-(2-chlorophenyl)ethane-1,2-dione (6j): Prepared from 1-(2-chlorophenyl)ethanone (386.5 mg, 2.5 mmol), anthracene (445.6 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 14 h following Method B. Eluent: ethyl acetate 5% in hexane; yellow solid (543.1 mg, 63%); m.p. 138–140 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.52 (s, 1 H), 8.22 (d, *J* = 7.6 Hz, 1 H), 7.95–7.94 (m, 4 H), 7.66–7.372 (m, 7 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 198.3, 191.2, 134.8, 132.2, 132.0, 131.0, 130.7, 130.3, 130.1, 129.2,

129.2, 128.0, 125.6, 124.4 ppm. IR (KBr film): $\tilde{\nu}$ = 3057, 2924, 2852, 1663, 1622, 1577, 1555, 1487, 1452, 1303, 1247, 1103 cm^{-1}. MS (ES^+): *m/z* = 345.1 $[M + H]^+$, 366.9 $[M + Na]^+$. $C_{22}H_{13}ClO_2$ (344.80): calcd. C 76.64, H 3.80; found C 76.48, H 3.96.

1-(Anthracen-9-yl)-2-(4-chlorophenyl)ethane-1,2-dione (6k): Prepared from 1-(4-chlorophenyl)ethanone (386.5 mg, 2.5 mmol), anthracene (445.6 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 15 h following Method B. Eluent: ethyl acetate 5% in hexane; yellow solid (586.2 mg, 68%); m.p. 165–167 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.55 (s, 1 H), 8.20–8.19 (m, 2 H), 7.99–7.89 (m, 4 H), 7.53–7.51 (m, 2 H), 7.42–7.40 (m, 4 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 196.8, 188.8, 140.5, 131.1, 131.0, 129.9, 129.5, 129.0,128.9, 128.6, 128.2, 127.0, 124.6, 123.2 ppm. IR (KBr film): $\tilde{\nu}$ = 3046, 2926, 2851, 1672, 1657, 1622, 1584, 1568, 1401, 1252 cm^{-1}. MS (ES^+): *m/z* = 345.1 $[M + H]^+$, 366.9 $[M + Na]^+$. $C_{22}H_{13}ClO_2$ (344.80): calcd. C 76.64, H 3.80; found C 76.58, H 3.92.

1-(Anthracen-9-yl)-2-(4-bromophenyl)ethane-1,2-dione (6l): Prepared from 1-(4-bromophenyl)ethanone (497.6 mg, 2.5 mmol), anthracene (554.8 g, 5.0 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 12 h following Method B. Eluent: ethyl acetate 10% in hexane; light yellow solid (603.0 mg, 62%); m.p. 168–170 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.55 (s, 1 H) 8.12–8.10 (m, 2 H), 7.99–7.88 (m, 4 H), 7.69–7.67 (m, 2 H), 7.42–7.40 (m, 4 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 197.8, 190.1, 132.6, 132.1, 132.1, 131.0, 130.5, 130.1, 129.9, 129.2, 128.0, 125.7, 124.2 ppm. IR (KBr film): $\tilde{\nu}$ = 3083, 3045, 2955, 2924, 2853, 1674, 1657, 1581, 1555, 1482, 1443, 1397, 1252 cm^{-1}. MS (ES^+): *m/z* = 389.4 $[M + H]^+$. $C_{22}H_{13}BrO_2$ (389.25): calcd. C 67.88, H 3.37; found C 67.79, H 3.43.

1-(Anthracen-9-yl)-2-(2-hydroxyphenyl)ethane-1,2-dione (6m): Prepared from 1-(2-hydroxyphenyl)ethanone (340.4 mg, 2.5 mmol), anthracene (445.6 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 14 h following Method B. Eluent: ethyl acetate 15% in hexane; light orange solid (407.9, 50%); m.p. 135–137 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 11.31 (s, 1 H), 8.58 (s, 1 H) 8.21 (d, *J* = 7.6 Hz, 1 H), 8.01–7.97 (m, 4 H), 7.59 (t, *J* = 7.6 Hz, 1 H), 7.45–7.43 (m, 4 H), 7.07–7.00 (m, 2 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 196.5, 195.6, 164.5, 138.2, 132.6, 132.3, 130.9, 130.2, 129.7, 129.3, 128.2, 125.7, 124.2, 120.0, 119.1, 115.7 ppm. IR (KBr film): $\tilde{\nu}$ = 3054, 3026, 3002, 2957, 2925, 2854, 1662, 1622, 1578, 1578, 1486, 1452, 1303, 1246 cm^{-1}. MS (ES^+): *m/z* = 327.0 $[M + H]^+$, 349.0 $[M + Na]^+$. $C_{22}H_{14}O_3$ (326.35): calcd. C 80.97, H 4.32; found C 80.87, H 4.12.

1-(Anthracen-9-yl)-2-(4-hydroxyphenyl)ethane-1,2-dione (6n): Prepared from 1-(4-hydroxyphenyl)ethanone (340.4 mg, 2.5 mmol), anthracene (445.6 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 16 h following Method B. Eluent: ethyl acetate 20% in hexane; yellow solid (465.0, 57%); m.p. 215–217 °C. ^1H NMR [400 MHz, $(CD_3)_2CO$]: δ = 8.85 (s, 1 H), 8.28–8.06 (m, 6 H), 7.61–7.56 (m, 4 H), 7.16–7.14 (m, 2 H), 5.05 (s, 1 H) ppm. ^{13}C NMR [100 MHz, $(CD_3)_2CO$]: δ = 198.6, 189.7, 163.8, 133.3, 131.3, 131.0, 129.7, 129.2, 127.8, 127.7, 125.9, 125.7, 124.3, 116.2 ppm. IR (KBr film): $\tilde{\nu}$ = 3384, 3056, 2926, 1692, 1649, 1595, 1569, 1514, 1445, 1296, 1227, 1162, 1111 cm^{-1}. MS (ES^+): *m/z* = 349.0 $[M + Na]^+$. $C_{22}H_{14}O_3$ (326.35): calcd. C 80.97, H 4.32; found C 80.73, H 4.23.

1-(Anthracen-9-yl)-2-(3-nitrophenyl)ethane-1,2-dione (6o): Prepared from 1-(3-nitrophenyl)ethanone (412.9 mg, 2.5 mmol), anthracene (445.6 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 15 h following Method B. Eluent: ethyl acetate 20% in hexane; light orange solid (568.5 mg, 64%); m.p. 132–134 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 9.08 (s, 1 H), 8.60–8.57 (m, 2 H), 8.51 (d, *J* = 7.6 Hz, 1 H), 8.02–8.00 (m, 2 H), 7.89–7.87 (m, 2 H), 7.76 (t, *J* = 7.6 Hz, 1 H), 7.45–7.43 (m, 4 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 197.0, 188.5, 148.7, 136.1, 133.6, 132.7, 131.0, 130.5, 130.2, 129.4, 129.0, 128.8, 128.3, 125.8, 125.5, 124.1 ppm. IR (KBr film): $\tilde{\nu}$ = 3056, 2923, 2852, 1676, 1649, 1609, 1554, 1533, 1441, 1354, 1249, 1079 cm^{-1}. MS (ES^+): *m/z* = 378.1 $[M + Na]^+$. $C_{22}H_{13}NO_4$ (355.35): calcd. C 74.36, H 3.69, N 3.94; found C 74.53, H 3.55, N 3.89.

1-(Anthracen-9-yl)-2-(4-nitrophenyl)ethane-1,2-dione (6p): Prepared from 1-(4-nitrophenyl)ethanone (412.9 mg, 2.5 mmol), anthracene (445.6 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 15 h following Method B. Eluent: ethyl acetate 20% in hexane; yellowish solid (586.3 mg, 66%); m.p. 187–189 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.67 (s, 1 H) 8.50–8.40 (m, 4 H), 8.09–8.07 (m, 2 H), 7.95–7.93 (m, 2 H), 7.52–7.50 (m, 4 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 197.0, 189.1, 151.1, 136.8, 132.6, 131.7, 130.9, 130.2, 129.4, 129.0, 128.3, 125.8, 124.2, 124.0 ppm. IR (KBr film): $\tilde{\nu}$ = 3113, 3074, 3054, 2925, 2855, 1678, 1624, 1598, 1555, 1525, 1444, 1408, 1353, 1345, 1321, 1266, 1254 cm^{-1}. GC–MS: *m/z* (%) = 355 (11) $[M]^+$ 355, 205 (80), 177 (100), 150 (92), 104 (62). $C_{22}H_{13}NO_4$ (355.35): calcd. C 74.36, H 3.69, N 3.94; found C 74.47, H 3.71, N 3.98.

***N*-[3-{2-(Anthracen-9-yl)-2-oxoacetyl}phenyl]acetamide (6q):** Prepared from *N*-(3-acetylphenyl)acetamide (443.0 mg, 2.5 mmol), anthracene (445.6 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 15 h following Method B. Eluent: ethyl acetate 25% in hexane; yellow solid (670.5 mg, 73%); m.p. 181–183 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.56 (s, 1 H), 8.13–7.88 (m, 7 H), 7.50–7.36 (m, 6 H), 1.99 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 197.8, 190.8, 168.7, 138.8, 132.6, 132.1, 130.9, 130.1, 130.1, 130.0, 129.2, 128.1, 126.6, 126.5, 125.7, 124.3, 121.1, 24.5 ppm. IR (KBr film): $\tilde{\nu}$ = 3305, 3087, 3054, 2925, 2854, 1668, 1612, 1587, 1554, 1514, 1485, 1368, 1304, 1250, 1175 cm^{-1}. MS (ES^+): *m/z* = 390.0 $[M + Na]^+$. $C_{24}H_{17}NO_3$ (367.40): calcd. C 78.46, H 4.66, N 3.81; found C 78.58, H 4.51, N 3.92.

1-(Anthracen-9-yl)-2-(2,4-dimethylphenyl)ethane-1,2-dione (6r): Prepared from 1-(2,4-dimethylphenyl)ethanone (370.5 mg, 2.5 mmol), anthracene (445.6 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 15 h following Method B. Eluent: ethyl acetate 8% in hexane; yellow solid (490.7 mg, 58%); m.p. 134–136 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.50 (s, 1 H), 8.11 (d, *J* = 8.0 Hz, 1 H), 8.03–7.93 (m, 4 H), 7.41–7.30 (m, 4 H), 7.15–7.11 (m, 2 H), 2.50 (s, 3 H), 2.33 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 198.6, 193.3, 144.7, 142.7, 133.8, 133.3, 131.8, 131.0, 130.7, 130.1, 129.1, 128.1, 127.8, 126.7, 125.5, 124.5, 22.1, 21.8 ppm. IR (KBr film): $\tilde{\nu}$ = 3079, 3059, 2926, 1670, 1658, 1594, 1554, 1522, 1449, 1253, 1119 cm^{-1}. MS (ES^+): *m/z* = 361.0 $[M + Na]^+$. $C_{24}H_{18}O_2$ (338.41): calcd. C 85.18, H 5.36; found C 85.27, H 5.42.

1-(Anthracen-9-yl)-2-mesitylethane-1,2-dione (6s): Prepared from 1-mesitylethanone (405.6 mg, 2.5 mmol), anthracene (445.6 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 16 h following Method B. Eluent: ethyl acetate 10% in hexane; orange solid (466.9 mg, 53%); m.p. 108–110 °C. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.52 (s, 1 H), 8.00–7.98 (m, 2 H), 7.76–7.74 (m, 2 H), 7.44–7.42 (m, 4 H), 6.85

(s, 2 H), 2.26 (s, 3 H), 2.21 (s, 6 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 198.4, 193.2, 140.3, 135.5, 133.6, 132.2, 131.0, 130.4, 129.3, 129.0, 129.0, 127.2, 125.7, 125.6, 124.7, 21.3, 20.5 ppm. IR (KBr film): $\tilde{\nu}$ = 3057, 3003, 2917, 2853, 1697, 1670, 1613, 1445, 1371, 1242, 1152, 1104 cm^{-1}. MS (ES$^+$): *m*/*z* = 375.0 [M + Na]$^+$. $C_{25}H_{20}O_2$ (352.43): calcd. C 85.20, H 5.72; found C 85.23, H 5.94.

1-(Naphthalen-2-yl)-2-phenylethane-1,2-dione (9a): Prepared from 1-(naphthalen-2-yl)ethanone (425.5 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and benzene (8 mL) over 18 h following Method A. Eluent: ethyl acetate 10% in hexane; light yellow solid (384.0 mg, 59%); m.p. 76–78 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.41 (s, 1 H), 8.11–7.89 (m, 6 H), 7.69–7.51 (m, 5 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 194.7, 136.4, 134.9, 133.6, 133.1, 132.3, 130.3, 130.0, 129.9, 129.6, 129.2, 129.1, 128.0, 127.2, 123.6 ppm. IR (KBr film): $\tilde{\nu}$ = 3084, 3057, 3031, 2973, 2927, 1671, 1659, 1626, 1596, 1580, 1466, 1449, 1438, 1396, 1371, 1354, 1325, 1307, 1276, 1251, 1211, 1190, 1176, 1153, 1126 cm^{-1}. MS (ES$^+$): *m*/*z* = 260.9 [M + H]$^+$, 282.9 [M + Na]$^+$. $C_{18}H_{12}O_2$ (260.29): calcd. C 83.06, H 4.65; found C 83.19, H 4.71.

1-(Naphthalen-2-yl)-2-(*p*-tolyl)ethane-1,2-dione (9b): Prepared from 1-(naphthalen-2-yl)ethanone (425.5 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and toluene (6 mL) over 16 h following Method B. Eluent: ethyl acetate 10% in hexane; yellow solid (350.0 mg, 51%); m.p. 90–92 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.40 (s, 1 H), 8.09 (d, *J* = 8.0 Hz, 1 H), 7.96–7.87 (m, 5 H), 7.63 (t, *J* = 6.8 Hz, 1 H), 7.54 (t, *J* = 8.0 Hz, 1 H), 7.31 (d, *J* = 8.0 Hz, 2 H), 2.43 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 194.9, 194.4, 146.3, 136.3, 133.5, 132.6, 132.3, 130.1, 129.9, 129.8, 129.5, 129.1, 127.9, 127.2, 126.1, 123.7, 22.0 ppm. IR (KBr film): $\tilde{\nu}$ = 3063, 2924, 2853, 1666, 1625, 1602, 1572, 1464, 1353, 1316, 1249, 1216, 1187, 1171, 1124 cm^{-1}. $C_{19}H_{14}O_2$ (274.32): calcd. C 83.19, H 5.14; found C 83.36, H 5.29.

1-(2,5-Dimethylphenyl)-2-(naphthalen-2-yl)ethane-1,2-dione (9c): Prepared from 1-(naphthalen-2-yl)ethanone (425.5 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and *p*-xylene (6 mL) over 16 h following Method B. Eluent: ethyl acetate 10% in hexane; yellowish solid (439.7 mg, 61%); m.p. 75–77 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.36 (s, 1 H), 8.04–8.02 (m, 1 H), 7.92–7.83 (m, 3 H), 7.58 (t, *J* = 7.2 Hz, 1 H), 7.50 (t, *J* = 7.2 Hz, 1 H), 7.40 (s, 1 H), 7.24 (d, *J* = 7.6 Hz, 1 H), 7.18 (d, *J* = 6.4 Hz, 1 H), 2.63 (s, 3 H), 2.21 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 197.0, 195.1, 138.4, 136.3,135.8, 134.7, 133.5, 133.4, 132.5, 132.4, 130.5, 129.9, 129.5, 129.1, 128.0, 127.1, 123.9, 21.5, 20.8 ppm. IR (KBr film): $\tilde{\nu}$ = 3061, 3023, 2957, 2924, 2855, 1678, 1665, 1623, 1596, 1570, 1497, 1466, 1438, 1387, 1380, 1361, 1309, 1277, 1249, 1221, 1196, 1162, 1129 cm^{-1}. GC–MS: *m*/*z* = 288 [M]$^+$. $C_{20}H_{16}O_2$ (288.35): calcd. C 83.31, H 5.59; found C 83.46, H 5.63.

1-(4-Methoxyphenyl)-2-(naphthalen-2-yl)ethane-1,2-dione (9d): Prepared from 1-(naphthalen-2-yl)ethanone (425.5 mg, 2.5 mmol), anisole (270.4 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 14 h following Method B. Eluent: ethyl acetate 15% in hexane; yellowish solid (508.1 mg, 70%); m.p. 69–71 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.34 (s, 1 H), 8.03 (d, *J* = 8.8 Hz, 1 H) 7.94–7.82 (m, 5 H), 7.57 (t, *J* = 7.2 Hz, 1 H), 7.49 (t, *J* = 7.2 Hz, 1 H), 6.93–6.91 (m, 2 H), 3.82 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.0, 193.3, 165.0, 136.3, 133.4, 132.4, 132.4, 130.5, 129.9, 129.9, 129.1, 128.0, 127.1, 126.2, 123.7, 114.4, 55.7 ppm. IR (KBr film): $\tilde{\nu}$ = 3058, 3006, 2933, 2839, 1663, 1628, 1599, 1574, 1509, 1464, 1422, 1313, 1252, 1219, 1167 cm^{-1}. MS (ES$^+$): *m*/*z* = 312.9 [M + Na]$^+$. $C_{19}H_{14}O_3$ (290.32): calcd. C 78.61, H 4.86; found C 78.70, H 4.90.

1-(Naphthalen-1-yl)-2-(naphthalen-2-yl)ethane-1,2-dione (10a): Prepared from 1-(naphthalen-2-yl)ethanone (425.5 mg, 2.5 mmol), naphthalene (320.4 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 15 h following Method B. Eluent: ethyl acetate 15% in hexane; brownish yellow solid (535.3 mg, 69%); m.p. 128–130 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.28 (d, *J* = 8.8 Hz, 1 H), 8.41 (s, 1 H), 8.08–8.05 (m, 2 H), 7.92–7.82 (m, 5 H), 7.72–7.68 (m, 1 H), 7.59–7.55 (m, 2 H), 7.47 (t, *J* = 7.6 Hz, 1 H), 7.41 (t, *J* = 7.6 Hz, 1 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 197.2, 194.7, 136.3, 136.0, 135.2, 134.1, 133.5, 132.4, 131.0, 130.7, 129.9, 129.5, 129.5, 129.2, 128.8, 128.7, 127.9, 127.2, 127.1, 126.0, 124.5, 123.9 ppm. IR (KBr film): $\tilde{\nu}$ = 3063, 3048, 2926, 2855, 1679, 1660, 1623, 1594, 1571, 1508, 1464, 1437, 1387, 1370, 1357, 1290, 1261, 1243, 1218, 1198, 1165, 1098 cm^{-1}. MS (ES$^+$): *m*/*z* = 333.0 [M + Na]$^+$. $C_{22}H_{14}O_2$ (310.35): calcd. C 85.14, H 4.55; found C 85.16, H 4.39.

1-(Furan-2-yl)-2-(naphthalen-1-yl)ethane-1,2-dione (10b): Prepared from 1-(furan-2-yl)ethanone (275.3 mg, 2.5 mmol), naphthalene (320.4 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 16 h following Method B. Eluent: ethyl acetate 15% in hexane; brownish yellow solid (237.7 mg, 38%); m.p. 64–66 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.13 (d, *J* = 8.4 Hz, 1 H), 8.11 (d, *J* = 8.0 Hz, 1 H), 7.98 (d, *J* = 7.2 Hz, 1 H), 7.92 (d, *J* = 8.0 Hz, 1 H), 7.75–7.69 (m, 2 H), 7.60 (t, *J* = 7.6 Hz, 1 H), 7.50 (t, *J* = 8.0 Hz, 1 H), 7.41 (d, *J* = 3.6 Hz, 1 H), 6.62–6.61 (m, 1 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 194.2, 181.3, 150.3, 149.3, 135.9, 134.5, 134.1, 131.1, 129.3, 128.8, 128.4, 127.1, 125.8, 124.4, 124.0, 113.7 ppm. IR (KBr film): $\tilde{\nu}$ = 3163, 3139, 3123, 3058, 3045, 3009, 2924, 2853, 1658, 1651, 1595, 1570, 1558, 1509, 1463, 1399, 1315, 1268, 1243, 1231, 1215, 1193, 1159, 1080, 1031 cm^{-1}. MS (ES$^+$): *m*/*z* = 272.9 [M + Na]$^+$. $C_{16}H_{10}O_3$ (250.25): calcd. C 76.79, H 4.03; found C 76.86, H 4.13.

1-(Naphthalen-1-yl)-2-(thiophen-2-yl)ethane-1,2-dione (10c): Prepared from 1-(thiophen-2-yl)ethanone (315.5 mg, 2.5 mmol), naphthalene (320.4 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 16 h following Method B. Eluent: ethyl acetate 15% in hexane; yellow sticky solid (379.5 mg, 57%). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 9.16 (d, *J* = 8.4 Hz, 1 H), 8.12 (d, *J* = 8.0 Hz, 1 H), 8.00 (d, *J* = 7.2 Hz, 1 H), 7.94 (d, *J* = 8.0 Hz, 1 H), 7.86–7.83 (m, 2 H), 7.75–7.71 (m, 1 H), 7.62 (t, *J* = 7.6 Hz, 1 H), 7.52 (t, *J* = 7.6 Hz, 1 H), 7.20 (t, *J* = 4.4 Hz, 1 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 194.7, 186.3, 140.3, 136.7, 136.6, 135.9, 134.8, 134.0, 131.1, 129.3, 128.8, 128.8, 128.5, 127.1, 125.8, 124.4 ppm. IR (Neat): $\tilde{\nu}$ = 3091, 3052, 3011, 2953, 1654, 1591, 1572, 1508, 1461, 1435, 1409, 1353, 1304, 1291, 1263, 1226, 1178, 1106, 1078, 1053 cm^{-1}. MS (ES$^+$): *m*/*z* = 267.2 [M + H]$^+$, 288.9 [M + Na]$^+$. $C_{16}H_{10}O_2S$ (266.31): calcd. C 72.16, H 3.78, S 12.04; found C 72.29, H 3.79, S 11.97.

1-(Anthracen-9-yl)-2-(thiophen-2-yl)ethane-1,2-dione (10d): Prepared from 1-(thiophen-2-yl)ethanone (315.5 mg, 2.5 mmol), anthracene (445.6 mg, 2.5 mmol), SeO_2 (554.8 mg, 5.0 mmol), *p*TsOH·H_2O (475.6 mg, 2.5 mmol), and acetonitrile (6 mL) over 14 h following Method B. Eluent: ethyl acetate 20% in hexane; yellowish solid (363.9 mg, 46%); m.p. 145–147 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.55 (s, 1 H), 8.23 (d, *J* = 3.6 Hz, 1 H), 8.00–7.98 (m, 2 H), 7.86–7.84 (m, 3 H), 7.44–7.40 (m, 4 H), 7.25 (t, *J* = 4.4 Hz, 1 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 197.7, 182.3, 138.1, 137.7, 137.2, 134.1, 133.5, 131.4, 130.9, 130.2, 129.8, 129.1, 129.1, 127.7, 127.2, 125.6, 124.4 ppm. IR (KBr film): $\tilde{\nu}$ =

3133, 3091, 3052, 2926, 1696, 1678, 1637, 1592, 1445, 1407, 1354, 1343, 1262, 1108 cm^{-1}. MS (ES^+): *m/z* = 338.9 $[M + Na]^+$. $C_{20}H_{12}O_2S$ (316.37): calcd. C 75.93, H 3.82, S 10.14; found C 75.81, H 3.80, S 10.23.

Supporting Information (see footnote on the first page of this article): Copies of ^1H and ^{13}C NMR and IR spectra for all new compounds.

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