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Visible-light-mediated synthesis of amides from aldehydes and amines via *in-situ* acid chloride formation

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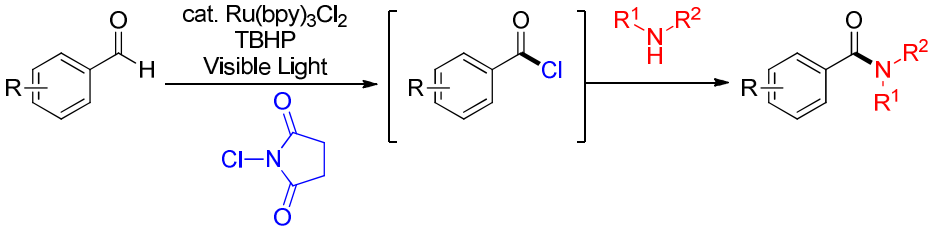
ABSTRACT



An efficient visible-light photocatalysis-based one-pot amide synthesis method was developed; visible-light irradiation of a mixture of an aldehyde, *tert*-butyl hydrogen peroxide, and *N*-chlorosuccinimide using $Ru(bpy)_3Cl_2$ photocatalyst afforded an acid chloride, which subsequently reacted with amine to yield the corresponding amide. The reaction was used to synthesize moclobemide and a D3 receptor intermediate.

Introduction

The amide bond is one of the most important bonds in biological macromolecules, and almost

25% of the drug molecules contain an amide bond.[1] Because of the high stability, polarity, and conformational diversity of amide bonds, they have been used in synthetic intermediates to functional materials.[1g] Owing to the importance of amide bonds, intensive efforts have been made to develop methods for the construction of the amide bonds. Conventionally, amide bonds are constructed by coupling of carboxylic acid derivatives with amines using certain activating groups [Figure 1. (1)].[2] Recently, a plethora of methods have been developed for the construction of amide bonds, including the transition-metal-catalyzed[3] and *N*-heterocyclic carbene (NHC)-catalyzed[4] oxidative amidation of aldehydes with amines.[5–7] However, these direct oxidative amidation of aldehydes has some limitations, mainly the limited substrate scope of amines, and thus the acylation of amines by activated acid derivatives is still the major method for the synthesis of amides. Therefore, alternative efforts have been made for the conversion of aldehydes to activated intermediates that can be utilized in situ for the synthesis of amides. Recently, Yamamoto, Barbas, and Luca independently reported the synthesis of amides from aldehydes through the formation of activated intermediates via acyl radicals [Figure 1. (2)].[8,9,10] The facile synthesis of active intermediates from aldehydes prompted us to develop a visible-light-induced one-pot method for the synthesis of amides. In the process, an acyl radical generated from an aldehyde by visible-light photocatalysis using *tert*-butyl hydrogen peroxide (TBHP) as the oxidant was converted to the corresponding acid chloride intermediate by abstracting a Cl radical from *N*-chlorosuccinimide (NCS).[11,12] Then, the addition of an amine to the acid chloride intermediate afforded the corresponding amide [Figure 1. (3)].

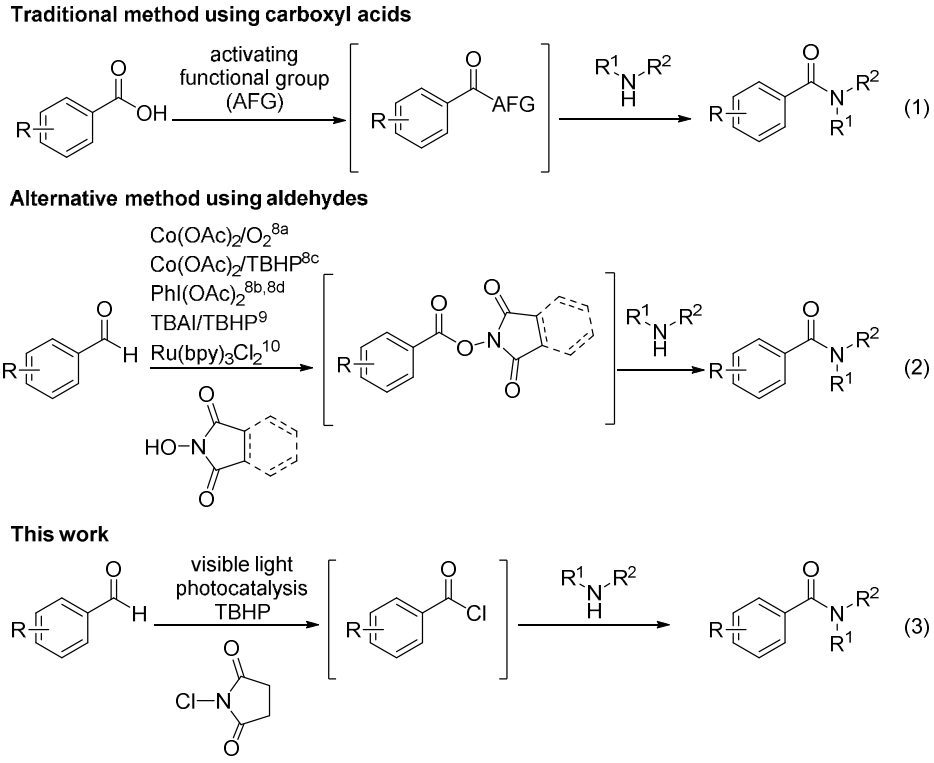


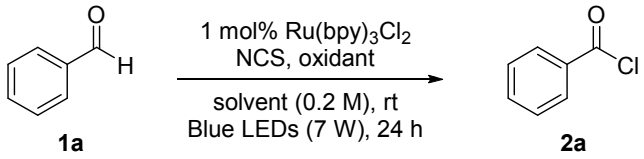
Figure 1. Amide bond formation via active intermediates.

Results and Discussion

First, benzaldehyde (**1a**) was converted into benzoyl chloride using TBHP and NCS as the source of Cl in CH_3CN, and only 30% of benzoyl chloride (**2a**) was obtained (Table 1, entry 1). We envisioned that the combined use of visible-light photocatalytic conditions would accelerate the reaction since TBHP can produce radicals under visible-light conditions.[13,14] To our delight, the use of $Ru(bpy)_3Cl_2$ under visible-light irradiation increased the yield of **2a** to 78% (Table 1, entry 2). A control experiment showed that the reaction requires visible-light irradiation for a satisfactory conversion of benzaldehyde to benzoyl chloride (Table 1, entry 3). A decrease in the amount of NCS decreased the yield of the reaction, and a further increase in the amount of NCS did not increase the yield (Table 1, entries 4–6). On the other hand, the yield of the reaction

increased with a less amount of TBHP, even a catalytic amount of TBHP was effective contrary to the previous report,[12] indicating that TBHP can either be regenerated during the course of the reaction or acts as an initiator (Table 1, entries 7–11).

Table 1. Oxidation of aldehyde to acid halide[a]



1a **2a**

entry	Oxidant (equiv)	equiv of NCS	solvent	variations	yield (%)[b] **2a**
1	^{t}BuOOH (4.0)	3.0	CH_3CN	no $Ru(bpy)_3Cl_2$	30
2	^{t}BuOOH (4.0)	3.0	CH_3CN		78
3	^{t}BuOOH (4.0)	3.0	CH_3CN	no light	32
4	^{t}BuOOH (4.0)	1.5	CH_3CN		17
5	^{t}BuOOH (4.0)	2.0	CH_3CN		56
6	^{t}BuOOH (4.0)	3.5	CH_3CN		70
7	^{t}BuOOH (2.5)	3.0	CH_3CN		82
8	**^{t}BuOOH (1.3)**	**3.0**	**CH_3CN**		**84**[c]
9	^{t}BuOOH (1.0)	3.0	CH_3CN		80
10	^{t}BuOOH (0.6)	3.0	CH_3CN		75
11	^{t}BuOOH (0.25)	3.0	CH_3CN		68
12	^{t}BuOOH (1.3)	-	CH_3CN	NBS (3.0 equiv)	46[d]
13	^{t}BuOOH (1.3)	3.0	CH_2Cl_2		84
14	^{t}BuOOH (1.3)	3.0	DMF		0
15	^{t}BuOOH (1.3)	3.0	CH_3OH		38
16	**^{t}BuOOH (1.3)**	**3.0**	**CH_3CN**	**0.25 M**	**85**
17	^{t}BuOOH (1.3)	3.0	CH_3CN	0.1 M	64
18	^{t}BuOOH (1.3)	3.0	CH_3CN	2.0 mol% cat.	84
19	^{t}BuOOH (1.3)	3.0	CH_3CN	0.5 mol% cat.	80
20	^{t}BuOOH (1.3)	3.0	CH_3CN	0.1 mol% cat.	77
21	O_2	3.0	CH_3CN		trace
22	-	3.0	CH_3CN	Degassed CH_3CN	0

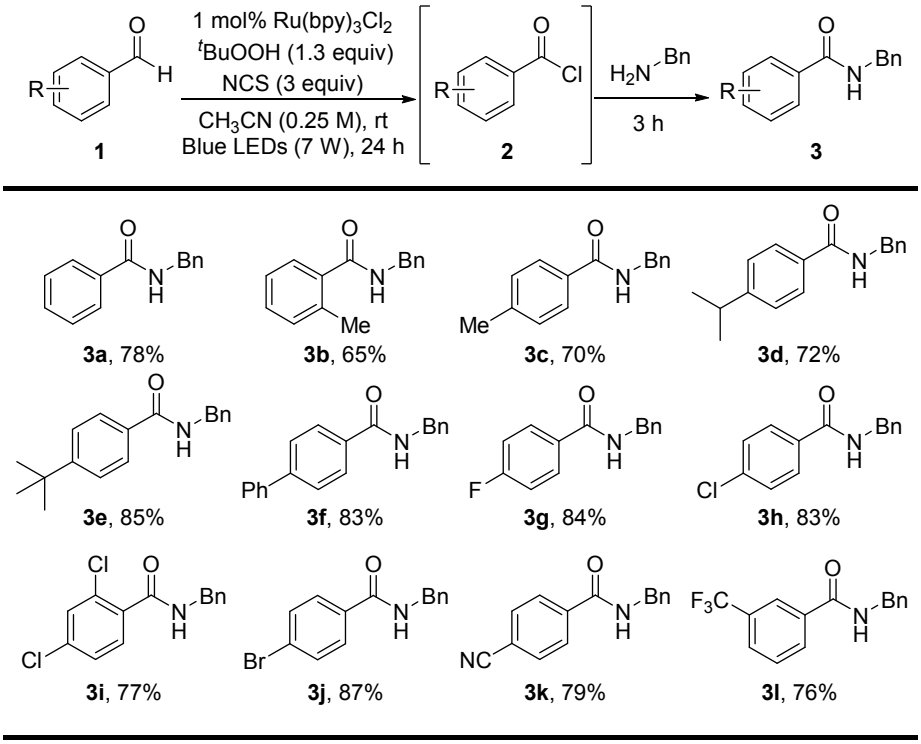
[a] reaction scale: **1a** (0.1 mmol), [b] yields based on GCMS using dodecane as the internal standard, [c] addition of benzyl amine afforded the corresponding amide in 78% yield, [d] yield of benzoyl bromide, which was formed along with benzoic acid and *tert*-butyl ester.

The use of *N*-bromosuccinimide (NBS) instead of NCS afforded a mixture of undesired products because of the higher reactivity of benzoyl bromide compared to benzoyl chloride (Table 1, entry 12). Different photocatalysts including $Ru(phen)_3Cl_2$, *fac*-$Ir(ppy)_3$, *fac*-$Ir(dFppy)_3$, and $Ir(dtbbpy)(ppy)_2PF_6$ were investigated; they showed a similar reactivity. Among the catalysts, $Ru(bpy)_3Cl_2$ was selected because of its low cost and easy availability. CH_3CN was the best solvent (Table 1, entries 13–15). The concentration studies showed that the use of 0.25 M concentration provided the best results (Table 1, entries 8, 16, and 17). Although a satisfactory conversion was achieved even by using 0.1 mol% of the photocatalyst, 1 mol% of the photocatalyst was selected for further studies for the reproducible results (Table 1, entries 8 and 18–20). When O_2 was used as the oxidant, the yield decreased significantly (Table 1, entries 21 and 22).

With the optimized conditions for the conversion of aldehydes to acid chlorides, a one-pot method was developed for the synthesis of amides. The addition of benzyl amine to a reaction mixture of benzoyl chloride (**2a**) afforded *N*-benzylbenzamide (**3a**) in 78% yield. This one-pot method was applied for the synthesis of amides from diverse benzaldehyde derivatives 1 (Table 2). The reactions of aldehydes bearing both electron-rich and deficient aryl substituents proceeded smoothly, affording the corresponding amides **3** in good yields. The substituents at the ortho position of the aryl ring did not affect the reactivity (**3b** and **3i**). The reaction tolerated the presence of a nitrile group (**3k**) and halides including fluoro (**3g**), chloro (**3h**), **3i**), and bromo (**3j**). However, some side reactions were also observed. In the reactions of **1b**, **1c**, and **1d**, the benzylic chlorination afforded 5–10% of chlorinated products along with the desired amides **3b**, **3c**, and **3d**, respectively. In the reaction of **1f**, aryl chlorinated products were also detected. Aliphatic aldehydes were not suitable substrates because the decarbonylative products were

obtained.[15] This is probably because of the lower stability of aliphatic acyl radicals compared to the benzoyl radical, causing the degradation of radicals before the abstraction of a chlorine radical from NCS.

Table 2. Amide Synthesis from aldehydes via acid chlorides: substrate scope with respect to benzaldehydes[a,b]

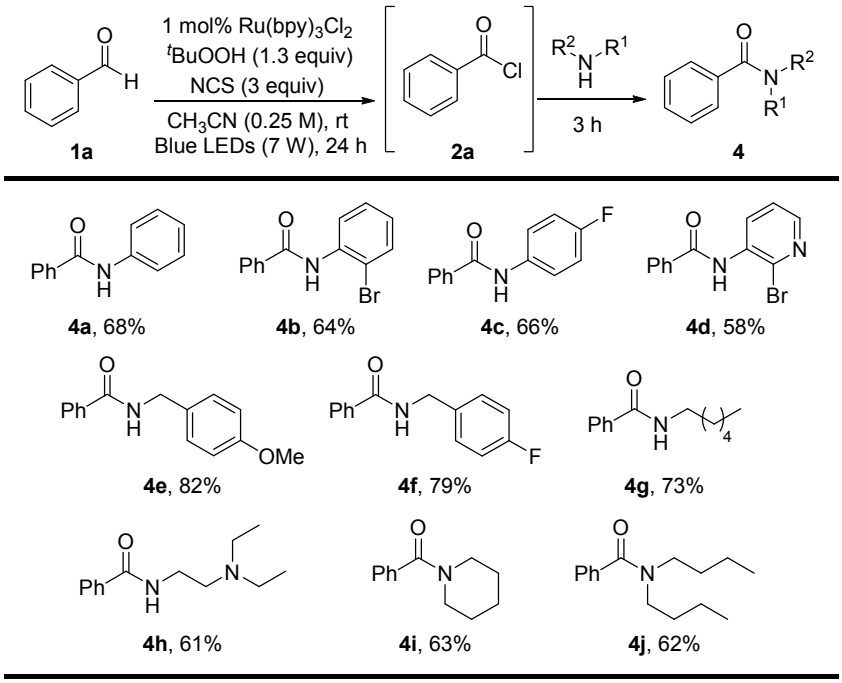


[a] reaction scale: **1** (1.0 mmol), benzyl amine (1.5 mmol). [b] Isolated yields based on the average of two runs.

Further, we studied the substrate scope with respect to different amines for the synthesis of amides (Table 3). Both aromatic and aliphatic amines participated well in the reaction. In general, benzyl amines (**4e** and **4f**) showed better reactivity. The successful amide formation with an aminopyridine (**4d**) indicates that the reaction can be applied to the synthesis of amide bonds containing heterocycles, important structural motifs in many applications including

pharmaceuticals.

Table 3. Amide Synthesis from aldehydes via acid chlorides: Substrate scope with respect to amines[a,b]

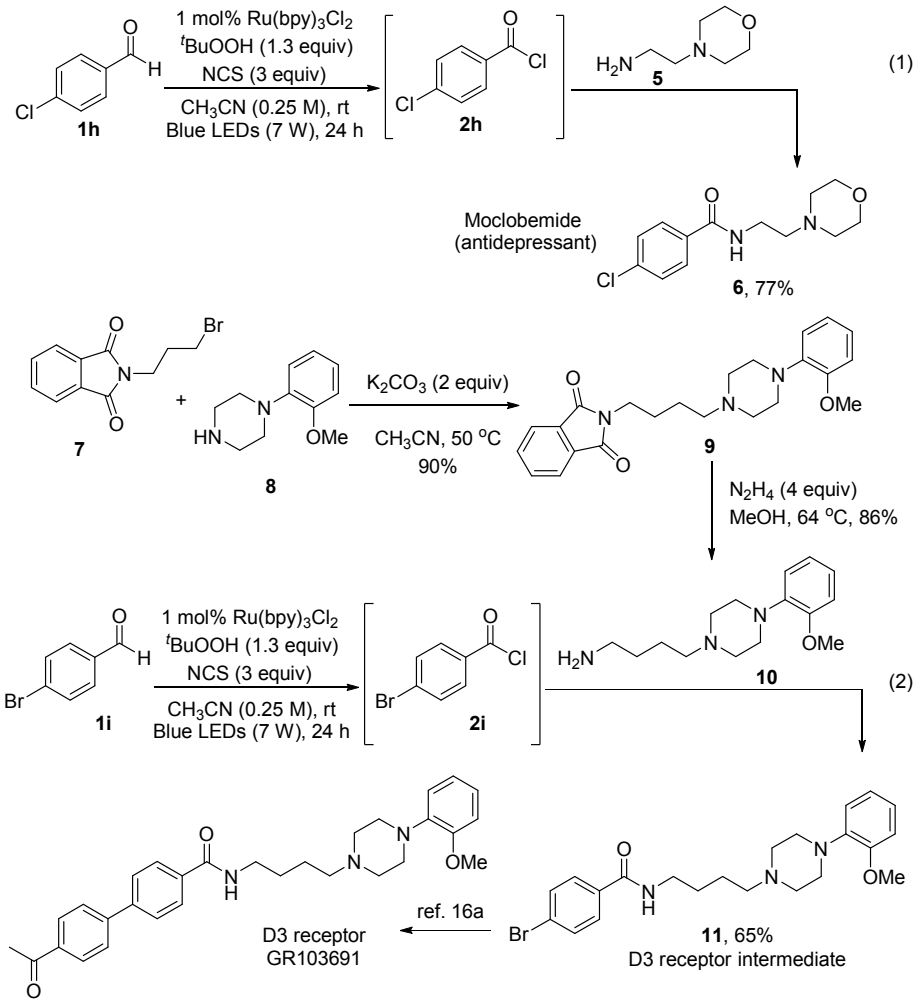


[a] reaction scale: **1** (1.0 mmol), amine (1.5 mmol). [b] Isolated yields based on the average of two runs.

The facile reaction developed in this study prompted us to synthesize some drug molecules using the reaction conditions. An antidepressant moclobemide (**6**)[16] was synthesized from 4-chlorobenzaldehyde (**1h**) and 2-morpholinoethan-1-amine (**5**) in 77% yield [Scheme 1. (1)]. A D3 receptor intermediate **11** was also synthesized from 4-bromobenzaldehyde (**1i**) and an amine intermediate **10** in 65% yield; compound **11** was converted to D3 receptor GR103691 following

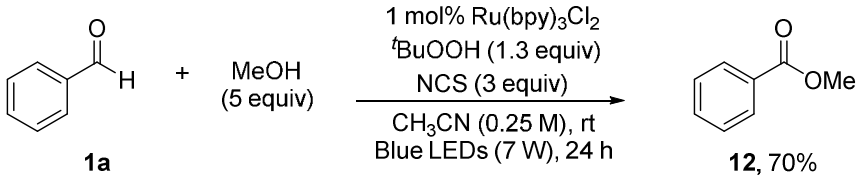
a known procedure[17a] [Scheme 1. (2)]. These results indicate that the developed method for the synthesis of amides is applicable to the synthesis of natural products and pharmaceutically and industrially relevant compounds.

Scheme 1. Synthesis of antidepressant Moclobemide 6 and D3 receptor GR103691.



Next, a one-pot method was investigated for the synthesis of esters from aldehydes and alcohols instead of amines. Although various alcohols, such as methanol, ethanol, isopropanol, hexanol, benzyl alcohol, and phenol were tried, only reaction with methanol afforded the corresponding methyl benzoate **12** in a reasonable yield (Scheme 2).

Scheme 2. Visible-light-mediated ester synthesis from benzaldehyde and methanol.



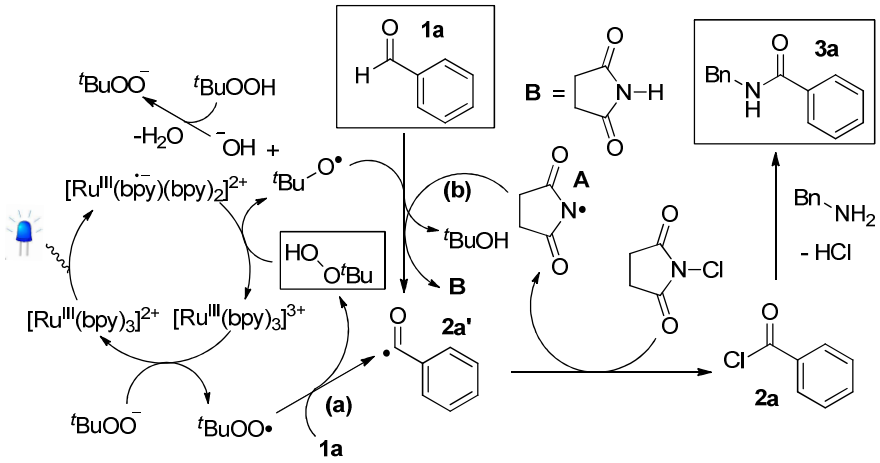


Figure 2. Proposed mechanism

Based on the results of the above mentioned studies, a plausible mechanism is proposed for the synthesis of amides using benzaldehyde **1a** and benzyl amine outlined in Figure 2.[18] The photoexcitation of $[Ru^{II}(bpy)_3]^{2+}$ produces the metal-to-ligand charge-transfer excited state $[Ru^{III}bpy^{\bullet-}(bpy)_2]^{2+}$, which is oxidatively quenched by a single-electron transfer to ^{t}BuOOH, generating the key intermediates ^{t}BuO• and $^{-}$OH, along with $[Ru^{III}(bpy)_3]^{3+}$. Hydrogen abstraction by ^{t}BuO• from benzaldehyde (**1a**) affords acyl radical **2a′** and ^{t}BuOH.[11] This acyl radical further abstracts a Cl radical from NCS, generating succinimide radical intermediate **A** and benzoyl chloride (**2a**). Then, the addition of benzyl amine to the reaction mixture affords N-benzylbenzamide (**3a**) by a substitution reaction. On the other hand, a single-electron transfer from ^{t}BuOO^{-}, which is generated by the deprotonation of ^{t}BuOOH with $^{-}$OH, to $[Ru^{III}(bpy)_3]^{3+}$

regenerates the photocatalyst, $[Ru^{II}(bpy)_3]^{2+}$.[11] The conversion of aldehydes to acid chlorides works even with a sub-stoichiometric amount of TBHP (Table 1, entries 9 and 10); this is supported by the following pathways: (a) ^{t}BuOO• abstracts a H• from aldehyde **1a**, affording **2a′** intermediate and regenerating TBHP, (b) the H• abstraction by succinimide radical **A** from aldehyde C–H also generates **2a′** with succinimide **B**.

In conclusion, we developed an efficient one-pot method for the synthesis of amides from aldehydes and amines. The amide bonds were constructed from aldehydes via the in-situ formation of acid chlorides using $Ru(bpy)_3Cl_2$ as the photocatalyst and TBHP as the oxidant under visible-light irradiation. The new method is a practical and efficient for the synthesis of amides from different benzaldehyde derivatives and diverse aliphatic and aromatic amines. The process was successfully applied to the synthesis of drug molecules, an antidepressant moclobemide and a D3 receptor intermediate.

EXPERIMENTAL SECTION

General experimental procedure for amide synthesis

An oven-dried resealable tube, equipped with a magnetic stir bar, was charged with an aldehyde (1.0 mmol), $Ru(bpy)_3Cl_2$ (1 mol%), TBHP (1.3 equiv), NCS (3.0 equiv), and CH_3CN (0.25 M). The tube was stoppered with a silicone septa screw-cap and placed under blue LEDs at room temperature. After 24 h irradiation, amine (1.5 equiv) was added, and the reaction was stirred for 3 h. The progress of the reaction was monitored by TLC and GC. After the completion of the reaction, the reaction mixture was diluted with ethyl acetate. After the aqueous work-up, the

organic layers were combined, dried over $MgSO_4$, and concentrated in vacuo. The residue was purified by flash column chromatography, affording the desired amide product.

In the reactions for **4h**, **6**, and **11**, before the addition of amines, the reaction mixture was concentrated in vacuo, dissolved in a 1% TEA/hexane solution and filtered.

Analytic Data for amides

***N*-benzylbenzamide (3a):**[5e] white solid (165 mg, 78%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.77 (d, J = 7.8 Hz, 2H), 7.48–7.42 (m, 1H), 7.40–7.34 (m, 2H), 7.34–7.28 (m, 4H), 7.28–7.24 (m, 1H), 6.84 (bs, 1H), 4.57 (d, J = 6.0 Hz, 2H).^{13}C NMR (151 MHz, $CDCl_3$) δ 167.6, 138.5, 134.5, 131.6, 128.8, 128.7, 127.9, 127.6, 127.2, 44.2. IR (neat): ν_{max} = 3318, 3058, 1639, 1538, 1310, 691 cm^{-1}; R_f 0.45 (hexane/ethyl acetate 2:1).

***N*-benzyl-2-methylbenzamide (3b):**[19b] white solid (146 mg, 65%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.39 − 7.27 (m, 7H), 7.24 − 7.15 (m, 2H), 6.20 (bs, 1H), 4.60 (d, J = 6.0 Hz, 2H), 2.44 (s, 3H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 170.0, 138.1, 136.5, 136.1, 131.1, 130.0, 128.8, 127.8, 127.2, 126.7, 125.7, 43.9, 19.8. IR (neat): ν_{max} = 3308, 1647, 1541, 1200, 1000, 735, 698 cm^{-1}; R_f 0.36 (hexane/ethyl acetate 3:1).

***N*-benzyl-4-methylbenzamide (3c):**[5e] white solid (158 mg, 70%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.69 (d, J = 8.4 Hz, 2H), 7.38–7.32 (m, 4H), 7.31–7.27 (m, 1H), 7.22 (d, J = 8.4 Hz, 2H), 6.47 (bs, 1H), 4.63 (d, J = 6.0 Hz, 2H), 2.39 (s, 3H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 167.5, 142.2, 138.5, 131.7, 129.4, 129.0, 128.1, 127.8, 127.2, 44.3, 21.6. IR (neat): ν_{max} = 3309, 1637, 1544, 1506, 907, 841, 721 cm^{-1}; R_f 0.36 (hexane/ethyl acetate 3:1).

***N*-benzyl-4-isopropylbenzamide (3d):** white solid (182 mg, 72%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.73 (d, J = 8.4 Hz, 2H), 7.38–7.33 (m, 4H), 7.32–7.26 (m, 1H), 7.28 (d, J = 8.4 Hz, 2H), 6.39 (bs, 1H), 4.65 (d, J = 6.0 Hz, 2H), 2.95 (hept, J = 7.2 Hz, 1H), 1.26 (d, J = 6.6 Hz, 6H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 167.5, 153.0, 138.5, 132.1, 129.0, 128.1, 127.8, 127.3, 126.9, 44.3, 34.3, 24.0. IR (neat): ν_{max} = 3319, 1636, 1541, 1506, 1311, 852, 697 cm^{-1}; R_f 0.45 (hexane/ethyl acetate 3:1).

***N*-benzyl-4-(tert-butyl)benzamide (3e):**[19f] white solid (227 mg, 85%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.77 (d, *J* = 8.4 Hz, 2H), 7.43 (d, *J* = 8.4 Hz, 2H), 7.37–7.32 (m, 4H), 7.31–7.28 (m, 1H), 6.48 (bs, 1H), 4.64 (d, *J* = 6.0 Hz, 2H), 1.44 (s, 9H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 167.4, 150.1, 138.3, 132.6, 129.0, 128.1, 127.8, 127.2, 126.5, 44.3, 40.3, 26.7. IR (neat): ν_{max} = 3318, 1732, 1708, 1540, 1152, 698, 645 cm^{-1}; R_f 0.52 (hexane/ethyl acetate 2:1).

***N*-benzyl-[1,1'-biphenyl]-4-carboxamide (3f):**[19c] white solid (239 mg, 83%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.87 (d, *J* = 8.4 Hz, 2H), 7.63 (d, *J* = 8.4 Hz, 2H), 7.61–7.57 (m, 2H), 7.47–7.43 (m, 2H), 7.40–7.37 (m, 1H), 7.37–7.33 (m, 4H), 7.32–7.28 (m, 1H), 6.66 (bs, 1H), 4.66 (d, *J* = 6.0 Hz, 2H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 167.3, 144.5, 140.1, 138.4, 133.1, 129.1, 129.0, 128.2, 128.1, 127.8, 127.7, 127.4, 127.4, 44.3. IR (neat): ν_{max} = 3326, 1701, 1636, 1541, 745, 692 cm^{-1}; R_f 0.52 (hexane/ethyl acetate 2:1).

***N*-benzyl-4-fluorobenzamide (3g):**[7e] white solid (193 mg, 84%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.79 (dd, $J_{H\text{-}F}$ = 9.0 Hz, $J_{H\text{-}H}$ = 5.4 Hz, 2H), 7.36–7.31 (m, 4H), 7.31–7.27 (m, 1H), 7.07 (dd, $J_{H\text{-}F}$ = 9.0 Hz, $J_{H\text{-}H}$ = 8.4 Hz, 2H), 6.62 (bs, 1H), 4.60 (d, *J* = 6.0 Hz, 2H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 166.6, 164.9 (d, *J* = 252.17 Hz), 138.3, 130.7 (d, *J* = 3.02 Hz), 129.5 (d, *J* = 9.06 Hz), 129.0, 128.1, 127.8, 115.8, 115.7, 44.4. IR (neat): ν_{max} = 1654, 1497, 1264, 906, 726 cm^{-1}; R_f 0.39 (hexane/ethyl acetate 3:1).

***N*-benzyl-4-chlorobenzamide (3h):**[5e] white solid (204 mg, 83%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.72 (d, *J* = 8.4 Hz, 2H), 7.38 (d, *J* = 8.4 Hz, 2H), 7.37–7.31 (m, 4H), 7.31–7.27 (m, 1H), 6.54 (bs, 1H), 4.61 (d, *J* = 5.4 Hz, 2H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 166.5, 138.2, 138.0, 132.9, 129.0, 128.6, 128.1, 127.9, 44.4. IR (neat): ν_{max} = 3309, 1637, 1552, 1487, 1320, 1013, 849, 711, 669 cm^{-1}; R_f 0.58 (hexane/ethyl acetate 2:1).

***N*-benzyl-2,4-dichlorobenzamide (3i):** white solid (216 mg, 77%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.58 (d, *J* = 7.8 Hz, 1H), 7.39 (d, *J* = 2.4 Hz, 1H), 7.38–7.32 (m, 4H), 7.32–7.26 (m, 2H), 6.65 (bs, 1H), 4.61 (d, *J* = 6.0 Hz, 2H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 165.6, 137.7, 137.0, 133.5, 131.7, 131.4, 130.2, 129.0, 128.0, 127.9, 127.7, 44.5. IR (neat): ν_{max} = 3263, 1644, 1587, 1540, 830, 743, 693 cm^{-1}; R_f 0.58 (hexane/ethyl acetate 2:1).

***N*-benzyl-4-bromobenzamide (3j):**[7e] white solid (252 mg, 87%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.65 (d, *J* = 8.4 Hz, 2H), 7.56 (d, *J* = 8.4 Hz, 2H), 7.38–7.31 (m, 4H), 7.31–7.28 (m, 1H), 6.42 (bs, 1H), 4.62 (d, *J* = 5.4 Hz, 2H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 166.6, 138.1, 133.4,

132.0, 129.1, 128.8, 128.2, 128.0, 126.5, 44.5. IR (neat): νmax = 3311, 1638, 1549, 1322, 1011, 847, 669 cm^{-1}; R_f 0.58 (hexane/ethyl acetate 2:1).

***N*-benzyl-4-cyanobenzamide (3k):**[19c] white solid (187 mg, 79%); ^1H NMR (600 MHz, $CDCl_3$) δ 7.87 (d, *J* = 8.4, 2H), 7.64 (d, *J* = 8.4, 2H), 7.36–7.24 (m, 5H),7.23 (bs, 1H) 4.58 (d, *J* = 5.6, Hz, 2H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 165.9, 138.2, 137.8, 132.4, 128.8, 127.9, 127.8, 118.1, 114.9, 44.2. IR (neat): ν_{max} = 3309, 1708, 1540, 1158, 906, 722 cm^{-1}; R_f 0.41 (hexane/ethyl acetate 3:1).

***N*-benzyl-3-(trifluoromethyl)benzamide (3l):**[19d] white solid (212 mg, 76%); ^1H NMR (600 MHz, $CDCl_3$) δ 8.05 (t, *J* = 1.8 Hz, 1H), 7.96 (d, *J* = 7.8 Hz, 1H), 7.74 (d, *J* = 7.8 Hz, 1H), 7.53 (dd, *J* = 7.8 Hz, 1H), 7.37–7.32 (m, 4H), 7.32–7.27 (m, 1H), 6.77 (bs, 1H), 4.62 (d, *J* = 5.4 Hz, 2H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 166.2, 138.0, 135.4, 131.3 (q, *J* = 32.78 Hz), 130.5, 129.4, 129.0, 128.3 (q, *J* = 3.6 Hz), 128.1, 128.0, 124.3 (q, *J* = 3.6 Hz), 123.9 (q, *J* = 272.71 Hz), 44.5. IR (neat): ν_{max} = 3295, 1641, 1541, 1333, 1278, 1168, 1125, 696 cm^{-1}; R_f 0.41 (hexane/ethyl acetate 3:1).

***N*-phenylbenzamide (4a):**[5b] white solid (134 mg, 68%); ^1H NMR (600 MHz, DMSO-d_6) δ 10.03 (bs, 1H), 7.93 (dd, *J* = 8.4, 1.2 Hz, 2H), 7.75 (dd, *J* = 8.4, 1.2 Hz, 2H), 7.49 (tt, *J* = 7.2, 1.2 Hz, 1H), 7.44 (dd, *J* = 8.4, 1.2 Hz, 2H), 7.27 (dd, *J* = 8.4, 7.2 Hz, 2H), 7.04 (tt, *J* = 7.2, 1.2 Hz, 1H). ^{13}C NMR (151 MHz, DMSO-d_6) δ 164.7, 137.9, 134.0, 130.0, 127.2, 126.9, 126.4, 122.4, 119.3. IR (neat): ν_{max} = 3347, 1652, 1525, 1407, 1216, 827, 716, 644 cm^{-1}; R_f 0.41 (hexane/ethyl acetate 3:1).

***N*-(2-bromophenyl)benzamide (4b):**[19g] white solid (177 mg, 64%); ^1H NMR (600 MHz, $CDCl_3$) δ 8.56 (dd, *J* = 8.4, 1.2 Hz, 1H), 8.47 (bs, 1H), 7.94 (d, *J* = 8.4 Hz, 2H), 7.63–7.55 (m, 2H), 7.55–7.49 (m, 2H), 7.38 (ddd, *J* = 8.4, 7.2, 1.2 Hz, 1H), 7.02 (ddd, *J* = 7.8, 7.2, 1.2 Hz, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 165.5, 136.0, 134.8, 132.5, 132.4, 129.2, 128.8, 127.3, 125.5, 122.0, 113.9. IR (neat): ν_{max} = 3280, 1653, 1529, 1435, 1308, 750, 706 cm^{-1}; R_f 0.51 (hexane/ethyl acetate 5:1).

***N*-(4-fluorophenyl)benzamide (4c):**[5b] white solid (142 mg, 66%); ^1H NMR (600 MHz, DMSO-d_6) δ 10.22 (bs, 1H), 7.97–7.90 (m, 2H), 7.79 (dd, J_{H-F} = 9 Hz, J_{H-H} = 5.4 Hz, 2H), 7.53 (tt, *J* = 7.2, 1.6 Hz, 1H), 7.48 (m, 2H), 7.11–7.05 (m, 2H). ^{13}C NMR (151 MHz, DMSO-d_6) δ 163.9, 156.8 (d, *J* = 241.6), 133.9, 133.3, 129.8, 126.6, 126.0, 120.6, 113.3 (d, *J* = 22.65). IR (neat): ν_{max} = 3343, 1654, 1537, 1438, 750, 690 cm^{-1}; R_f 0.45(hexane/ethyl acetate 3:1).

***N*-(2-bromopyridin-3-yl)benzamide (4d):**[19a] white solid (161 mg, 58%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.86 (dd, *J* = 8.4, 1.8 Hz, 1H), 8.49 (bs, 1H), 8.13 (dd, *J* = 4.8, 1.8 Hz, 1H), 7.93 (dd, *J* = 8.4, 1.2 Hz, 2H), 7.60 (tt, *J* = 7.5, 1.6 Hz, 1H), 7.53 (dd, *J* = 7.8, 7.7 Hz, 2H), 7.32 (dd, *J* = 7.8, 4.8 Hz, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 165.7, 144.8, 134.0, 133.9, 133.6, 132.8, 130.3, 129.3, 128.9, 127.3, 123.9. IR (neat): ν_{max} = 3403, 1684, 1508, 1488, 1381, 1297, 1048, 706 cm^{-1}; R_f 0.34 (hexane/ethyl acetate 3:1).

***N*-(4-methoxybenzyl)benzamide (4e):**[4f] white solid (198 mg, 82%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.78 (d, *J* = 8.4 Hz, 2H), 7.48 (tt, *J* = 7.5, 1.4 Hz, 1H), 7.41 (dd, *J* = 7.8, 7.7 Hz, 2H), 7.28 (dd, *J* = 8.4, 6.6 Hz, 2H), 6.88 (d, *J* = 8.4 Hz, 2H), 6.40 (s, 1H), 4.57 (d, *J* = 6.0 Hz, 2H), 3.80 (s, 3H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 167.5, 159.3, 134.7, 131.7, 130.5, 129.5, 128.8, 127.1, 114.4, 55.5, 43.9. IR (neat): ν_{max} = 3308, 3062, 1636, 1534, 1511, 1301, 1246, 1033, 695 cm^{-1}; R_f 0.36 (hexane/ethyl acetate 2:1).

***N*-(4-fluorobenzyl)benzamide (4f):**[19c] white solid (181 mg, 79%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.81 – 7.77 (m, 2H), 7.51 (tt, *J* = 7.5, 1.4 Hz, 1H), 7.44 (dd, *J* = 7.8, 7.7 Hz, 2H), 7.33 (dd, *J* = 8.4, 5.4 Hz, 2H), 7.04 (dd, *J* = 8.7, 2H), 6.41 (bs, 1H), 4.62 (d, *J* = 5.4 Hz, 2H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 167.5, 163.5 (d, *J* = 326 Hz), 134.4 (d, *J* = 48 Hz), 131.9, 129.8 (d, *J* = 10 Hz), 128.9, 127.1, 115.9, 115.8, 43.6. IR (neat): ν_{max} = 3310, 1637, 1541, 1510, 1222, 692 cm^{-1}; R_f 0.43 (hexane/ethyl acetate 2:1).

***N*-hexylbenzamide (4g):**[7d] colorless liquid (150 mg, 73%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.76 (dd, *J* = 8.4, 7.2 Hz, 2H), 7.44 (tt, *J* = 7.2, 1.7 Hz, 1H), 7.36 (dd, *J* = 7.8 Hz, 2H), 6.60 (bs, 1H), 3.40 (td, *J* = 7.2, 6.0 Hz, 2H), 1.57 (tt, *J* = 7.2 Hz, 2H), 1.40–1.30 (m, 2H), 1.30–1.23 (m, 4H), 0.86 (t, *J* = 7.2 Hz, 3H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 167.8, 135.0, 131.3, 128.6, 127.0, 40.3, 31.6, 29.7, 26.8, 22.7, 14.1. IR (neat): ν_{max} = 3310, 1635, 1540, 1490, 1309, 693 cm^{-1}; R_f 0.40 (hexane/ethyl acetate 3:1).

***N*-(2-(diethylamino)ethyl)benzamide (4h):**[19c] colorless liquid (134 mg, 61%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.77 (bs, 1H), 8.01 (dd, *J* = 8.4, 1.2 Hz, 2H), 7.44 (tt, *J* = 7.5, 1.6 Hz, 1H), 7.39 (dd, *J* = 7.2, 6.0 Hz, 2H), 3.84 (dd, *J* = 10.8, 6.0 Hz, 2H), 3.22 (t, *J* = 5.4 Hz, 2H), 3.11 (q, *J* = 7.2 Hz, 4H), 1.34 (t, *J* = 7.2 Hz, 6H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 167.9, 133.3, 132.0, 128.7, 127.7, 53.0, 48.5, 36.0, 9.1. IR (neat): ν_{max} = 3363, 1637, 1539, 1488, 1308, 694 cm^{-1}; R_f 0.42 (ethyl acetate/methanol 2:1).

phenyl(piperidin-1-yl)methanone (4i):[5e] colorless liquid (119 mg, 63%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.43–7.35 (m, 5H), 3.77–3.65 (m, 2H), 3.40–3.28 (m, 2H), 1.76–1.60 (m, 4H), 1.59 – 1.41 (m, 2H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 170.6, 136.5, 129.5, 128.5, 127.0, 49.0, 43.4, 26.7, 25.8, 24.7. IR (neat): ν_{max} = 1713, 1625, 1444, 1274, 1003, 707 cm^{-1}; R_f 0.37 (hexane/ethyl acetate 2:1).

***N,N*-dibutylbenzamide (4j):**[7a] colorless liquid (145 mg, 62%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.38 – 7.30 (m, 5H), 3.48 (t, *J* = 7.8 Hz, 2H), 3.18 (t, *J* = 7.8 Hz, 2H), 1.65 (tt, *J* = 7.8 Hz, 2H), 1.46 (tt, *J* = 7.8 Hz, 2H), 1.43–1.32 (m, 2H), 1.21-1.05 (m, 2H), 0.97 (t, *J* = 7.2 Hz, 3H), 0.76 (t, *J* = 7.2 Hz, 3H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 171.4, 137.2, 128.8, 128.1, 126.2, 48.6, 44.3, 30.6, 29.5, 20.1, 19.5, 13.7, 13.4. IR (neat): ν_{max} = 2956, 1627, 1421, 1296, 1100, 698 cm^{-1}; R_f 0.49 (hexane/ethyl acetate 5:1).

4-chloro-*N*-(2-morpholinoethyl)benzamide (6):[16] white solid (207 mg, 77%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.71 (d, *J* = 8.4 Hz, 2H), 7.40 (d, *J* = 8.4 Hz, 2H), 6.79 (bs, 1H), 3.75–3.65 (m, 4H), 3.58–3.50 (m, 2H), 2.63–2.56 (m, 2H), 2.57–2.43 (m, 4H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 166.5, 137.8, 133.2, 129.0, 128.6, 67.1, 57.0, 53.5, 36.3. IR (neat): ν_{max} = 3309, 1646, 1541, 1487, 1312, 1116, 1093, 860 cm^{-1}; R_f 0.50 (ethyl acetate/methanol 4:1).

2-(4-(4-(2-methoxyphenyl)piperazin-1-yl)butyl)isoindoline-1,3-dione (9):[17b] Yellowish solid; ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.82 (dd, *J* = 5.4, 3.0 Hz, 2H), 7.69 (dd, *J* = 5.4, 3.0 Hz, 2H), 6.97 (ddd, *J* = 7.8, 7.2, 2.4 Hz, 1H), 6.92 (dd, *J* = 7.8, 2.4 Hz, 1H), 6.89 (ddd, *J* = 7.5, 1.4 Hz, 1H)6.83 (dd, *J* = 7.8, 1.2 Hz, 1H), 3.84 (s, 3H), 3.71 (t, *J* = 7.8 Hz, 2H), 3.21–2.95 (m, 4H), 2.73–2.55 (m, 4H), 2.44 (t, *J* = 7.8 Hz, 2H), 1.72 (tt, *J* = 7.8 Hz, 2H), 1.57 (tt, *J* = 7.8 Hz, 2H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 168.6, 152.4, 141.5, 134.0, 132.3, 123.3, 123.0, 121.1, 118.3, 111.3, 58.2, 55.4, 53.5, 50.7, 38.0, 26.7, 24.3. IR (neat): ν_{max} = 3280, 1653, 1529, 1435, 1308, 750, 706 cm^{-1}; R_f 0.68 (ethyl acetate/methanol 10:1).

4-(4-(2-methoxyphenyl)piperazin-1-yl)butan-1-amine (10):[17b] greenish yellow viscous liquid; ^{1}H NMR (600 MHz, $CDCl_3$) δ 6.98 (dd, *J* = 7.8, 7.2 Hz, 1H), 6.94 (d, *J* = 7.6 Hz, 1H), 6.91 (dd, *J* = 7.8, 7.2 Hz, 1H), 6.85 (d, *J* = 7.8 Hz, 1H), 3.85 (s, 3H), 3.25–3.0 (m, 4H), 2.73 (t, *J* = 7.2 Hz, 2H), 2.70–2.58 (m, 4H), 2.42 (t, *J* = 7.2 Hz, 2H), 2.33 (bs, 2H), 1.58 (tt, *J* = 7.2 Hz, 2H), 1.51 (tt, *J* = 7.2 Hz, 2H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 152.5, 141.5, 123.1, 121.2, 118.4, 111.4, 58.7, 55.6, 53.6, 50.8, 42.0, 31.6, 24.6. IR (neat): ν_{max} = 1499, 1450, 1238, 1118, 1025, 746 cm^{-1}; R_f 0.52 (chloroform/methanol/ammonia solution 85:13:2).

4-bromo-N-(4-(4-(2-methoxyphenyl)piperazin-1-yl)butyl)benzamide (11):[17b] light green solid (290 mg, 65%); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.63 (d, *J* = 8.4 Hz, 2H), 7.54 (d, *J* = 8.4 Hz, 2H), 7.0 (dd, *J* = 7.6 Hz, 1H), 6.92 (dd, *J* = 7.6 Hz, 1H), 6.87 (d, *J* = 7.8 Hz, 1H), 6.85(d, *J* = 7.8 Hz, 1H), 3.85 (s, 3H), 3.46 (td, *J* = 6.0 Hz, 2H), 3.15–2.95 (m, 4H), 2.70–2.55 (m, 4H), 2.46 (t, *J* = 7.2 Hz, 2H), 1.72 – 1.58 (m, 4H), ^{13}C NMR (151 MHz, $CDCl_3$) δ 167.0, 152.4, 141.3, 134.1, 131.9, 128.8, 126.0, 123.2, 121.2, 118.36, 111.38, 58.2, 55.6, 53.61, 50.63, 40.3, 27.6, 24.6. IR (neat): ν_{max} = 3289, 1634, 1541, 1499, 1239, 1010, 729 cm^{-1}; R_f 0.45 (ethyl acetate/methanol 4:1).

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SUPPORTING INFORMATION

Characterization and spectral data of compounds are available free of charge via the Internet at http://pubs.acs.org.

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