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Iodine-mediated Domino Oxidative Cyclization: One-pot Synthesis of 1,3,4-Oxadiazoles via Oxidative Cleavage of C(sp^2)-H or C(sp)-H Bond

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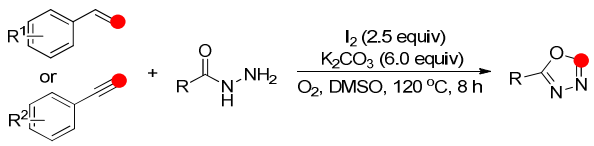


Abstract: An I_2 promoted, metal-free domino protocol for one-pot synthesis of 1,3,4-oxadiazoles has been developed via oxidative cleavage of C(sp^2)-H or C(sp)-H bond, followed by cyclization and deacylation. In this reaction, the use of K_2CO_3 as a base is found to be an essential factor in the cyclization and the C-C bond cleavage. This procedure proceeded smoothly in moderate to high yields with good functional-group compatibility.

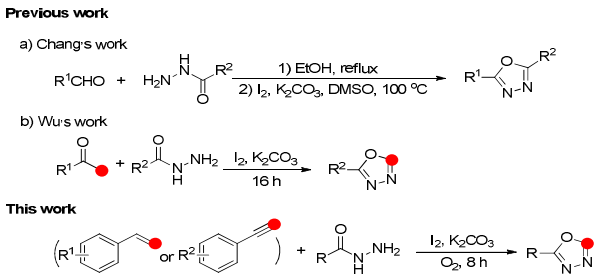
Domino strategy, due to its applicability to various types of reactions like radical,[1] pericyclic,[2] photochemical,[3] and transition metal-mediated[4] reaction, has been extensively examined for the synthesis of organic compounds in modern synthetic chemistry.[5] In the past decade, significant progress has been achieved in developing domino reactions. For example, Jiang and co-workers have developed a series of domino protocols to construct heterocycle compounds.[6] Wu's group have successfully led to various different compounds by the use of the

multiplicative effect of a coupled domino strategy.[7] In addition, the efficiency and significance of domino strategy for one pot synthesis of 2-acylbenzothiazoles from multiform substrates were also demonstrated.[7b] Inspired by these studies, we present a route through domino cyclization for the formation of 1,3,4-oxadiazoles using simple and commercially available reagents via oxidative cleavage of C(sp^2)-H or C(sp)-H bond.

1,3,4-oxadiazoles are non-naturally occurring five-membered aromatic heterocycles which have been widely used in many compounds with biological and pharmaceutical activities,[8] such as antifungal, antiviral, and antibacterial properties.[9] Consequently, a number of methods have been reported for the synthesis of the 1,3,4-oxadiazoles skeleton via oxidative cyclization of N-acylhydrazones[10] or the dehydrative cyclization of 1,2-diacylhydrazines.[11] In 2013, Chang's group described an I_2-mediated oxidative C-O bond formation for the synthesis of 1,3,4-oxadiazoles from aldehydes and hydrazides (Scheme 1, a).[9] Subsequently, Wu's group demonstrated a direct annulation of hydrazides to 1,3,4-oxadiazoles via oxidative C(CO)-C(Methyl) bond cleavage of methyl ketones (Scheme 1, b),[12] where a wide range of ketones and hydrazides were used as substrates, showing good functional group tolerance and high selectivity. Furthermore, Cu-catalyzed decarboxylative coupling domino reaction for the formation of 2-(1,3,4-oxadiazol-2-yl)

anilines derivatives[13] was disclosed. However, to our knowledge, I_2-mediated oxidative cyclization for the synthesis of 1,3,4-oxadiazoles from hydrazides and styrene or phenyl acetylene has not yet been reported. Herein, we choose hydrazides and styrene or phenyl acetylene derivatives as the substrates to synthesize 1,3,4-oxadiazoles in the presence of O_2 as the oxidant and K_2CO_3 as the base, achieving the desired products in moderate to good yields.

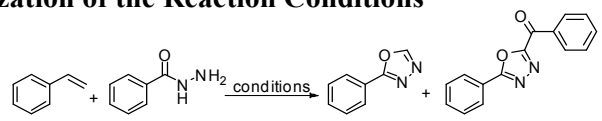
Scheme 1. Synthetic approaches to 1,3,4-oxadiazoles



Initial investigation was conducted by employing reaction of **2a** (0.2 mmol) with **1a** (0.6 mmol) in the presence of I_2 (0.5 mmol), and K_2CO_3 (0.6 mmol) in DMSO (2.0 mL) under air at 120 ^oC for 8 h, resulting in 6% yield of **3aa** (Table 1, entry 1). To identify the appropriate conditions for the reaction, firstly, various oxidants were examined, such as TBHP, DDQ, IBX, $PhI(OAc)_2$, and dioxygen (1 atm). These results illustrated that dioxygen displayed the best ability in this transformation, and a 34% yield of **3aa** was obtained (Table 1, entries 2-6). Meanwhile, the yield of side product **4aa** was also increased to 50% when dioxygen (1 atm) was used as the oxidant. In order to improve the yield of **3aa**, different bases

were tested, including K_2CO_3, NaOH, *t*-BuOK, K_3PO_4, Cs_2CO_3, and $NaHCO_3$. The results promoted us to use K_2CO_3 to conduct this reaction (Table 1, entries 6-11). After screening the amount of K_2CO_3, we found that the substrate showed the highest activity for this process when 6.0 equiv K_2CO_3 was added, affording **3aa** in 85% yield (Table 1, entry 16). Then different solvents were also evaluated for this conversion, and DMSO effectively facilitated the reaction. Additionally, changing the amount of iodine did not give better result (Table 1, entries 19-23).

Table 1. Optimization of the Reaction Conditions[a]



1a **2a** **3aa** **4aa**

Entry	Oxidant	I_2 (equiv)	Base (equiv)	Solvent	Yield**3aa**[b]/**4aa** (%)
1[c]	-	2.5	K_2CO_3(3.0)	DMSO	6/38
2[d]	TBHP	2.5	K_2CO_3(3.0)	DMSO	17/40
3[d]	DDQ	2.5	K_2CO_3(3.0)	DMSO	0/10
4[d]	IBX	2.5	K_2CO_3(3.0)	DMSO	10/34
5[d]	$PhI(OAc)_2$	2.5	K_2CO_3(3.0)	DMSO	7/38
6	O_2	2.5	K_2CO_3(3.0)	DMSO	34/50
7	O_2	2.5	NaOH(3.0)	DMSO	0/0
8	O_2	2.5	t-BuOK(3.0)	DMSO	0/0
9	O_2	2.5	K_3PO_4(3.0)	DMSO	21/65
10	O_2	2.5	Cs_2CO_3(3.0)	DMSO	27/40
11	O_2	2.5	$NaHCO_3$(3.0)	DMSO	0/7
12	O_2	2.5	K_2CO_3(1.0)	DMSO	0/0
13	O_2	2.5	K_2CO_3(2.0)	DMSO	5/50
14	O_2	2.5	K_2CO_3(4.0)	DMSO	68/0
15	O_2	2.5	K_2CO_3(5.0)	DMSO	75/0
16	**O_2**	**2.5**	**K_2CO_3(6.0)**	**DMSO**	**85/0**
17	O_2	2.5	K_2CO_3(6.0)	DMF	0/0
18	O_2	2.5	K_2CO_3(6.0)	Toluene	0/0
19	O_2	0.5	K_2CO_3(6.0)	DMSO	0/0
20	O_2	1.0	K_2CO_3(6.0)	DMSO	0/0
21	O_2	1.5	K_2CO_3(6.0)	DMSO	41/6
22	O_2	2.0	K_2CO_3(6.0)	DMSO	51/8

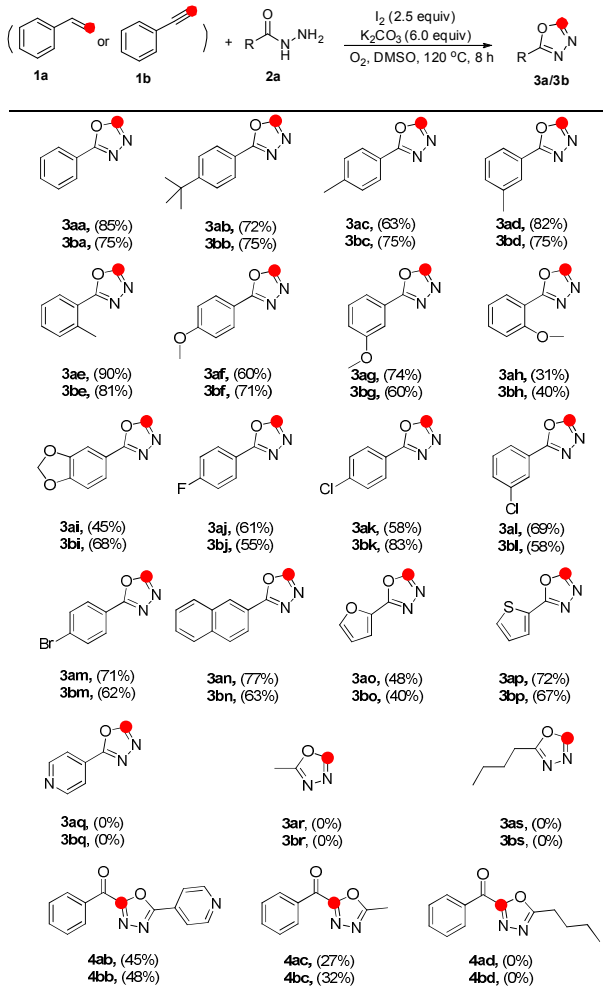
23	O_2	3.0	K_2CO_3(6.0)	DMSO	68/6

[a]Reaction conditions: **1a** (0.6 mmol), **2a** (0.2 mmol) in DMSO (2.0 mL) at 120 °C for 8 h. Dioxygen (1 atm) was used. [b]Yields of isolated products. [c]Open air. [d]Oxidant (1.5 equiv). TBHP = terbutylhydroperoxide (5.0-6.0 M in decane), DDQ = 2,3-dichloro-5,6-dicyanobenzoquinone, IBX = 2-iodoxybenzoic acid, DMSO = demethyl sulfoxide, DMF = N,N-dimethyl formamide.

With the optimized conditions in hand, the reaction scope was tested using styrene (**1a**) with different hydrazides (Table 2). A variety of hydrazides proceeded smoothly to afford the corresponding products in moderate to high yields (31-90%). Various functional groups on the aryl ring of the hydrazides demonstrated that both electron-withdrawing and electron-donating groups, including methyl, *tert*-butyl, methoxy, 3,4-methylenedioxy, fluoro, chloro and bromo, were compatible well with this reaction (**3aa-3am**). 2-Naphthyl hydrazine also exhibited good reactivity to give the desired product (**3an**) in 77% yield. Moreover, heteroaryl hydrazides like furanyl and thienyl hydrazides could undergo the tandem reaction smoothly to receive the expected products **3ao** and **3ap** in 48% and 72% yields. Unexpectedly, when we used pyridyl hydrazide as the substrate, the desired product **3aq** was not detected, but the undeacylative by-product **4ab** was obtained in 45% yield. In addition, alkyl hydrazides, such as acetohydrazide and valeric acid hydrazide, were also investigated. However, only the acetohydrazide could proceed to afford the undeacylative product **4ac** in 27% yield; the substrate of valetric acid hydrazide was failed to accomplish this reaction (**3as** and **4ad**).

Subsequently, we explored the limitations of different hydrazides reacting with phenyl acetylene (**1b**). As expected, we were pleased to find that the reaction of **1b** with a range of hydrazides transformed well under the optimized conditions to give the corresponding 1,3,4-oxadiazoles products (Table 2, **3ba-3bp**) in 40-83% yields. Disappointedly, pyridyl hydrazide, acetohydrazide and valeric acid hydrazide did not give desired products (**3bq-3bs**).

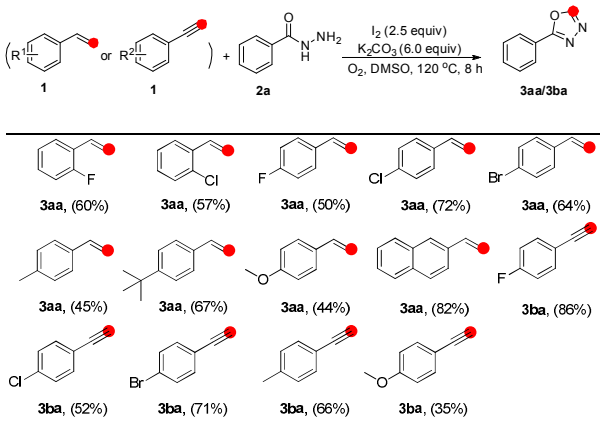
Table 2. Synthesis of 1,3,4-oxadiazoles from styrene or phenyl acetylene and hydrazides[a]



[a]Reaction conditions: **2a** (0.2 mmol), **1a** or **1b** (0.6 mmol), I_2 (0.5 mmol) and K_2CO_3 (1.2 mmol) in DMSO (2.0 mL) at 120 °C under dioxygen (1 atm) for 8 h.

To further expand the scope of the substrates, various styrene and phenyl acetylene derivatives were carried out under the standard conditions. As shown in Table 3, the results indicated that substituents at different positions of the aryl ring did not affect the efficiency obviously. Further studies proved that either electron-withdrawing or electron–donating groups of **1a** and **1b** all can be converted to the corresponding products in moderate to good yields (35-86%). Vinylnaphthalene was also tolerated in this transformation to produce the desired 2-phenyl-1,3,4-oxadiazole in 82% yield.

Table 3. Synthesis of 2-phenyl-1,3,4-oxadiazole from various styrene or phenyl acetylene and benzoylhydrazide[a]

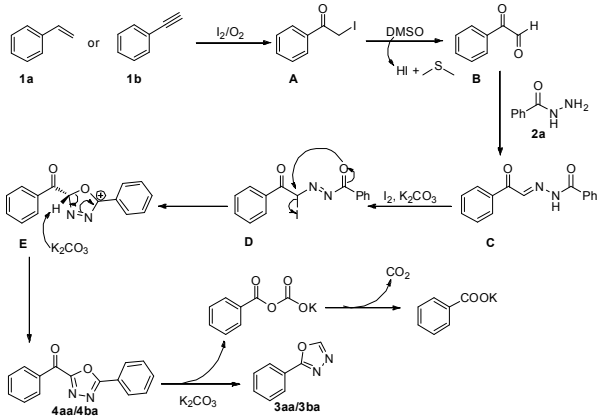


[a]Reaction conditions: **2a** (0.2 mmol), **1a** or **1b** (0.6 mmol), I_2 (0.5 mmol) and K_2CO_3 (1.2 mmol) in DMSO (2.0 mL) at 120 °C under dioxygen (1 atm) for 8 h.

On the basis of the above results and literature reports,[5a,9,12,14] the proposed mechanism for the synthesis of 1,3,4-oxadiazoles is outlined in Scheme 2. Using **1a** or **1b** and **2a** as model substrates, initially, **1a** or **1b** is converted into phenacyl iodide (**A**)[5a,12,14] through consecutive

iodination and oxidation under I_2/O_2, then **A** is further converted into phenylglyoxal (**B**)[5a,12,14] by a subsequent Kornblum oxidation in DMSO.[7b] Benzoylhydrazide (**2a**) reacts with **B to** generate the C-acyl benzoylhydrazone (**C**).[12] Subsequently, K_2CO_3-promoted oxidative iodination of **C** gives an iodide intermediate **D,**[9,12] which would be converted to **E** via a S_N2'-type[9,12] cyclization, with a new C-O bond formed. Consequently, **4aa/4ba** is formed through subsequent deprotonation by base.[9] Finally, **4aa/4ba** would go through deacylation to generate the product **3aa/3ba** with the aid of K_2CO_3.[12]

Scheme 2. Possible mechanism



In summary, we have developed an I_2-mediated direct oxidative cyclization reaction for the synthesis of substituted 1,3,4-oxadiazoles in a one-pot manner. This procedure employs dioxygen as oxidant and K_2CO_3 as base. In addition, K_2CO_3 plays an important role in the cyclization and deacylation. This reaction system has broad substrates scope, providing a facile pathway for the synthesis of 1,3,4-oxadiazoles.

Experimental Section

General remarks. ^{1}H NMR spectra were obtained at 400 MHz in $CDCl_3$ with tetramethylsilane (δ = 0.00 ppm) as an internal standard. ^{13}C NMR spectra were recorded at 100 MHz and were calibrated with $CDCl_3$ (δ = 77.00 ppm). The high-resolution mass spectra (HRMS) were recorded on an FT-ICR mass spectrometer using electrospray ionization (ESI). The HRMS data and melting points for the same compounds that have been prepared by two methods were obtained only by the samples of **3aa-3ap** and **4aa-4ac**. Products were purified by flash chromatogrgraphy on 200-300 mesh silica gels. All melting points were determined without correction. Unless otherwise noted, commercially reagents were used without further purification.

General procedure for the synthesis of 3 (3aa/3ba as an example)

A test tube was charged with styrene or phenyl acetylene (0.6 mmol), I_2 (0.5 mmol) in DMSO (2.0 mL). The mixture was stirred at 120 ^{o}C for 6 h under 1 atm of dioxygen. After disappearance of the reactant, then added benzoylhydrazide (0.2 mmol), K_2CO_3 (1.2 mmol), and the mixture was heated at 120 ^{o}C for 2 h under 1 atm of dioxygen. Upon completion, the mixture was diluted with water and extracted with EtOAc (3×20 mL). The extract was washed with 10% $Na_2S_2O_3$ solution (w/w), dried over anhydrous Na_2SO_4 and evaporation. The residue was purified by column chromatography on silica gel (petroleum ether/EtOAc = 5:1) to yield the

desired product **3aa/3ba**.

2-phenyl-1,3,4-oxadiazole (**3aa**). Yellow solid. Yield 85% (24.8 mg). m. p. 33-34 °C (lit.[15] m. p. 34-36 °C); ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.50 (s, 1H), 8.11 – 8.07 (m, 2H), 7.59 – 7.50 (m, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.7, 152.6, 132.0, 129.1, 127.0, 123.4. HRMS (ESI) (*m/z*) calcd for $C_8H_6N_2O$ $[M+H]^+$ 147.0553; found 147.0556.

2-(4-(tert-butyl)phenyl)-1,3,4-oxadiazole (3ab). Light yellow solid. Yield 72% (29.1 mg). m. p. 95-98 °C (lit.[12] m. p. 94-97 °C); ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.46 (s, 1H), 8.04 – 8.00 (m, 2H), 7.56 – 7.53 (m, 2H), 1.36 (s, 9H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.8, 155.6, 152.4, 126.9, 126.1, 120.6, 35.0, 31.1. HRMS (ESI) (*m/z*) calcd for $C_{12}H_{14}N_2O$ $[M+H]^+$ 203.1179; found 203.1181.

2-(p-tolyl)-1,3,4-oxadiazole (3ac). Light yellow solid. Yield 63% (20.2 mg). m. p. 148-150 °C (lit.[12] m. p. 149-151 °C); ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.46 (s, 1H), 7.97 (d, *J* = 8.1 Hz, 2H), 7.32 (dd, *J* = 8.0, 0.5 Hz, 2H), 2.43 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.8, 152.3, 142.5, 129.7, 126.9, 120.6, 21.6. HRMS (ESI) (*m/z*) calcd for $C_9H_8N_2O$ $[M+H]^+$ 161.0746; found 161.0748.

2-(m-tolyl)-1,3,4-oxadiazole (3ad). Yellow oil. Yield 82% (26.2 mg). ^{1}H

NMR (400 MHz, $CDCl_3$): δ = 8.48 (s, 1H), 7.92 (s, 1H), 7.88 (d, *J* = 7.4 Hz, 1H), 7.44 – 7.35 (m, 2H), 2.44 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.9, 152.5, 139.0, 132.8, 129.0, 127.6, 124.2, 123.3, 21.3. HRMS (ESI) (*m/z*) calcd for $C_9H_8N_2O$ $[M+H]^+$ 161.0746; found 161.0747.

2-(o-tolyl)-1,3,4-oxadiazole (3ae). Yellow oil. Yield 90% (28.8 mg). ^1H NMR (400 MHz, $CDCl_3$): δ = 8.51 (s, 1H), 7.95 (d, *J* = 7.8 Hz, 1H), 7.43 (t, *J* = 7.4 Hz, 1H), 7.34 (dd, *J* = 13.1, 7.0 Hz, 2H), 2.72 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.8, 152.2, 138.5, 131.7, 131.4, 129.0, 126.1, 122.5, 22.0. HRMS (ESI) (*m/z*) calcd for $C_9H_8N_2O$ $[M+H]^+$ 161.0746; found 161.0749.

2-(4-methoxyphenyl)-1,3,4-oxadiazole (3af). Yellow solid. Yield 60% (21.1 mg). m. p. 55-57 ^oC (lit.[16] m. p. 61-62 ^oC); ^1H NMR (400 MHz, $CDCl_3$): δ = 8.43 (s, 1H), 8.01 (d, *J* = 8.8 Hz, 2H), 7.02 (d, *J* = 8.8 Hz, 2H), 3.88 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.6, 162.4, 152.1, 128.8, 115.9, 114.5, 55.4. HRMS (ESI) (*m/z*) calcd for $C_9H_8N_2O_2$ $[M+H]^+$ 177.0659; found 177.0661.

2-(3-methoxyphenyl)-1,3,4-oxadiazole (3ag). Light yellow solid. Yield 74% (26.0 mg). m. p. 105-107 ^oC (lit.[12] m. p. 105-107 ^oC); ^1H NMR (400 MHz, $CDCl_3$): δ 8.48 (s, 1H), 7.66 – 7.60 (m, 2H), 7.42 (t, *J* = 8.0 Hz, 1H), 7.09 (dd, *J* = 8.3, 2.4 Hz, 1H), 3.88 (s, 3H); ^{13}C NMR (100 MHz,

$CDCl_3$): δ = 164.6, 159.9, 152.6, 130.2, 124.5, 119.4, 118.4, 111.7, 55.4. HRMS (ESI) (*m/z*) calcd for $C_9H_8N_2O_2$ $[M+H]^+$ 177.0659; found 177.0660.

2-(2-methoxyphenyl)-1,3,4-oxadiazole (3ah). Light yellow solid. Yield 31% (11.0 mg). m. p. 50-52 °C (lit.[12] m. p. 49-51 °C); ^1H NMR (400 MHz, $CDCl_3$): δ 8.51 (s, 1H), 7.96 (dd, *J* = 7.8, 1.7 Hz, 1H), 7.56 – 7.51 (m, 1H), 7.10 (dd, *J* = 12.2, 4.6 Hz, 2H), 3.98 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$): δ 163.4, 157.8, 152.4, 133.3, 130.5, 120.7, 112.4, 111.9, 55.9. HRMS (ESI) (*m/z*) calcd for $C_9H_8N_2O_2$ $[M+H]^+$ 177.0659; found 177.0662.

2-(benzo[d][1,3]dioxol-5-yl)-1,3,4-oxadiazole (3ai). Light yellow solid. Yield 45% (17.0 mg). m. p. 146-148 °C (lit.[12] m. p. 147-149 °C); ^1H NMR (400 MHz, $CDCl_3$): δ = 8.43 (s, 1H), 7.63 (dd, *J* = 8.1, 1.7 Hz, 1H), 7.53 (d, *J* = 1.5 Hz, 1H), 6.93 (d, *J* = 8.2 Hz, 1H), 6.08 (s, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.5, 152.2, 150.8, 148.3, 122.2, 117.2, 108.9, 107.1, 101.9. HRMS (ESI) (*m/z*) calcd for $C_9H_6N_2O_3$ $[M+H]^+$ 191.0451; found 191.0453.

2-(4-fluorophenyl)-1,3,4-oxadiazole (3aj). Light yellow solid. Yield 61% (20.0 mg). m. p. 72-74 °C (lit.[17] m. p. 121.0-122.0 °C); ^1H NMR (400 MHz, $CDCl_3$): δ = 8.48 (s, 1H), 8.11 (dd, *J* = 8.9, 5.2 Hz, 2H), 7.23

(t, *J* = 8.6 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 165.1 (d, J = 223 Hz), 152.6, 129.4 (d, *J* = 8.9 Hz), 119.9 (d, *J* = 3.3 Hz), 116.6, 116.4. HRMS (ESI) (*m/z*) calcd for $C_8H_5FN_2O$ $[M+H]^+$ 165.0459; found 165.0460.

2-(4-chlorophenyl)-1,3,4-oxadiazole (3ak). Light yellow solid. Yield 58% (20.9 mg). m. p. 134-136 °C (lit.[16] 134-135 °C); ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.51 (s, 1H), 8.03 (dd, *J* = 8.6, 2.0 Hz, 2H), 7.51 (dd, *J* = 8.6, 1.9 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 163.9, 152.7, 138.2, 129.4, 128.3, 121.8. HRMS (ESI) (*m/z*) calcd for $C_8H_5ClN_2O$ $[M+H]^+$ 181.0163; found 181.0166.

2-(3-chlorophenyl)-1,3,4-oxadiazole (3al). Light yellow solid. Yield 69% (24.8 mg). m. p. 79-82 °C (lit.[18] 115.3 °C); ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.51 (s, 1H), 8.09 (d, *J* = 1.4 Hz, 1H), 7.99 (dd, *J* = 7.7, 1.1 Hz, 1H), 7.54 (dd, *J* = 5.0, 3.9 Hz, 1H), 7.48 (t, *J* = 7.9 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 163.6, 152.8, 135.2, 132.1, 130.5, 127.1, 125.2, 125.0. HRMS (ESI) (*m/z*) calcd for $C_8H_5ClN_2O$ $[M+H]^+$ 181.0163; found 181.0167.

2-(4-bromophenyl)-1,3,4-oxadiazole (3am). Light yellow solid. Yield 71% (32.0 mg). m. p. 139-142 °C (lit.[12] m. p. 140-143 °C); ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.51 (s, 1H), 7.95 (d, *J* = 8.6 Hz, 2H), 7.67 (d, *J* = 8.6

Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.0, 152.7, 132.4, 128.4, 126.7, 122.2. HRMS (ESI) (*m/z*) calcd for $C_8H_5BrN_2O$ $[M+H]^+$ 224.9658; found 224.9662.

2-(naphthalen-2-yl)-1,3,4-oxadiazole (3an). Light yellow solid. Yield 77% (30.2 mg). m. p. 58-60 °C (lit.[19] m. p. 60-62 °C); ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.57 (s, 1H), 8.52 (s, 1H), 8.16 – 8.12 (m, 1H), 7.95 (dd, *J* = 7.0, 4.5 Hz, 2H), 7.89 (d, *J* = 7.6 Hz, 1H), 7.62 – 7.54 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.9, 152.6, 134.7, 132.7, 129.0, 128.8, 128.0, 127.9, 127.6, 127.1, 123.1, 120.6. HRMS (ESI) (*m/z*) calcd for $C_{12}H_8N_2O$ $[M+H]^+$ 197.0710; found 197.0714.

2-(furan-2-yl)-1,3,4-oxadiazole (3ao). Yellow oil. Yield 48% (13.0 mg). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.45 (s, 1H), 7.68 (d, *J* = 1.0 Hz, 1H), 7.22 (d, *J* = 3.5 Hz, 1H), 6.63 (dd, *J* = 3.5, 1.8 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 157.5, 151.8, 146.0, 139.0, 114.6, 112.2. HRMS (ESI) (*m/z*) calcd for $C_6H_4N_2O_2$ $[M+H]^+$ 137.0346; found 137.0349.

2-(thiophen-2-yl)-1,3,4-oxadiazole (3ap). Yellow solid. Yield 72% (21.9 mg). m. p. 150-152 °C (lit.[12] m. p. 151-154 °C); ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.45 (s, 1H), 7.80 (dd, *J* = 2.9, 2.0 Hz, 1H), 7.59 (dd, *J* = 5.0, 1.2 Hz, 1H), 7.19 (ddd, *J* = 5.1, 3.7, 1.6 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 160.9, 151.9, 130.5, 130.1, 128.1, 124.5. HRMS (ESI) (*m/z*)

calcd for $C_6H_4N_2OS$ $[M+H]^+$ 153.0117; found 153.0119.

Phenyl(5-phenyl-1,3,4-oxadiazol-2-yl)methanone (4aa). Light yellow solid. Yield 65% (33.0 mg). m. p. 95-97 ^oC (lit.[12] m. p. 95-97 ^oC); ^1H NMR (400 MHz, $CDCl_3$): δ = 8.62 – 8.55 (m, 2H), 8.28 – 8.20 (m, 2H), 7.72 (dd, *J* = 10.5, 4.3 Hz, 1H), 7.65 – 7.54 (m, 5H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 177.6, 166.0, 160.9, 134.9, 134.3, 132.9, 130.9, 129.2, 128.8, 127.8, 122.8. HRMS (ESI) (*m/z*) calcd for $C_{15}H_{10}N_2O_2$ $[M+H]^+$ 251.0815; found 251.0819.

Phenyl(5-(pyridine-4-yl)-1,3,4-oxadiazol-2-yl)methanone (4ab). Light yellow solid. Yield 45% (23.0 mg). m. p. 148-150 ^oC (lit.[12] m. p. 148-151 ^oC); ^1H NMR (400 MHz, $CDCl_3$): δ = 8.89 (d, *J* = 5.9 Hz, 2H), 8.60 – 8.54 (m, 2H), 8.07 (dd, *J* = 4.5, 1.5 Hz, 2H), 7.73 (t, *J* = 7.4 Hz, 1H), 7.59 (t, *J* = 7.8 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 177.2, 164.0, 161.2, 151.0, 135.2, 133.9, 130.9, 130.0, 128.9, 120.9. HRMS (ESI) (*m/z*) calcd for $C_{14}H_9N_3O_2$ $[M+H]^+$ 252.0732; found 252.0736.

(5-methyl-1,3,4-oxadiazol-2-yl)(phenyl)methanone (4ac). Brown solid. Yield 27% (10.0 mg). m. p. 107-109 ^oC (lit.[12] m. p. 106-109 ^oC); ^1H NMR (400 MHz, $CDCl_3$): δ = 8.53 (d, *J* = 7.4 Hz, 2H), 7.70 (t, *J* = 7.4 Hz, 1H), 7.56 (t, *J* = 7.8 Hz, 2H), 2.70 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 177.6, 165.7, 161.4, 134.8, 134.0, 130.9, 128.7, 11.1. HRMS (ESI) (*m/z*)

calcd for $C_{10}H_8N_2O_2$ $[M+H]^+$ 189.0659; found 189.0660.

2-phenyl-1,3,4-oxadiazole (3ba). Yellow solid. Yield 75% (21.9 mg). m. p. 33-34 ^oC; ^1H NMR (400 MHz, $CDCl_3$): δ = 8.47 (s, 1H), 8.09 (dd, *J* = 8.1, 1.6 Hz, 2H), 7.58 – 7.51 (m, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.8, 152.6, 132.0, 129.1, 127.1, 123.5. HRMS (ESI) (*m/z*) calcd for $C_8H_6N_2O$ $[M+H]^+$ 147.0553; found 147.0556.

2-(4-(tert-butyl)phenyl)-1,3,4-oxadiazole (3bb). Light yellow solid. Yield 75% (30.3 mg). m. p. 95-98 ^oC; ^1H NMR (400 MHz, $CDCl_3$): δ = 8.45 (s, 1H), 8.01 (d, *J* = 8.5 Hz, 2H), 7.54 (d, *J* = 8.5 Hz, 2H), 1.36 (s, 9H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.8, 155.6, 152.4, 126.9, 126.1, 120.7, 35.1, 31.1. HRMS (ESI) (*m/z*) calcd for $C_{12}H_{14}N_2O$ $[M+H]^+$ 203.1179; found 203.1181.

2-(p-tolyl)-1,3,4-oxadiazole (3bc). Light yellow solid. Yield 75% (24.0 mg). m. p. 148-150 ^oC; ^1H NMR (400 MHz, $CDCl_3$): δ = 8.45 (s, 1H), 7.96 (d, *J* = 8.2 Hz, 2H), 7.32 (d, *J* = 8.0 Hz, 2H), 2.43 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.8, 152.3, 142.5, 129.7, 127.0, 120.7, 21.6. HRMS (ESI) (*m/z*) calcd for $C_9H_8N_2O$ $[M+H]^+$ 161.0746; found 161.0748.

2-(m-tolyl)-1,3,4-oxadiazole (3bd). Yellow oil. Yield 75% (24.0 mg). ^1H

NMR (400 MHz, $CDCl_3$): δ = 8.46 (s, 1H), 7.92 (s, 1H), 7.87 (d, *J* = 7.4 Hz, 1H), 7.43 – 7.34 (m, 2H), 2.44 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.9, 152.5, 139.0, 132.8, 129.0, 127.6, 124.2, 123.3, 21.3. HRMS (ESI) (*m/z*) calcd for $C_9H_8N_2O$ $[M+H]^+$ 161.0746; found 161.0747.

2-(o-tolyl)-1,3,4-oxadiazole (3be). Yellow oil. Yield 81% (25.9 mg).^1H NMR (400 MHz, $CDCl_3$): δ = 8.50 (s, 1H), 7.95 (d, *J* = 7.9 Hz, 1H), 7.43 (dd, *J* = 10.8, 4.2 Hz, 1H), 7.34 (dd, *J* = 13.6, 7.2 Hz, 2H), 2.72 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.9, 152.2, 138.6, 131.7, 131.4, 129.1, 126.2, 122.6, 22.0. HRMS (ESI) (*m/z*) calcd for $C_9H_8N_2O$ $[M+H]^+$ 161.0746; found 161.0749.

2-(4-methoxyphenyl)-1,3,4-oxadiazole (3bf). Yellow solid. Yield 71% (25.0 mg). m. p. 55-57 ^oC; ^1H NMR (400 MHz, $CDCl_3$): δ = 8.43 (s, 1H), 8.02 (d, *J* = 8.8 Hz, 2H), 7.02 (d, *J* = 8.8 Hz, 2H), 3.89 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.6, 162.4, 152.1, 128.8, 115.9, 114.5, 55.4. HRMS (ESI) (*m/z*) calcd for $C_9H_8N_2O_2$ $[M+H]^+$ 177.0659; found 177.0661.

2-(3-methoxyphenyl)-1,3,4-oxadiazole (3bg). Light yellow solid. Yield 60% (21.1 mg). m. p. 105-107 ^oC; ^1H NMR (400 MHz, $CDCl_3$): δ = 8.48 (s, 1H), 7.66 – 7.60 (m, 2H), 7.42 (t, *J* = 8.0 Hz, 1H), 7.11 – 7.07 (m, 1H), 3.88 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.6, 159.9, 152.6, 130.2,

124.5, 119.4, 118.4, 111.7, 55.4. HRMS (ESI) (*m/z*) calcd for $C_9H_8N_2O_2$ $[M+H]^+$ 177.0659; found 177.0660.

2-(2-methoxyphenyl)-1,3,4-oxadiazole (3bh). Light yellow solid. Yield 40% (14.1 mg). m. p. 50-52 ^oC; ^1H NMR (400 MHz, $CDCl_3$): δ = 8.49 (s, 1H), 7.96 (d, *J* = 7.7 Hz, 1H), 7.56 – 7.50 (m, 1H), 7.09 (t, *J* = 8.1 Hz, 2H), 3.97 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 163.4, 157.8, 152.4, 133.3, 130.5, 120.7, 112.4, 111.9, 55.9. HRMS (ESI) (*m/z*) calcd for $C_9H_8N_2O_2$ $[M+H]^+$ 177.0659; found 177.0662.

2-(benzo[d][1,3]dioxol-5-yl)-1,3,4-oxadiazole (3bi). Light yellow solid. Yield 68% (25.6 mg). m. p. 146-148 ^oC; ^1H NMR (400 MHz, $CDCl_3$): δ = 8.43 (s, 1H), 7.62 (dd, *J* = 8.2, 1.7 Hz, 1H), 7.52 (d, *J* = 1.6 Hz, 1H), 6.92 (d, *J* = 8.2 Hz, 1H), 6.07 (s, 2H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.4, 152.2, 150.8, 148.3, 122.2, 117.2, 108.8, 107.1, 101.8. HRMS (ESI) (*m/z*) calcd for $C_9H_6N_2O_3$ $[M+H]^+$ 191.0451; found 191.0453.

2-(4-fluorophenyl)-1,3,4-oxadiazole (3bj). Light yellow solid. Yield 55% (18.0 mg). m. p. 72-74 ^oC; ^1H NMR (400 MHz, $CDCl_3$): δ = 8.48 (s, 3H), 8.11 (dd, *J* = 8.9, 5.2 Hz, 6H), 7.24 (dd, *J* = 16.4, 7.9 Hz, 7H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 165.1 (d, J=223 Hz), 152.6, 129.4 (d, *J* = 8.9 Hz), 119.9 (d, *J* = 3.3 Hz), 116.6, 116.4. HRMS (ESI) (*m/z*) calcd for $C_8H_5FN_2O$ $[M+H]^+$ 165.0459; found 165.0460.

2-(4-chlorophenyl)-1,3,4-oxadiazole (3bk). Light yellow solid. Yield 83% (29.9 mg). m. p. 134-136 ^{o}C; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.49 (s, 1H), 8.03 (d, *J* = 8.6 Hz, 2H), 7.51 (d, *J* = 8.6 Hz, 2H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.0, 152.7, 138.3, 129.5, 128.3, 121.9. HRMS (ESI) (*m/z*) calcd for $C_8H_5ClN_2O$ $[M+H]^+$ 181.0163; found 181.0166.

2-(3-chlorophenyl)-1,3,4-oxadiazole (3bl). Light yellow solid. Yield 58% (20.9 mg). m. p. 79-82 ^{o}C; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.50 (s, 1H), 8.08 (s, 1H), 7.99 (d, *J* = 7.7 Hz, 1H), 7.54 (d, *J* = 8.1 Hz, 1H), 7.47 (t, *J* = 7.9 Hz, 1H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 163.6, 152.8, 135.3, 132.0, 130.5, 127.0, 125.2, 125.1. HRMS (ESI) (*m/z*) calcd for $C_8H_5ClN_2O$ $[M+H]^+$ 181.0163; found 181.0167.

2-(4-bromophenyl)-1,3,4-oxadiazole (3bm) Light yellow solid. Yield 62% (27.9 mg). m. p. 139-142 ^{o}C; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.50 (s, 1H), 7.95 (dd, *J* = 8.4, 1.2 Hz, 2H), 7.69 – 7.63 (m, 2H).^{13}C NMR (100 MHz, $CDCl_3$): δ = 164.0, 152.7, 132.4, 128.4, 126.7, 122.3. HRMS (ESI) (*m/z*) calcd for $C_8H_5BrN_2O$ $[M+H]^+$ 224.9658; found 224.9662.

2-(naphthalen-2-yl)-1,3,4-oxadiazole (3bn). Light yellow solid. Yield 63% (24.7 mg). m. p. 58-60 ^{o}C; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.53 (s, 1H), 8.51 (s, 1H), 8.11 (d, *J* = 8.6 Hz, 1H), 7.93 (d, *J* = 9.3 Hz, 2H), 7.86 (d, *J* = 7.5 Hz, 1H), 7.60 – 7.51 (m, 2H). ^{13}C NMR (100 MHz, $CDCl_3$): δ

= 164.8, 152.6, 134.6, 132.6, 129.0, 128.8, 128.0, 127.8, 127.5, 127.0, 123.0, 120.6. HRMS (ESI) (*m/z*) calcd for $C_{12}H_8N_2O$ $[M+H]^+$ 197.0710; found 197.0714.

2-(furan-2-yl)-1,3,4-oxadiazole (3bo). Yellow oil. Yield 40% (10.8 mg). ^1H NMR (400 MHz, $CDCl_3$): δ = 8.45 (s, 1H), 7.68 (d, *J* = 1.0 Hz, 1H), 7.22 (d, *J* = 3.5 Hz, 1H), 6.63 (dd, *J* = 3.5, 1.8 Hz, 1H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 157.5, 151.8, 146.0, 139.0, 114.6, 112.2. HRMS (ESI) (*m/z*) calcd for $C_6H_4N_2O_2$ $[M+H]^+$ 137.0346; found 137.0349.

2-(thiophen-2-yl)-1,3,4-oxadiazole (3bp). Yellow solid. Yield 67% (20.4 mg). m. p. 150-152 ^oC (lit.[12] m. p. 151-154 ^oC); ^1H NMR (400 MHz, $CDCl_3$): δ = 8.42 (s, 1H), 7.80 (dd, J = 3.7, 1.0 Hz, 1H), 7.58 (dd, J = 5.0, 1.0 Hz, 1H), 7.19 (dd, J = 5.0, 3.8 Hz, 1H). ^{13}C NMR (100 MHz, CDCl3): δ = 160.9, 151.9, 130.5, 130.2, 128.2, 124.6. HRMS (ESI) (*m/z*) calcd for $C_6H_4N_2OS$ $[M+H]^+$ 153.0117; found 153.0119.

Phenyl(5-phenyl-1,3,4-oxadiazol-2-yl)methanone (4ba). Light yellow solid. Yield 55% (27.9 mg). m. p. 95-97 ^oC; ^1H NMR (400 MHz, $CDCl_3$): δ = 8.58 (d, *J* = 7.2 Hz, 2H), 8.24 (d, *J* = 6.9 Hz, 2H), 7.74 – 7.69 (m, 1H), 7.65 – 7.54 (m, 5H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 177.6, 166.0, 160.9, 134.9, 134.3, 132.8, 131.0, 129.3, 128.8, 127.8, 122.8. HRMS (ESI) (*m/z*) calcd for $C_{15}H_{10}N_2O_2$ $[M+H]^+$ 251.0815; found 251.0819.

Phenyl(5-(pyridine-4-yl)-1,3,4-oxadiazol-2-yl)methanone (4bb). Light yellow solid. Yield 48% (24.5 mg). m. p. 148-150 ^{o}C; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.90 (d, *J* = 6.0 Hz, 2H), 8.57 (d, *J* = 7.3 Hz, 2H), 8.09 (d, *J* = 6.1 Hz, 2H), 7.74 (t, *J* = 7.4 Hz, 1H), 7.60 (t, *J* = 7.8 Hz, 2H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 177.3, 164.1, 161.2, 151.0, 135.2, 134.0, 131.0, 130.1, 128.9, 121.0. HRMS (ESI) (*m/z*) calcd for $C_{14}H_9N_3O_2$ $[M+H]^+$ 252.0732; found 252.0736.

(5-methyl-1,3,4-oxadiazol-2-yl)(phenyl)methanone (4bc). Brown solid. Yield 32% (11.9 mg); m. p. 107-109 ^{o}C; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.53 (dd, *J* = 8.4, 1.2 Hz, 2H), 7.72 – 7.68 (m, 1H), 7.56 (t, *J* = 7.8 Hz, 2H), 2.70 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 177.7, 165.8, 161.4, 134.9, 134.1, 130.9, 128.8, 11.2. HRMS (ESI) (*m/z*) calcd for $C_{10}H_8N_2O_2$ $[M+H]^+$ 189.0659; found 189.0660.

Supporting Information

Copies of ^{1}H and ^{13}C NMR spectra for all reaction products. The materials are available free of charge on the ACS Publications website.

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