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Iron-Catalyzed Oxidative 1,2-Carboacylation of Activated Alkenes with Alcohols: A Tandem Route to 3-(2-Oxoethyl)indolin-2-ones

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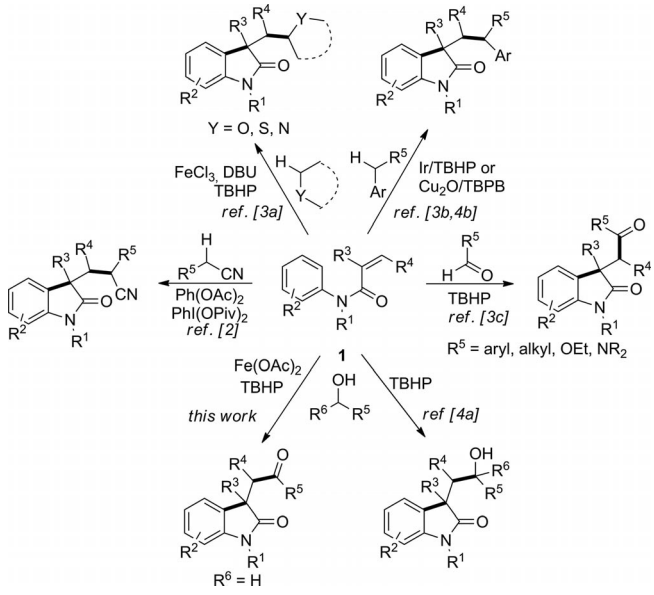
Oxindoles are important heterocyclic compounds that are found in a wide range of pharmaceutical agents and natural products. A new oxidative tandem route to the assembly of 3-(2-oxoethyl)indolin-2-ones from *N*-arylacrylamides and alcohols has been established by using inexpensive and environmentally benign iron catalysts and peroxides. In the presence of $Fe(OAc)_2$ and *tert*-butyl hydroperoxide, a variety of arylacrylamides underwent the oxidative 1,2-carboacylation reaction with alcohols to give the corresponding 3-(2-oxoethyl)indolin-2-ones in moderate to good yields.

Introduction

Oxidative difunctionalization of alkenes through C–H functionalization has emerged as a direct and highly atom-economical synthetic approach to a diverse range of products with important chemical, biological, and medicinal properties.[1] In this field, the oxidative C–H functionalization/carbocyclization of alkenes with two C–H bonds leading to the simultaneous formation of two carbon–carbon bonds is an elegant finding because their cyclic products are important skeletal units in natural products and bioactive molecules.[2–5] However, the oxidative C–H functionalization/carbocyclization of alkenes has not been much investigated (Scheme 1).[2–5] Liu and co-workers first reported oxidative difunctionalization of alkenes in *N*-arylacrylamides with the α-C(sp^3)–H bonds of alkyl nitriles using the $Pd(OAc)_2$/$PhI(OPiv)_2$ catalytic system, which provides a useful route to oxindoles.[2] Recently, our group,[3] the Li group,[3i] and the group of Guo and Duan[4] independently illustrated the oxidative difunctionalization of alkenes in *N*-arylacrylamides with various C–H bonds, including the C(sp^3)–H bond adjacent to a heteroatom[3a,4a] or an aryl group[3b,4b] and the carbonyl C(sp^2)–H bond[3c] using a transition metal and/or peroxide system.

Alcohols, which are fundamental bulk chemicals, are widely used as solvents and building blocks in organic synthesis and industry.[6] Direct functionalization of the C–H bond adjacent to a hydroxyl group still belongs to an important and challenging area of organic chemistry.[7,8]



Scheme 1. Oxidative difunctionalization of alkenes with two C–H bonds.

Tu[8a–d] and Liu[8e] have reported transition metal (Rh, Ru, Pd, Fe or Cu) catalyzed C–H functionalization/cross-coupling of alcohols with alkenes. Recently, Guo and Duan developed metal-free *tert*-butyl hydroperoxide (TBHP) mediated oxidative hydroxyalkylarylation of activated alkenes by direct C(sp^3)–H functionalization of alcohols, providing hydroxy-containing oxindoles in high yields.[4a] As part of our continuing interest in the oxidative C–H functionalization/carbocyclization of alkenes, we report here a tandem route to 3-(2-oxoethyl)indolin-2-ones by iron-catalyzed oxidative 1,2-carboacylation of activated alkenes with alcohols (Scheme 1). Carbonyl-containing oxindoles are fundamental structural motifs in pharmaceutical agents and natural products as well as versatile intermediates in organic synthesis.[9]

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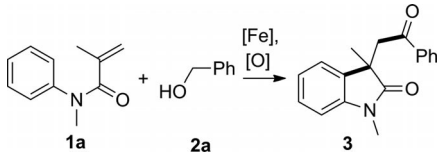
Results and Discussion

As shown in Table 1, we chose the reaction between *N*-methyl-*N*-phenylmethacrylamide (**1a**) and benzyl alcohol (**2a**) as a model with which to optimize the reaction conditions. Treatment of amide **1a** with alcohol **2a**, $Fe(OAc)_2$, and TBHP in EtOAc afforded the expected product **3** in 77% yield (entry 1). Therefore, a series of other Fe catalysts, including $FeCl_2$, $Fe(OTf)_2$, $FeCl_3$, $Fe(OTf)_3$, $Fe(acac)_3$, were examined (entries 2–6), however, these catalysts were less effective than $Fe(OAc)_2$. Interestingly, the reaction gave product **3** in the presence of TBHP without Fe catalysts, albeit in low yield (entry 7). Screening revealed that the amount of $Fe(OAc)_2$ affected the reaction, and 10 mol-% $Fe(OAc)_2$ was viable (entry 1 vs. entries 8–9). Subsequently, several oxidants, such as aqueous TBHP, di-*tert*-butyl peroxide (DTBP), 2,3-dichloro-5,6-dicyano-1,4-benzoquinone (DDQ), and O_2, were investigated (entries 10–13). Whereas both hydrous TBHP and DTBP displayed high activity for the reaction, DDQ and O_2 had no effect on the reaction. The effect of reaction temperature was also investigated, and it was found that conducting the reaction at 110 °C gave the best results (entries 1, 14 and 15). A number of other solvents, including *n*BuOAc, MeCN, dioxane and *N*,*N*-dimethylformamide (DMF), were also tested (entries 16–19). We found that *n*BuOAc was a suitable solvent for the reaction, with other solvents such as MeCN, dioxane, and DMF, suppressing the reaction.

Table 1. Screening optimal conditions.[a]



1a 2a 3

Entry	[M] (mol-%)	[O]	Solvent	*T* (°C)	Isolated yield [%]
1	$Fe(OAc)_2$ (10)	TBHP	EtOAc	110	77
2	$FeCl_2$ (10)	TBHP	EtOAc	110	65
3	$Fe(OTf)_2$ (10)	TBHP	EtOAc	110	51
4	$FeCl_3$ (10)	TBHP	EtOAc	110	15
5	$Fe(OTf)_3$ (10)	TBHP	EtOAc	110	59
6	$Fe(acac)_3$ (10)	TBHP	EtOAc	110	58
7	–	TBHP	EtOAc	110	28
8	$Fe(OAc)_2$ (5)	TBHP	EtOAc	110	41
9	$Fe(OAc)_2$ (15)	TBHP	EtOAc	110	75
10[b]	$Fe(OAc)_2$ (10)	TBHP	EtOAc	110	61
11	$Fe(OAc)_2$ (10)	DTBP	EtOAc	110	58
12	$Fe(OAc)_2$ (10)	DDQ	EtOAc	110	trace
13[c]	$Fe(OAc)_2$ (10)	–	EtOAc	110	trace
14	$Fe(OAc)_2$ (10)	TBHP	EtOAc	80	32
15	$Fe(OAc)_2$ (10)	TBHP	EtOAc	130	67
16	$Fe(OAc)_2$ (10)	TBHP	*n*BuOAc	110	76
17	$Fe(OAc)_2$ (10)	TBHP	MeCN	110	49
18	$Fe(OAc)_2$ (10)	TBHP	dioxane	110	trace
19	$Fe(OAc)_2$ (10)	TBHP	DMF	110	trace

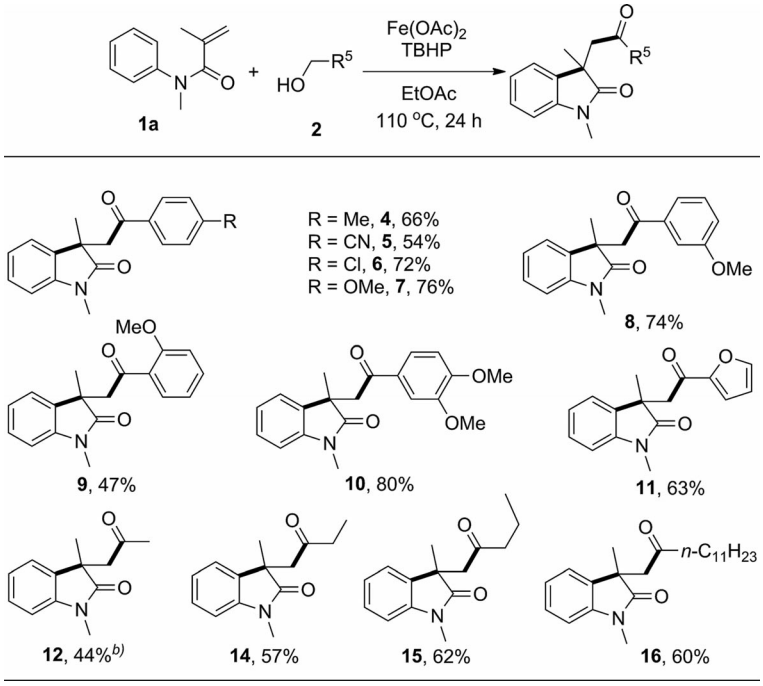
[a] Reaction conditions: **1a** (0.2 mmol), **2a** (3 equiv.), [Fe], [O] (2 equiv.), solvent (2 mL), 110 °C, 24 h. Unless specified otherwise, TBHP was used as a 5 M solution in decane. Some byproducts, including C–N decomposition products, were observed by GC–MS analysis, and the rest of alcohol **2a** was converted into the corresponding acid. [b] TBHP (70% in water). [c] O_2 (1 atm).

After determining the optimal conditions, we turned our attention to exploring the scope of this oxidative 1,2-carboacylation reaction (Table 2 and Table 3). As illustrated in Table 2, a variety of primary alcohols **2**, including benzyl alcohols, were compatible with the optimal conditions with respect to *N*-methyl-*N*-phenylmethacrylamide (**1a**), giving products **4**–**14**. Initially, a number of benzyl alcohols were investigated (products **4**–**11**). The results demonstrated that several substituents, including Me, CN, Cl and MeO groups, on the aromatic ring of the benzyl moiety were well-tolerated, and the reactive order of the substituents was *para*, *meta* > *ortho* (products **4**–**10**). For example, the reaction of *p*-tolylmethanol with amide **1a**, $Fe(OAc)_2$, and TBHP gave the desired product **4** in 66% yield. 4-CN- or 4-Cl-substituted benzyl alcohols were also viable for the reaction, giving the corresponding products **5** and **6** in good yields. Whereas (*p*-methoxyphenyl)methanol and (*m*-methoxyphenyl)methanol afforded products **7** and **8** in 76 and 74% yield, respectively, (*o*-methoxyphenyl)methanol gave a lower yield of the corresponding product **9** (47%). Notably, the optimized conditions were compatible with a heteroaryl methanol, furan-2-ylmethanol, resulting in the formation of **11** in 63% yield. In light of these results, other alkyl alcohols were examined and all were found to be suitable substrates, providing the desired products **12** and **14**–**16** in moderate yield. It is noteworthy that the reaction with ethanol offered another hydroxy-containing product, **13**, in 31% yield. Unfortunately, secondary alcohols were inert under these conditions.

We next set out to apply the iron-catalyzed oxidative tandem reaction to *N*-arylacrylamides **1** in the presence of (*p*-methoxyphenyl)methanol, $Fe(OAc)_2$, and TBHP (Table 3). *N*-Phenylmethacrylamides with a N–Bn group or a N–Ac group were found to be suitable substrates for the reaction (products **17** and **18**), but substrates with an N–H group did not react (product **19**). Gratifyingly, *N*-arylacrylamides **1**, bearing either electron-donating or electron-withdrawing substituents on the aromatic ring of the *N*-aryl moiety, were compatible with the optimized conditions, and the electron-donating groups displayed higher activity than electron-withdrawing groups (products **20**–**26**). Importantly, *ortho*-halo groups were perfectly tolerated, affording the corresponding products **23** and **24** in moderate yields. Using substrate amides with a *meta*-methyl group, gave a mixture of products **25** and **26**. It was noted that *N*-arylacrylamides with a phenyl group at the 2-position was also viable in the reaction with (*p*-methoxyphenyl)methanol, $Fe(OAc)_2$ and TBHP, providing the desired product **27** in 58% yield.

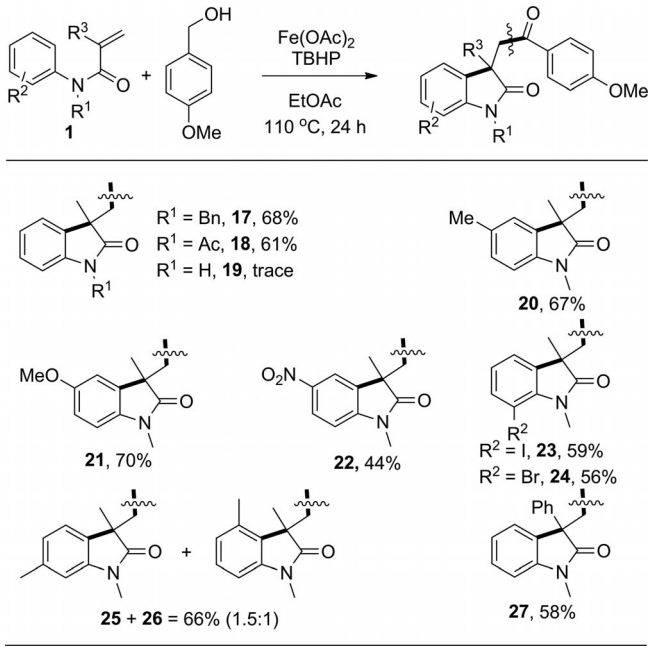
To understand the mechanism followed in the oxidative 1,2-carboacylation reaction, a number of control experiments were performed (Scheme 2).[10] These experiment showed that (*p*-methoxyphenyl)methanol could be readily converted into aldehyde **28** in the presence of $Fe(OAc)_2$ and TBHP [Equation (1)]. However, dodecan-1-ol display lower reactivity under the same conditions [Equation (2)]. Interestingly, substrate **13**, which was synthesized by a reported

Table 2. Screening scope of alcohols **2**.[a]



[a] Reaction conditions: **1a** (0.3 mmol), **2** (2 equiv.), $Fe(OAc)_2$ (10 mol-%), TBHP (3 equiv; 5 M in decane), EtOAc (2 mL), 110 °C, 24 h. [b] A mixture of **12** and byproduct **13** [3-(2-hydroxypropyl)-1,3-dimethylindolin-2-one] was isolated in 75% total yield with 1.4:1 ratio.

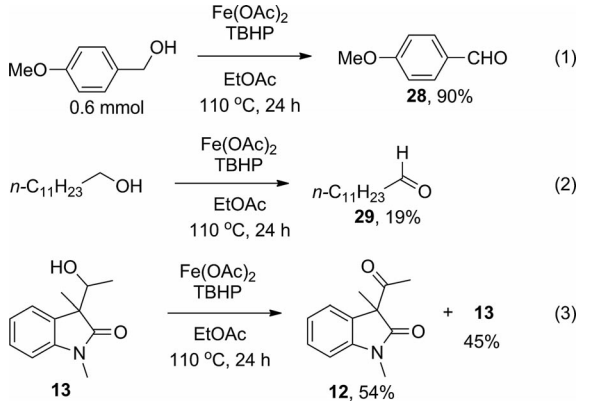
Table 3. Screening scope of *N*-arylacrylamides (**1**).[a]



[a] Reaction conditions: **1** (0.3 mmol), **2** (2 equiv.), $Fe(OAc)_2$ (10 mol-%), TBHP (3 equiv; 5 M in decane), EtOAc (2 mL), 110 °C, 24 h.

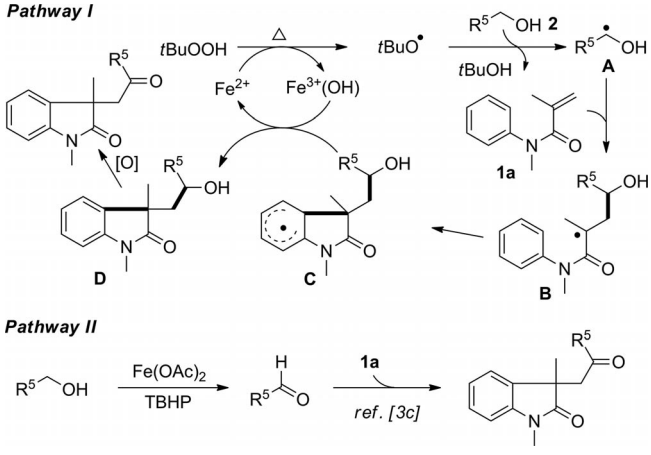
method,[4a] could be transformed into ketone **12** in 54% yield [Equation (3)]. These results suggest that the oxidative 1,2-carboacylation reaction may proceed through two different mechanisms.

Consequently, possible mechanisms as outlined in Scheme 3 were proposed on the basis of the present results.[5,6] Initially, TBHP is easily split by Fe^{2+} into a *tert*-butoxy radical and Fe^{3+}(OH) (Pathway I). The reaction of



Scheme 2. Control experiments.

a *tert*-butoxy radical with an alcohol affords radical intermediate A by abstracting the C(sp^3)–H bonds adjacent to an oxygen atom. Addition of radical intermediate **A** to the carbon–carbon double bond of substrate **1a** gave radical intermediate **B**, followed by intramolecular carbocyclization of radical intermediate **B** to furnish radical intermediate **C**. Hydrogen abstraction of radical intermediate **C** by Fe^{3+}(OH) takes place to produce 3-(2-hydroxyethyl)indolin-2-one **D**. The oxidation of **D** by $Fe(OAc)_2$ and TBHP gives the expected 3-(2-hydroxyethyl)indolin-2-one[10] and regenerates the active Fe^{II} species.



Scheme 3. Possible mechanisms.

Another pathway includes initial oxidation of the alcohol to give the aldehyde, followed by a acylation–carbocyclization tandem reaction to give the expected 3-(2-hydroxyethyl)indolin-2-one (Pathway II). It is noteworthy that benzyl alcohols may predominantly perform the reaction through Pathway II.

Conclusions

We have established an iron-catalyzed oxidative carboacylation tandem route to 3-(2-oxoethyl)indolin-2-ones. This tandem method is realized by 1,2-difunctionalization of the C–C double bond in *N*-arylacrylamides with alcohols by using inexpensive and environmentally benign iron catalysts; moreover, it provides a shortcut to carbonyl-containing oxindoles from alcohols.

Experimental Section

General Methods: All the materials and solvents were purchased from commercial suppliers and used without additional purification. IR measurements were performed with a FTIR Shimadzu DR-8000 spectrometer fitted with a Pike Technologies MIRacle Single Reflection ATR adapter. ^{1}H and ^{13}C NMR spectra were recorded with a Bruker DRX-500 spectrometer (^{1}H at 500 MHz and ^{13}C at 125 MHz) or a Bruker DRX-400 spectrometer (^{1}H at 400 MHz and ^{13}C at 100 MHz). NMR spectroscopic data were obtained in $CDCl_3$ unless otherwise noted. High-resolution mass spectra were recorded with a Bruker microTOF-QII (ESI) spectrometer. Preparative thin-layer chromatography was performed on silica gel plates with PF254 indicator. Flash column chromatography was performed with silica gel 60N unless otherwise noted.

Typical Experimental Procedure for the Fe-Catalyzed Synthesis of 3-(2-Oxoethyl)indolin-2-ones: To a Schlenk tube were added *N*-arylacrylamide **1** (0.3 mmol), alcohol **2** (2 equiv.), $Fe(OAc)_2$ (10 mol-%), TBHP (5 M in decane, 3 equiv), and EtOAc (2 mL). The tube was charged with argon and the mixture was stirred at 110 °C for 24 h until complete consumption of starting material was observed (reaction monitored by TLC and/or GC–MS analysis). When the reaction was complete, the reaction mixture was washed with brine, the aqueous phase was re-extracted with ethyl acetate, and the combined organic extracts were dried with Na_2SO_4, concentrated in vacuo, and the resulting residue was purified by silica gel column chromatography (hexane/ethyl acetate) to afford the desired product.

1,3-Dimethyl-3-(2-oxo-2-phenylethyl)indolin-2-one (3):[3c,5f] Colorless liquid. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.83 (d, J = 7.2 Hz, 2 H), 7.51 (t, J = 7.2 Hz, 1 H), 7.39 (t, J = 8.0 Hz, 2 H), 7.26 (t, J = 8.0 Hz, 1 H), 7.14 (d, J = 7.2 Hz, 1 H), 6.98 (t, J = 7.6 Hz, 1 H), 6.90 (d, J = 8.0 Hz, 1 H), 3.72 (d, J = 18.0 Hz, 1 H), 3.65 (d, J = 18.0 Hz, 1 H), 3.31 (s, 3 H), 1.44 (s, 3 H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 195.9, 180.4, 143.7, 136.3, 133.6, 133.0, 128.3, 127.8, 127.7, 122.0, 121.6, 108.0, 45.9, 45.1, 26.3, 24.8 ppm. MS (EI, 70 eV): *m*/*z* (%) = 279 (87) [M^+], 234 (22), 220 (8), 160 (100).

1,3-Dimethyl-3-[2-oxo-2-(*p*-tolyl)ethyl]indolin-2-one (4):[3c] Colorless oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.74 (d, J = 8.2 Hz, 2 H), 7.26 (t, J = 4.0 Hz, 1 H), 7.19 (d, J = 8.0 Hz, 1 H), 7.13 (d, J = 6.8 Hz, 1 H), 6.95 (t, J = 8.0 Hz, 1 H), 6.95 (t, J = 7.6 Hz, 1 H), 3.71 (d, J = 18.0 Hz, 1 H), 3.63 (d, J = 18.0 Hz, 1 H), 3.32 (s, 3 H), 2.37 (s, 3 H), 1.43 (s, 3 H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 195.6, 180.6, 143.9, 143.8, 133.9, 133.8, 129.1, 128.0, 127.7, 122.0, 121.7, 108.0, 45.8, 45.2, 26.4, 24.8, 21.5 ppm. MS (EI, 70 eV): *m*/*z* (%) = 293 (78) [M^+], 248 (23), 174 (30), 160 (79), 119 (100).

4-[2-(1,3-Dimethyl-2-oxoindolin-3-yl)acetyl]benzonitrile (5):[3c] Colorless oil. ^{1}H NMR (500 MHz, $CDCl_3$): δ = 7.90 (d, J = 8.0 Hz, 2 H), 7.70 (d, J = 8.5 Hz, 2 H), 7.27 (t, J = 7.5 Hz, 1 H), 7.13 (d, J = 7.0 Hz, 1 H), 6.99 (t, J = 7.5 Hz, 1 H), 6.91 (d, J = 8.0 Hz, 1 H), 3.67 (s, 2 H), 3.30 (s, 3 H), 1.45 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 194.9, 180.1, 143.7, 139.1, 133.1, 132.3, 128.3, 128.0, 122.3, 121.7, 117.7, 116.4, 108.2, 46.1, 45.2, 26.4, 24.8 ppm. MS (EI, 70 eV): *m*/*z* (%) = 304 (37) [M^+], 259 (13), 245 (7), 160 (100).

3-[2-(4-Chlorophenyl)-2-oxoethyl]-1,3-dimethylindolin-2-one (6): Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.76 (d, J = 8.4 Hz, 2 H), 7.36 (d, J = 8.4 Hz, 2 H), 7.26 (d, J = 7.6 Hz, 1 H), 7.13 (t, J = 7.2 Hz, 1 H), 6.98 (d, J = 7.6 Hz, 1 H), 6.90 (d, J = 8.0 Hz, 1 H), 3.67 (d, J = 18.0 Hz, 1 H), 3.62 (d, J = 18.0 Hz, 1 H), 3.30 (s, 3 H), 1.43 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 194.9, 180.4, 143.7, 139.6, 134.5, 133.5, 129.3, 128.7, 127.9, 122.2, 121.6, 108.1, 45.9, 45.2, 26.4, 24.8 ppm. IR (KBr): $\tilde{\nu}$ = 1702, 1686 cm^{-1}. MS (EI, 70 eV): *m*/*z* (%) = 315 (14) [M^+ + 2], 313 (37) [M^+], 268 (20), 168 (100). HRMS (ESI): *m*/*z* calcd. for $C_{18}H_{17}ClNO_2$ [M + H]$^+$ 314.0948; found 314.0957.

3-[2-(4-Methoxyphenyl)-2-oxoethyl]-1,3-dimethylindolin-2-one (7):[3c] Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.81 (d, J = 8.8 Hz, 2 H), 7.24 (t, J = 8.0 Hz, 1 H), 7.13 (d, J = 7.2 Hz, 1 H), 6.96 (t, J = 7.6 Hz, 1 H), 6.89 (d, J = 8.0 Hz, 1 H), 6.85 (d, J = 8.8 Hz, 2 H), 3.81 (s, 3 H), 3.68 (d, J = 18.0 Hz, 1 H), 3.59 (d, J = 17.6 Hz, 1 H), 3.30 (s, 3 H), 1.42 (s, 3 H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 194.5, 180.7, 163.5, 143.8, 133.9, 130.2, 129.5, 127.7, 122.1, 121.7, 113.6, 108.1, 55.4, 45.6, 45.3, 26.4, 24.9 ppm. MS (EI, 70 eV): *m*/*z* (%) = 309 (28) [M^+], 264 (10), 174 (13), 160 (28), 135 (100).

3-[2-(3-Methoxyphenyl)-2-oxoethyl]-1,3-dimethylindolin-2-one (8):[3c] Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.45 (d, J = 7.6 Hz, 1 H),7.32 (t, J = 4.0 Hz, 2 H), 7.28 (t, J = 6.4 Hz, 1 H), 7.13 (d, J = 8.0 Hz, 1 H), 7.06–7.05 (m, 1 H), 6.97 (t, J = 7.2 Hz, 1 H), 6.90 (d, J = 7.6 Hz, 1 H), 3.77 (s, 3 H), 3.71 (d, J = 18.0 Hz, 1 H), 3.64 (d, J = 18.0 Hz, 1 H), 3.31 (s, 3 H), 1.43 (s, 3 H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 195.9, 180.5, 159.6, 143.7, 137.6, 133.6, 129.4, 127.7, 122.1, 121.6, 120.5, 119.9, 111.8, 108.0, 55.3, 46.0, 45.2, 26.3, 24.9 ppm. MS (EI, 70 eV): *m*/*z* (%) = 309 (96) [M^+], 264 (28), 250 (11), 160 (100).

3-[2-(2-Methoxyphenyl)-2-oxoethyl]-1,3-dimethylindolin-2-one (9):[3c] Colorless oil. ^{1}H NMR (500 MHz, $CDCl_3$): δ = 7.42–7.37 (m, 2 H), 7.24 (t, J = 7.5 Hz, 1 H), 7.13 (d, J = 5 Hz, 1 H), 6.97 (t, J = 7.5 Hz,1 H), 6.91 (d, J = 10 Hz, 1 H), 6.88–6.85 (m, 2 H), 3.91 (s, 3 H), 3.76 (d, J = 15 Hz, 1 H), 3.67 (d, J = 20 Hz, 1 H), 3.28 (s, 3 H), 1.39 (s, 3 H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 198.0, 180.8, 158.6, 143.8, 134.0, 133.6, 130.3, 127.6, 122.6, 122.0, 121.8, 120.5, 111.3, 108.0, 55.5, 51.2, 45.7, 26.4, 25.0 ppm. MS (EI, 70 eV): *m*/*z* (%) = 309 (49) [M^+], 264 (12), 160 (33), 135 (100).

3-[2-(3,4-Dimethoxyphenyl)-2-oxoethyl]-1,3-dimethylindolin-2-one (10): Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.54 (d, J = 8.4 Hz, 1 H),7.33 (d, J = 2.0 Hz, 1 H), 7.25 (t, J = 7.2 Hz, 1 H), 7.14 (d, J = 7.2 Hz, 1 H), 6.98 (t, J = 7.6 Hz, 1 H), 6.91 (t, J = 8.0 Hz, 1 H), 6.84 (d, J = 8.4 Hz, 1 H), 3.92 (s, 3 H), 3.83 (s, 3 H), 3.69 (d, J = 17.6 Hz, 1 H), 3.63 (d, J = 17.6 Hz, 1 H), 3.31 (s, 3 H), 1.43 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 194.7, 180.7, 153.3, 148.9, 143.7, 133.9, 129.6, 127.7, 122.6, 122.1, 121.7, 110.0, 109.8, 108.1, 56.0, 55.8, 45.5, 45.3, 26.4, 25.0 ppm. IR (KBr): $\tilde{\nu}$ = 1692, 1684 cm^{-1}. MS (EI, 70 eV): *m*/*z* (%) = 339 (43) [M^+], 294 (14), 165 (100). HRMS (ESI): *m*/*z* calcd. for $C_{20}H_{22}NO_4$ [M + H]$^+$ 340.1549; found 340.1556.

3-[2-(Furan-2-yl)-2-oxoethyl]-1,3-dimethylindolin-2-one (11):[3c] Colorless oil. ^{1}H NMR (500 MHz, $CDCl_3$): δ = 7.51 (m, 1 H), 7.24 (t, J = 10 Hz, 1 H), 7.16 (d, J = 10 Hz, 1 H), 7.07 (d, J = 5 Hz, 1 H), 6.97 (t, J = 7.5 Hz, 1 H), 6.87 (d, J = 5 Hz, 1 H), 6.47 (d, J = 5 Hz, 1 H), 3.58 (d, J = 10 Hz, 1 H), 3.44 (d, J = 7.5 Hz, 1 H), 3.29 (s, 3 H), 1.43 (s, 3 H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 185.4, 180.3, 152.4, 146.2, 143.8, 133.4, 127.9, 122.2, 122.1, 116.9, 112.3, 108.1, 45.4, 45.3, 26.4, 24.5 ppm. MS (EI, 70 eV): *m*/*z* (%) = 269 (36) [M^+], 224 (15), 210 (12), 160 (100).

1,3-Dimethyl-3-(2-oxopropyl)indolin-2-one (12) and 3-(2-Hydroxypropyl)-1,3-dimethylindolin-2-one (13):[5f] Ratio **12/13** = 1.4:1; colorless oil. ^{1}H NMR (500 MHz, $CDCl_3$): δ = 7.31–7.24 (m, 2 H), 7.19 (d, J = 7.2 Hz, 0.85 H), 7.14 (d, J = 7.2 Hz, 1 H), 7.09 (t, J = 7.6 Hz, 0.83 H), 7.01 (t, J = 7.6 Hz, 1 H), 6.87 (d, J = 7.6 Hz, 1.78 H), 4.15–4.08 (m, 0.85 H), 3.82 (br. s, 0.73 H), 3.26 (s, 3 H), 3.23 (s, 2.5 H), 3.13 (d, J = 18.0 Hz, 1 H), 3.08 (d, J = 18.4 Hz, 1 H), 1.98 (s, 3 H), 1.97–1.93 (m, 1 H), 1.79–1.73 (m, 0.94 H), 1.35 (s, 2.5 H), 1.33 (s, 3 H), 1.15 (d, J = 6.0 Hz, 2.5 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 204.6, 181.9, 180.3, 143.6, 142.3, 134.9, 133.3, 128.0, 127.8, 123.0, 122.4, 122.2, 121.7, 108.4, 108.1, 64.6, 50.4, 47.3, 46.0, 45.1, 29.9, 26.3, 24.3, 23.9, 22.8 ppm. MS (EI, 70 eV) for **12**: *m*/*z* (%) = 217 (82) [M^+], 174 (50), 160 (100); MS (EI, 70 eV) for **13**: *m*/*z* (%) = (30) [M^+] [%] 219, 174 (52), 161 (100).

1,3-Dimethyl-3-(2-oxobutyl)indolin-2-one (14): Colorless oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.25 (d, J = 7.6 Hz, 1 H), 7.13 (d, J = 7.6 Hz, 1 H), 7.00 (t, J = 7.6 Hz, 1 H), 6.87 (d, J = 7.6 Hz, 1 H), 3.27 (s, 3 H), 3.08 (s, 2 H), 2.39–2.19 (m, 2 H), 1.33 (s, 3 H), 0.89 (t, J = 7.2 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 207.4, 180.4, 143.7, 133.5, 127.9, 122.2, 121.7, 108.2, 49.3, 45.1, 35.9, 26.4, 24.5, 7.3 ppm. IR (KBr): $\tilde{\nu}$ = 1703, 1692 cm^{-1}. MS (EI, 70 eV): *m*/*z* (%) = 231 (59) [M^+], 174 (77), 160 (100). HRMS (ESI): *m*/*z* calcd. for $C_{14}H_{18}NO_2$ [M + H]$^+$ 232.1338; found 232.1342.

1,3-Dimethyl-3-(2-oxopentyl)indolin-2-one (15):[3c] Colorless oil. ^{1}H NMR (500 MHz, $CDCl_3$): δ = 7.25–7.23 (m, 1 H), 7.14 (d, J = 5 Hz, 1 H), 7.02 (t, J = 7.5 Hz, 1 H), 6.88 (d, J = 2.5 Hz, 1 H), 3.28 (s, 3 H), 3.08 (s, 2 H), 2.29–2.12 (m, 2 H), 1.46–1.41 (m, 2 H), 1.33 (s, 3 H), 0.77 (t, J = 7.5 Hz, 3 H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 206.9, 180.4, 143.7, 133.5, 127.8, 122.1, 121.7, 108.1, 49.7, 45.2, 44.7, 29.7, 24.4, 16.9, 13.5 ppm. MS (EI, 70 eV): *m*/*z* (%) = 246 (13) [M^+ + 1], 245 (70) [M^+], 200 (5), 174 (100).

1,3-Dimethyl-3-(2-oxotridecyl)indolin-2-one (16): Colorless oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.24 (t, J = 7.6 Hz, 1 H), 7.12 (d, J = 7.2 Hz, 1 H), 6.99 (d, J = 7.6 Hz, 1 H), 6.85 (d, J = 7.6 Hz, 1 H), 3.26 (s, 3 H), 3.07 (s, 2 H), 2.31–2.16 (m, 2 H), 1.41–1.39 (m, 2 H), 1.33 (s, 3 H), 1.26–1.24 (m, 16 H), 0.87 (t, J = 7.2 Hz, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 207.0, 180.3, 143.6, 133.5, 127.8, 122.1, 121.6, 108.9, 49.6, 45.1, 42.8, 31.8, 29.5 (2 C), 29.3, 29.2, 28.9, 26.3, 24.4, 23.4, 22.6, 14.0 ppm. IR (KBr): $\tilde{\nu}$ = 1701, 1698 cm^{-1}. MS (EI, 70 eV): *m*/*z* (%) = 357 (26) [M^+], 174 (71), 160 (100). HRMS (ESI): *m*/*z* calcd. for $C_{23}H_{36}NO_2$ [M + H]$^+$ 358.2746; found 358.2751.

1-Benzyl-3-[2-(4-methoxyphenyl)-2-oxoethyl]-3-methylindolin-2-one (17):[3c] Yellow solid; m.p. 101.2–102.3 °C (uncorrected). ^{1}H NMR (500 MHz, $CDCl_3$): δ = 7.85 (d, J = 9.0 Hz, 2 H), 7.41 (d, J = 7.5 Hz, 2 H), 7.33 (t, J = 7.5 Hz, 2 H), 7.25 (d, J = 9.0 Hz, 1 H), 7.13 (d, J = 7.0 Hz, 1 H), 7.08 (d, J = 9.0 Hz, 1 H), 6.92 (t, J = 7.5 Hz, 1 H), 6.86 (d, J = 9.0 Hz, 2 H), 6.72 (d, J = 8.0 Hz, 1 H), 5.08 (d, J = 16.0 Hz, 1 H), 4.95 (d, J = 16.0 Hz, 1 H), 3.81 (s, 3 H), 3.71–3.69 (m, 2 H), 1.48 (s, 3 H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 194.5, 180.8, 163.6, 142.9, 136.3, 134.0, 130.4, 128.7, 127.7, 127.4, 127.3, 122.2, 121.8, 113.7, 109.3, 55.5, 45.6, 44.0, 25.6 ppm. MS (EI, 70 eV): *m*/*z* (%) = 385 (42) [M^+], 250 (9), 235 (50), 135 (100).

1-Acetyl-3-[2-(4-methoxyphenyl)-2-oxoethyl]-3-methylindolin-2-one (18): Colorless oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.32 (d, J = 8.0 Hz, 2 H), 7.82 (d, J = 8.8 Hz, 2 H), 7.31–7.27 (m, 1 H), 7.13–7.09 (m, 2 H), 6.88 (d, J = 8.8 Hz, 2 H), 3.85 (s, 3 H), 3.81 (d, J = 18.0 Hz, 1 H), 3.68 (d, J = 18.0 Hz, 1 H), 2.74 (s, 3 H),1.48 (s, 3 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 194.2, 181.5, 171.3, 163.8, 140.0, 132.9, 130.3, 128.8, 128.1, 124.8, 120.9, 116.7, 113.7, 55.5, 47.0, 45.7, 26.7, 26.1 ppm. IR (KBr): $\tilde{\nu}$ = 1723, 1691,

1685 cm^{-1}. MS (EI, 70 eV): *m/z* (%) = 337 (32) [M^+], 264 (4), 135 (100). HRMS (ESI): *m/z* calcd. for $C_{20}H_{20}NO_4$ [M + H]$^+$ 338.1392; found 338.1398.

3-[2-(4-Methoxyphenyl)-2-oxoethyl]-1,3,5-trimethylindolin-2-one (20):[3c] Colorless oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.83 (d, *J* = 78.8 Hz, 2 H), 7.04 (d, *J* = 8.0 Hz, 1 H), 6.94 (s, 1 H), 6.87 (d, *J* = 8.8 Hz, 2 H), 6.78 (d, *J* = 8.0 Hz, 1 H), 3.84 (s, 3 H), 3.65 (d, *J* = 17.6 Hz, 1 H), 3.59 (d, *J* = 18.0 Hz, 1 H), 3.29 (s, 3 H), 2.27 (s, 3 H), 1.41 (s, 3 H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 194.6, 180.6, 163.4, 141.4, 133.9, 131.5, 130.2, 129.5, 127.9, 122.7, 113.5, 107.8, 55.4, 45.6, 45.3, 26.4, 25.0, 21.1 ppm. MS (EI, 70 eV): *m/z* (%) = 323 (87) [M^+], 278 (30), 174 (88), 135 (100).

5-Methoxy-3-[2-(4-methoxyphenyl)-2-oxoethyl]-1,3-dimethylindolin-2-one (21):[3c] Colorless oil. ^{1}H NMR (500 MHz, $CDCl_3$): δ = 7.82 (d, *J* = 9.0 Hz, 2 H), 6.85 (d, *J* = 9.0 Hz, 2 H), 6.80–6.74 (m, 3 H), 3.82 (s, 3 H), 3.72 (s, 3 H), 3.64 (d, *J* = 18.0 Hz, 1 H), 3.59 (d, *J* = 17.5 Hz, 1 H), 3.28 (s, 3 H), 1.42 (s, 3 H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 194.5, 180.3, 163.4, 155.6, 137.3, 135.3, 130.2, 129.4, 113.5, 111.3, 109.8, 108.2, 55.6, 55.3, 45.7, 45.3, 26.4, 24.9 ppm. MS (EI, 70 eV): *m/z* (%) = 339 (94) [M^+], 294 (40), 280 (12), 254 (11), 190 (100), 135 (91).

3-[2-(4-Methoxyphenyl)-2-oxoethyl]-1,3-dimethyl-5-nitroindolin-2-one (22):[3c] Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.26 (d, *J* = 8.8 Hz, 1 H), 7.99 (d, *J* = 2.4 Hz, 1 H), 7.81 (d, *J* = 9.2 Hz, 2 H), 6.98 (d, *J* = 8.4 Hz, 1 H), 6.88 (d, *J* = 8.8 Hz, 2 H), 3.85 (s, 3 H), 3.78 (d, *J* = 18.0 Hz, 1 H), 3.73 (d, *J* = 18.0 Hz, 1 H), 3.39 (s, 3 H), 1.46 (s, 3 H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 194.2, 180.8, 163.9, 149.8, 143.1, 135.0, 130.3, 128.8, 125.3, 117.3, 113.8, 107.6, 55.5, 46.0, 45.1, 26.9, 24.7 ppm. MS (EI, 70 eV): *m/z* (%) = 354 (13) [M^+], 306 (10), 219 (30), 205 (39), 135 (100).

7-Iodo-3-[2-(4-methoxyphenyl)-2-oxoethyl]-1,3-dimethylindolin-2-one (23):[3c] Red oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.81 (d, *J* = 8.8 Hz, 2 H), 7.63 (d, *J* = 8.8 Hz, 1 H), 7.04 (d, *J* = 7.2 Hz, 1 H), 6.87 (d, *J* = 8.8 Hz, 2 H), 6.66 (t, *J* = 7.6 Hz, 1 H), 3.84 (s, 3 H), 3.69 (s, 3 H), 3.64 (s, 2 H), 1.39 (s, 3 H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 194.3, 181.5, 163.6, 144.2, 140.2, 137.1, 130.2, 129.2, 123.8, 121.2, 113.6, 71.9, 55.4, 45.9, 44.8, 30.3, 25.3 ppm. MS (EI, 70 eV): *m/z* (%) = 435 (43) [M^+], 300 (6), 286 (13), 135 (100).

7-Bromo-3-[2-(4-methoxyphenyl)-2-oxoethyl]-1,3-dimethylindolin-2-one (24):[3c] Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.82 (d, *J* = 8.8 Hz, 2 H), 7.34 (d, *J* = 8.4 Hz, 1 H), 7.01 (d, *J* = 7.2 Hz, 1 H), 6.87 (d, *J* = 8.8 Hz, 2 H), 6.80 (t, *J* = 7.6 Hz, 1 H), 3.84 (s, 3 H), 3.69 (s, 3 H), 3.65 (s, 2 H), 1.40 (s, 3 H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 194.3, 181.2, 163.6, 141.2, 137.2, 133.3, 130.2, 129.2, 123.2, 120.5, 113.6, 102.6, 55.4, 46.0, 45.0, 30.0, 25.3 ppm. MS (EI, 70 eV): *m/z* (%) = 389 (12) [M^+ + 2], 387 (12) [M^+], 240 (6), 238 (6), 135 (100).

3-[2-(4-Methoxyphenyl)-2-oxoethyl]-1,3,6-trimethylindolin-2-one (25) and 3-[2-(4-Methoxyphenyl)-2-oxoethyl]-1,3,4-trimethylindolin-2-one (26):[3c] Ratio **25**/**26** = 1.5:1; Colorless oil. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.84–7.80 (m, 3.2 H), 7.15 (t, *J* = 8.0 Hz, 1 H), 7.01 (d, *J* = 7.6 Hz, 0.7 H), 6.86 (d, *J* = 8.80 Hz, 3.3 H), 6.79–6.72 (m, 2.5 H), 3.92 (d, *J* = 17.6 Hz, 1 H), 3.81 (s, 5 H), 3.68–3.55 (m, 2.4 H), 3.29 (s, 2 H), 3.28 (s, 3 H), 2.36 (s, 2 H), 2.29 (s, 3 H), 1.48 (s, 3 H), 1.41 (s, 2 H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 194.7, 181.0, 180.6, 163.4, 144.0, 143.8, 137.7, 132.7, 130.9, 130.5, 130.2, 130.1, 129.5, 129.2, 128.6, 127.5, 124.7 (2 C), 122.5, 121.4, 113.6, 113.5, 109.1, 105.9, 55.3, 46.1, 45.6, 45.1, 44.5, 29.6, 26.4, 26.3, 24.9, 22.8, 21.7, 18.1 ppm. MS (EI, 70 eV) for **25**: *m/z* (%) = 323 (79) [M^+], 308 (6), 174 (77), 173 (83), 135 (100); MS (EI, 70 eV) for **26**: *m/z* [%] = 323 (60) [M^+], 264 (4), 174 (68), 135 (100).

3-[2-(4-Methoxyphenyl)-2-oxoethyl]-1-methyl-3-phenylindolin-2-one (27):[3c] Yellow oil. ^{1}H NMR (500 MHz, $CDCl_3$): δ = 7.83 (d, *J* = 8.5 Hz, 2 H), 7.44 (d, *J* = 7.5 Hz, 2 H), 7.33–7.29 (m, 3 H), 7.25 (t, *J* = 7.5 Hz, 2 H), 7.03 (t, *J* = 7.5 Hz, 1 H), 6.93 (d, *J* = 7.5 Hz, 1 H), 6.86 (d, *J* = 8.5 Hz, 2 H), 4.12 (d, *J* = 18.0 Hz, 1 H), 4.04 (d, *J* = 18.0 Hz, 1 H), 3.83 (s, 3 H), 3.29 (s, 3 H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 194.2, 178.7, 163.5, 144.8, 139.7, 131.7, 130.3, 129.5, 128.6, 128.3, 127.5, 126.7, 124.1, 122.1, 113.6, 108.4, 55.4, 53.2, 46.6, 26.7 ppm. MS (EI, 70 eV): *m/z* (%) = 371 (56) [M^+], 326 (10), 278 (15), 236 (24), 222 (75), 135 (100).

Supporting Information (see footnote on the first page of this article): ^{1}H and ^{13}C NMR spectra for 3-(2-oxoethyl)indolin-2-one products.

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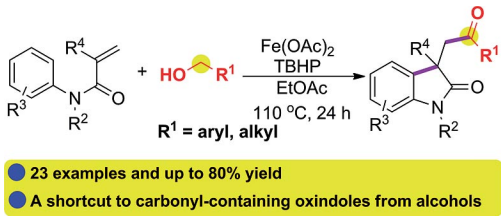
Iron-Catalyzed Cyclization



A route to 3-(2-oxoethyl)indolin-2-ones is presented that involves iron-catalyzed oxidative tandem reaction of *N*-arylacrylamides with alcohols. This method represents a new 1,2-difunctionalization of activated alkenes with alcohols, and provides a shortcut to carbonyl-containing oxindoles from alcohols.

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Iron-Catalyzed Oxidative 1,2-Carboacylation of Activated Alkenes with Alcohols: A Tandem Route to 3-(2-Oxoethyl)indolin-2-ones

Keywords: Domino reactions / Cyclization / Iron / Oxidation / Alkenes / Alcohols

