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Photoredox-Induced Functionalization of Alkenes for the Synthesis of Substituted Imidazolines and Oxazolidines

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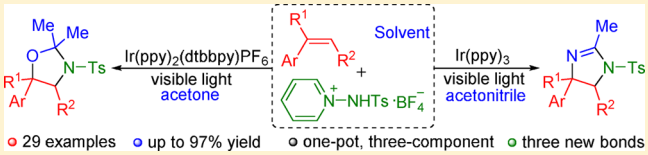
Supporting Information

ABSTRACT: A one-pot, three-component cascade reaction combining photoredox catalyzed radical addition and formal [3 + 2] annulation was developed. With this approach, highly concise syntheses of imidazoline and oxazolidine derivatives have been achieved. The advantages of this transformation are good to excellent yields, mild reaction conditions, operational simplicity, and easy accessibility of raw materials.



INTRODUCTION

Nitrogenous heterocyclic compounds are widely found in nature.[1] Some examples include imidazolines and oxazolidines, which have been commonly used as biologically active molecules,[2] organic synthetic intermediates,[3] and organocatalysts.[4] Especially, imidazolines have long been used as auxiliary groups[5] and metal complexation ligands[6] for asymmetric synthesis. Therefore, as shown in Scheme 1, it is not surprising that the syntheses of imidazoline and oxazolidine derivatives have been extensively investigated. In general, 1,2-diamine and 1,2-amino alcohol, which are not easily available, can be smoothly transformed into the corresponding imidazoline and oxazolidine under harsh conditions,[7] respectively. Besides, Lewis acid mediated [3 + 2] cycloaddition of *N*-tosylaziridines with nitriles or carbonyls, as an alternative approach, was applied to the syntheses of imidazolines or oxazolidines.[8] However, *N*-tosylaziridines had to be synthesized from 1,2-amino alcohol.[9] Recently, Yeung et al. developed the cationic Br or Cl initiated imidazoline synthesis from olefin by a two-step transformation.[10] Therefore, the development of novel methods to transform the unactivated alkenes directly into imidazolines and oxazolidines under mild reaction conditions will be highly desired.

More recently, owing to its ability to implement the incredible conversion reactions under benign conditions, visible-light photocatalysis has attracted a great deal of attention from the chemistry community.[11] Recent studies have shown that the *N*-centered radical, generated from the unactivated substrates by photoredox catalysis, could be employed in the cyclization and cross-coupling reactions.[12] We envisioned that the construction of imidazolines or oxazolidines from alkenes might also be realized through visible-light photoredox catalysis under mild conditions.

RESULTS AND DISCUSSION

To accomplish this goal, we first examined the reaction of styrene and *N*-Ts-protected 1-aminopyridinium[13] at 20 °C in the presence of acetonitrile, a 25 W blue LED strip, and 1 mol % of $Ru(bpy)_3(PF_6)_2$. The functionalization of alkenes for the synthesis of substituted imidazolines occurred under these conditions. The choice of photocatalysts was a significant factor to promote this reaction. When $Ir(ppy)_3$ was used in place of $Ru(bpy)_3(PF_6)_2$, **3a** was obtained in 59% yield (entry 3 in Table 1), while no reaction occurred with other organic photocatalysts such as Methylene blue and Eosin Y. To further improve the yield of the desired product, 2-methyl-4-phenyl-1-tosyl-4,5-dihydro-1*H*-imidazole (**3a**), the equivalent ratio of *N*-centered radical source to styrene was increased, and the yield was dramatically improved (to 95%, entry 8 in Table 1). Finally, control experiments revealed that both the photocatalyst and the light source were necessary for the synthesis of imidazoline derivatives.

Having successfully identified the optimal reaction conditions, we next turned our attention to examining the scope of the substrates. As shown in Table 2, a variety of substituted alkenes were readily converted to the corresponding imidazolines using $Ir(ppy)_3$ as the photocatalyst. Interestingly, when *N*-benzenesulfonyl protected 1-aminopyridinium salt was used instead of the *N*-Ts-protected reactant, the yield of the product was only slightly diminished (**3a** vs **3b**). Substrates with electron-withdrawing (**3c**−**3g**) and electron-donating (**3h**−**3k**) groups in the aromatic ring, including a halogen atom and a methyl group, could proceed smoothly to give the corresponding substituted imidazolines in good to excellent yields. Notably, the substrate with a bulky *tert*-butyl group was also converted into the corresponding target compound **3k** in 84% yield. Furthermore, transformations of the indene and 1,2-dihydronaphthalene were observed as well in high yields without any undesired diastereomers (**3l**−**3m**). In addition, α-methylstyrenes were also readily transformed into the imidazolines bearing a tetrasubstituted carbon quaternary stereocenter

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Scheme 1. Synthesis of Imidazoline and Oxazolidine Derivatives

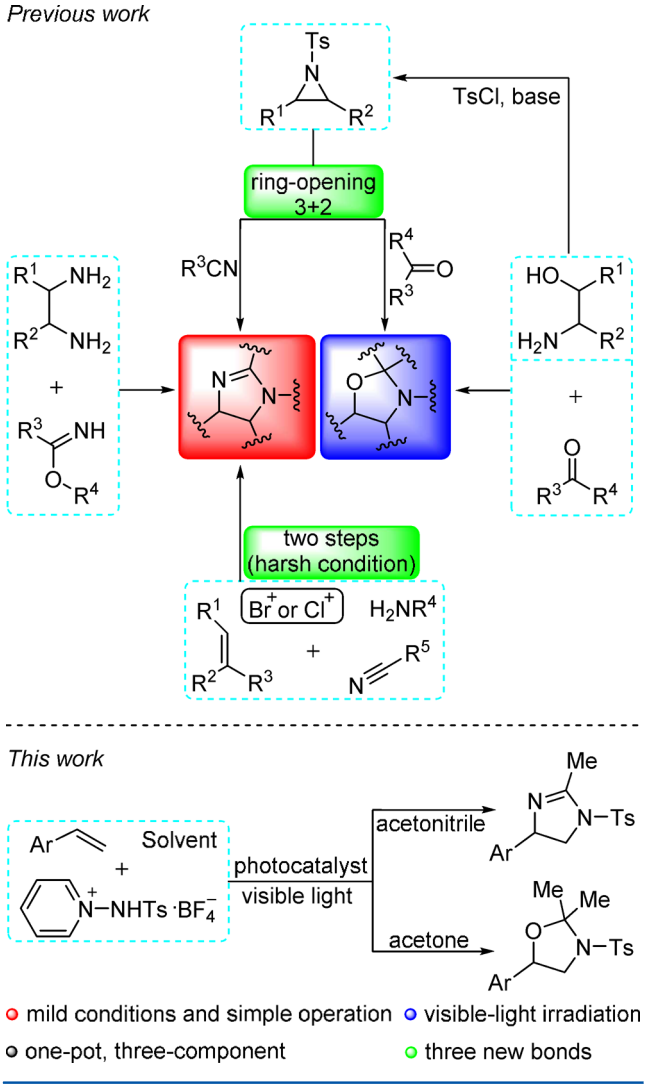
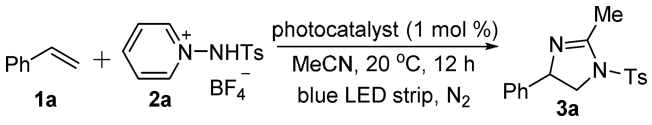


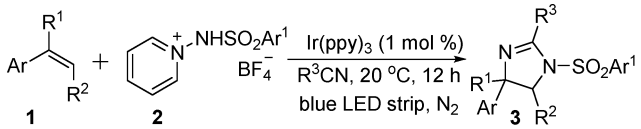
Table 1. Screening of the Reaction Conditions for the Synthesis of Imidazoline[a]

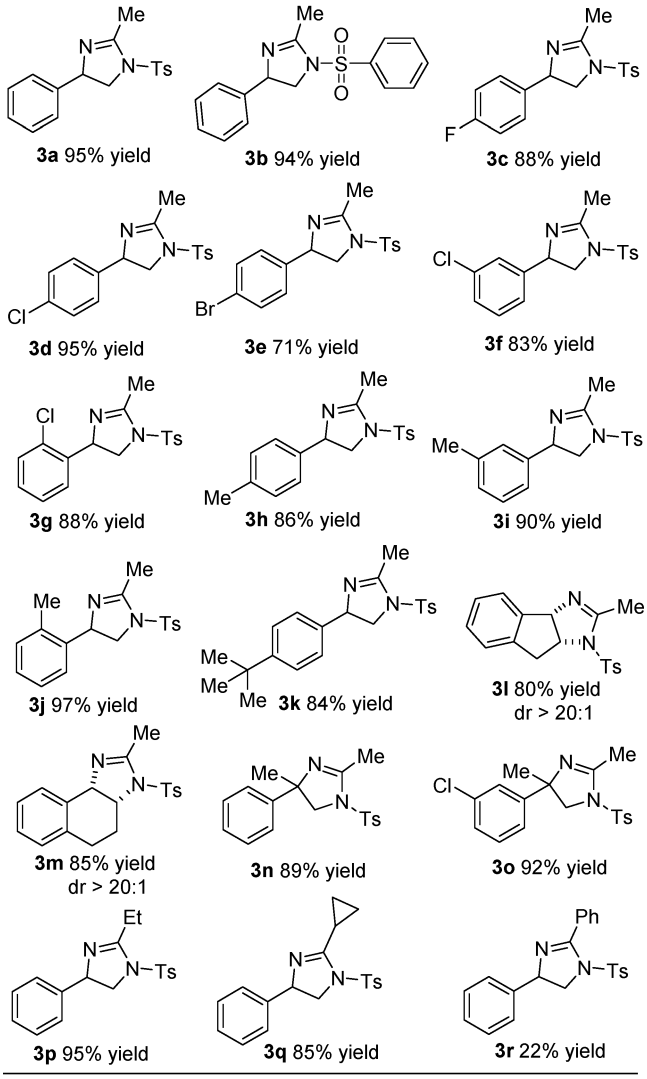


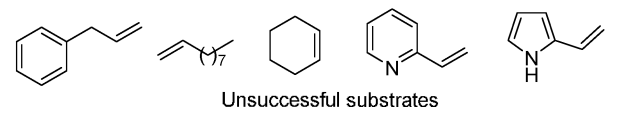
entry	photocatalyst	1a:2a	yield (%)[b]
1	$Ru(bpy)_3(PF_6)_2$	1:1.5	52
2	$Ir(ppy)_2(dtbbpy)PF_6$	1:1.5	58
3	$Ir(ppy)_3$	1:1.5	59
4	Methylene blue	1:1.5	0
5[c]	Eosin Y	1:1.5	0
6	$Ir(ppy)_3$	1:2	68
7	$Ir(ppy)_3$	1:3	78
8	$Ir(ppy)_3$	1:4	95
9	–	1:4	0
10[d]	$Ir(ppy)_3$	1:4	0

[a]Unless otherwise noted, reaction conditions are as follows: **1a** (0.2 mmol), **2a** (1.5–4 equiv), photocatalyst (0.002 mmol), CH_3CN (4 mL), 25 W blue LED strip, 20 °C, under a N_2 atmosphere. [b]Isolated yield. [c]25 W white LED strip was used. [d]In the absence of a light source.

Table 2. Scope of the Substrates for the Synthesis of Imidazoline Derivatives[a,b]







[a]Reaction conditions: substrate **1** (0.2 mmol), substrate **2** (0.8 mmol), $Ir(ppy)_3$ (0.002 mmol) in R^3CN (anhydrous, 4 mL) at 20 °C under irradiation by 25 W blue LED strip for 12 h. [b]Isolated yields.

moiety in high yield (**3n–3o**). Except for acetonitrile, other nitriles, such as propiononitrile, cyclopropane-carbonitrile, and benzonitrile, could also achieve this kind of transformation under the photoredox catalysis conditions (**3p–3r**). However, the transformation of some heterocyclic aromatic and non-conjugated alkenes was not success.

Encouraged by the results described above, we expected that the substituted oxazolidines might also be generated under similar reaction conditions. The transformation was smoothly realized (entry 1 in Table 3) under similar reaction conditions when $Ir(ppy)_3$ was used as the photocatalyst, and the

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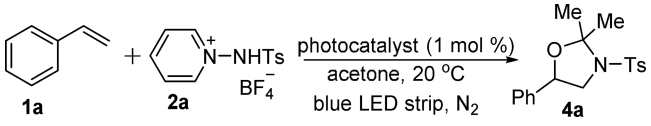
Table 3. Screening of the Reaction Conditions for the Synthesis of Oxazolidine[a]



entry	photocatalyst	time (h)	1a:2a	yield (%)[b]
1	$Ir(ppy)_3$	12	1:1.5	80
2	$Ru(bpy)_3(PF_6)_2$	3	1:1.5	77
3	$Ir(ppy)_2(dtbbpy)PF_6$	2.5	1:1.5	88
4	$Ir(ppy)_2(dtbbpy)PF_6$	2.5	1:2	55
5	$Ir(ppy)_2(dtbbpy)PF_6$	2.5	1:1	75

[a]Unless otherwise noted, reaction conditions are as follows: substrate **1a** (0.2 mmol), **2a** (1−2 equiv), photocatalyst (0.002 mmol), acetone (anhydrous, 4 mL), 25 W blue LED strip, 20 °C, under a N_2 atmosphere. [b]Isolated yield.

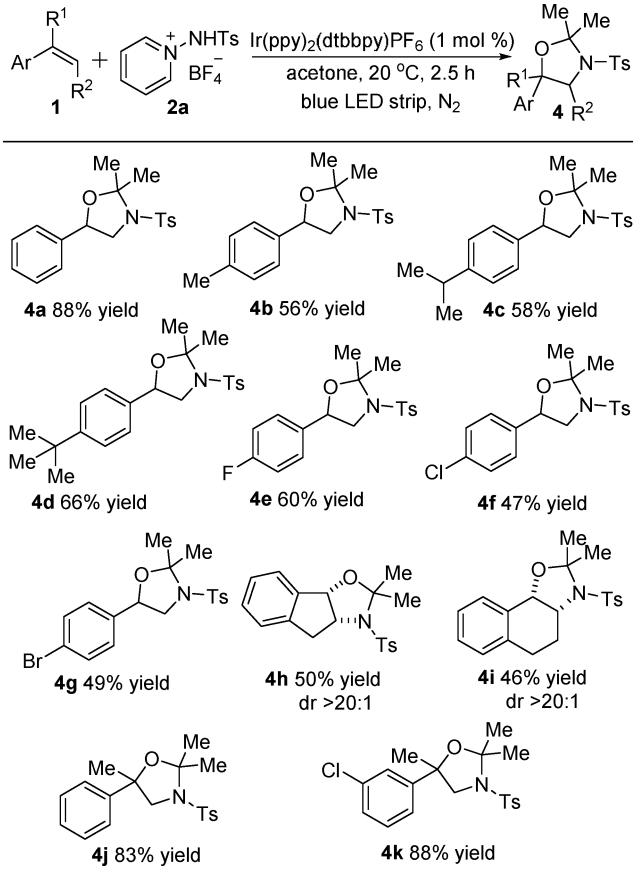
examination of the photocatalysts indicated that $Ir(ppy)_2(dtbbpy)PF_6$ produced the highest yield and resulted in the shortest time for reaction completion. Interestingly, the optimal equivalent ratio of the *N*-centered radical source to styrene was 1:1.5 in this case; either increasing or decreasing the equivalent ratio would reduce the yield of substituted oxazolidine (entries 3−5 in Table 3).

With the reaction conditions optimized, we next sought to examine the scope of olefins that can be applied to this cyclization reaction. As shown in Table 4, this protocol permitted the direct three-component cyclization of the *N*-centered radical and acetone with a wide range of unactivated olefins. Furthermore, both electron-withdrawing and -donating substituents in the aromatic ring were well accommodated in this conversion under the photoredox catalysis. A variety of substituted oxazolidines were synthesized by this simple method in good to high yields (**4a**−**4i**). Likewise, *α*-methylstyrenes could also be easily transformed into the oxazolidines bearing an all-carbon quaternary stereocenter moiety in high yields (**4j**−**4k**). In a word, the reaction described herein makes an important complementation to the transformation mentioned above.

The configuration of compound **4i** was determined by X-ray crystal structure analysis. The data of X-ray crystal structures can be found in the Supporting Information.[14]

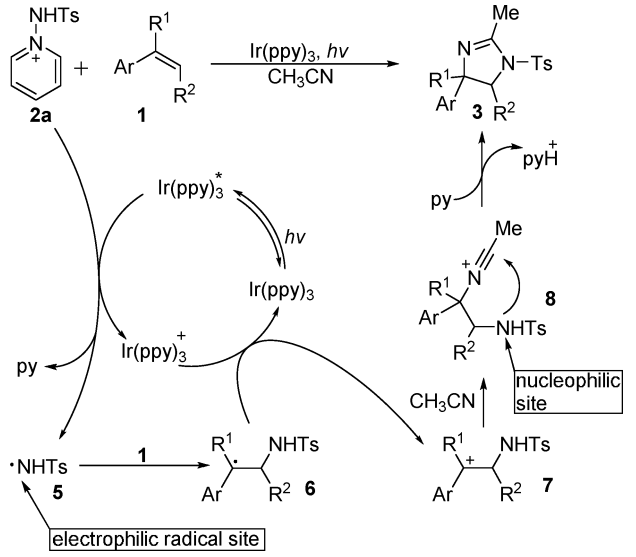
Control experiments have shown that both the photocatalyst and visible light were involved in the synthesis of substituted imidazolines. On the basis of our previous work as well as other reports,[15] a plausible reaction mechanism is illustrated in Scheme 2. Irradiation of the photoredox catalyst $Ir(ppy)_3$ with a blue LED strip will generate the photoexcited species $*Ir(ppy)_3$, which is a strong electron donor to reduce the substrate **2a**.[16] The *N*-Ts-protected 1-aminopyridinium **2a** accepts a single electron from $*Ir(ppy)_3$ and then provides the highly activated *N*-centered radical (**5**) and the oxidized $Ir(ppy)_3^+$. The species **5** will react with alkene (**1**) and then afford the stabilized carbon radical (**6**). $Ir(ppy)_3^+$ is a powerful oxidant which will be reduced by species **6** and then converted to the ground state photocatalyst. The generated carbocationic intermediate (**7**) is attacked by nitrile to form the highly reactive intermediate (**8**). Finally, the cyclization of intermediate **8** leads to the formation of the imidazoline product. Likewise, the reaction for synthesis of oxazolidine follows the similar mechanism as we proposed. We also confirmed that this nitrogen radical had multiple reaction sites.

Table 4. Scope of the Substrate for the Preparation of Oxazolidine Derivatives[a,b]



[a]Reaction conditions: substrate **1** (0.2 mmol), substrate **2a** (0.3 mmol), $Ir(ppy)_2(dtbbpy)PF_6$ (0.002 mmol) in acetone (anhydrous, 4 mL) at 20 °C under irradiation by 25 W blue LED strip for 2.5 h. [b]Isolated yields.

Scheme 2. Proposed Mechanism



CONCLUSION

In conclusion, we have developed a direct and concise approach to achieve the visible light photoredox-induced functionalization of alkenes for the synthesis of imidazoline and oxazolidine

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derivatives. This method achieved a one-pot, three-component cascade reaction through the combination of photoredox catalyzed radical addition and formal [3 + 2] annulation. In contrast to traditional methods, the advantages of this transformation are good to excellent yields, mild reaction conditions, operational simplicity, and easy accessibility of raw materials.

EXPERIMENTAL SECTION

General Information. All glassware was thoroughly oven-dried. Chemicals and solvents were either purchased from commercial suppliers or purified by standard techniques. Thin-layer chromatography plates were visualized by exposure to ultraviolet light and/or staining with phosphomolybdic acid followed by heating on a hot plate. Flash chromatography was carried out using silica gel (200−300 mesh). ^{1}H NMR and ^{13}C NMR spectra were recorded on a 400 MHz spectrometer. The spectra were recorded in deuterochloroform ($CDCl_3$) as solvent at room temperature; ^{1}H and ^{13}C NMR chemical shifts are reported in ppm relative to the residual solvent peak. The residual solvent signals were used as references, and the chemical shifts were converted to the TMS scale ($CDCl_3$: δ_H = 7.26 ppm, δ_C = 77.0 ppm). Data for ^{1}H NMR are reported as follows: chemical shift (δ ppm), multiplicity (s = singlet, d = doublet, t = triplet, m = multiplet, dd = doublet, br = broad), integration, coupling constant (Hz), and assignment. Data for ^{13}C NMR are reported as chemical shift. IR spectra were recorded on an FT-IR instrument and are reported in wave numbers (cm^{-1}). HRMS spectra using ESI were recorded on an ESI-FTMS mass spectrometer.

General Procedure for the Synthesis of Imidazoline Derivatives. Substrate **1** (0.2 mmol) and *N*-Ts-protected 1-aminopyridinium **2** (0.8 mmol) were added to a solution of photocatalyst $Ir(ppy)_3$ (1 mol %) in nitrile (4 mL) at room temperature. The heterogeneous mixture was degassed by three cycles of freeze−pump−thaw and then placed in the irradiation apparatus equipped with a 25 W blue light-emitting diode (LED) strip. The resulting mixture was stirred at 20 °C until the starting material was completely consumed after 12 h. Upon completion of the reaction, the reaction mixture was evaporated under reduced pressure, and the resulting crude mixture was purified on silica gel flash column chromatography using ethyl acetate/hexanes (1/2) eluent to give the corresponding imidazoline derivatives.

General Procedure for the Synthesis of Oxazolidine Derivatives. Substrate **1** (0.2 mmol) and *N*-Ts-protected 1-aminopyridinium **2** (0.3 mmol) were added to a solution of photocatalyst $Ir(ppy)_2(dtbbpy)PF_6$ (1 mol %) in acetone (4 mL) at room temperature. The heterogeneous mixture was degassed by three cycles of freeze−pump−thaw and then placed in the irradiation apparatus equipped with a 25 W blue light-emitting diode (LED) strip. The resulting mixture was stirred at 20 °C until the starting material was completely consumed after 2.5 h. Upon completion of the reaction, the reaction mixture was evaporated under reduced pressure, and the resulting crude mixture was purified on silica gel flash column chromatography using ethyl acetate/hexanes (1/4) eluent to give the corresponding oxazolidine derivatives.

*2-Methyl-4-phenyl-1-tosyl-4,5-dihydro-1H-imidazole (**3a**).*[8d] Colorless oil; 59.9 mg, 95% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 2.39 (s, 3H), 2.44 (s, 3H), 3.62 (t, J = 9.5 Hz, 1H), 4.17 (t, J = 10.1 Hz, 1H), 4.98 (t, J = 9.0 Hz, 1H), 7.04 (dd, J = 6.9, 1.6 Hz, 2H), 7.21−7.28 (m, 3H), 7.34 (d, J = 8.1 Hz, 2H), 7.74 (d, J = 8.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 16.7, 21.5, 55.4, 66.5, 126.3, 127.1, 127.5, 128.6, 130.0, 135.1, 141.5, 144.7, 156.3; IR (KBr, cm^{-1}): 3370, 3282, 3063, 2924, 1646, 1598, 1495, 1359, 1242, 1166, 1115, 1024, 969, 815, 701, 659, 593, 547. HRMS (ESI) for $C_{17}H_{19}N_2O_2S$ [M + H]$^+$ calcd 315.1162, found 315.1172

*2-Methyl-4-phenyl-1-(phenylsulfonyl)-4,5-dihydro-1H-imidazole (**3b**).* Colorless oil; 56.4 mg, 94% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 2.41 (s, 3H), 3.64 (dd, J = 9.6, 8.1 Hz, 1H), 4.19 (t, J = 10.1 Hz, 1H), 5.00 (t, J = 8.8 Hz, 1H), 7.04 (dd, J = 6.9, 1.4 Hz, 2H), 7.22−7.29 (m, 3H), 7.57 (t, J = 7.7 Hz, 2H), 7.66 (t, J = 7.4 Hz, 1H), 7.74 (d, J = 7.8 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 16.8, 55.5, 66.6, 126.3, 127.1, 127.7, 128.7, 129.5, 133.7, 138.1, 141.4, 156.2; IR (KBr, cm^{-1}): 3278, 3064, 2927, 1647, 1543, 1448, 1360, 1243, 1169, 1116, 1092, 1024, 970, 757, 730, 690, 600, 570. HRMS (ESI) for $C_{16}H_{17}N_2O_2S$ [M + H]$^+$ calcd 301.1005, found 301.1004.

*4-(4-Fluorophenyl)-2-methyl-1-tosyl-4,5-dihydro-1H-imidazole (**3c**).*[17] Colorless oil; 58.5 mg, 88% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 2.39 (d, J = 1.4 Hz, 3H), 2.46 (s, 3H), 3.58 (dd, J = 9.8, 7.8 Hz, 1H), 4.16 (t, J = 10.1 Hz, 1H), 4.97 (t, J = 8.7 Hz, 1H), 6.94 (t, J = 8.7 Hz, 2H), 7.00−7.03 (m, 2H), 7.35 (d, J = 8.1 Hz, 2H), 7.74 (d, J = 8.3 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 16.8, 21.5, 55.5, 65.9, 115.5 (d, J = 21.3 Hz), 127.2, 128.0 (d, J = 8.1 Hz), 130.1, 137.4 (d, J = 3.1 Hz), 144.8, 156.6, 162.1 (d, J = 244.5 Hz); IR (KBr, cm^{-1}): 3283, 3060, 2927, 1646, 1599, 1511, 1360, 1225, 1166, 1114, 1092, 1016, 971, 836, 710, 657, 593, 546. HRMS (ESI) for $C_{17}H_{18}FN_2O_2S$ [M + H]$^+$ calcd 333.1068, found 333.1076.

*4-(4-Chlorophenyl)-2-methyl-1-tosyl-4,5-dihydro-1H-imidazole (**3d**).*[17] Colorless oil; 66.0 mg, 95% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 2.39 (d, J = 1.4 Hz, 3H), 2.40 (s, 3H), 3.57 (dd, J = 9.8, 7.8 Hz, 1H), 4.17 (t, J = 10.1 Hz, 1H), 4.97 (t, J = 8.9 Hz, 1H), 6.98 (d, J = 8.4 Hz, 2H), 7.23 (d, J = 8.4 Hz, 2H), 7.35 (d, J = 8.1 Hz, 2H), 7.73 (d, J = 8.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 16.8, 21.6, 55.4, 65.8, 127.2, 127.7, 128.8, 130.1, 133.4, 135.0, 140.1, 144.9, 156.9; IR (KBr, cm^{-1}): 3276, 3059, 2926, 1647, 1598, 1493, 1360, 1166, 1115, 1091, 1015, 971, 816, 736, 656, 593, 547. HRMS (ESI) for $C_{17}H_{18}ClN_2O_2S$ [M + H]$^+$ calcd 349.0772, found 349.0782.

*4-(4-Bromophenyl)-2-methyl-1-tosyl-4,5-dihydro-1H-imidazole (**3e**).*[17] Colorless oil; 55.7 mg, 71% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 2.39 (s, 3H), 2.45 (s, 3H), 3.56 (dd, J = 9.8, 7.9 Hz, 1H), 4.16 (t, J = 10.1 Hz, 1H), 4.95 (t, J = 8.7 Hz, 1H), 6.92 (d, J = 8.3 Hz, 2H), 7.32−7.38 (m, 4H), 7.72 (d, J = 8.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 16.7, 21.5, 55.3, 65.8, 121.4, 127.1, 128.0, 130.1, 131.6, 135.0, 140.6, 144.8, 156.8; IR (KBr, cm^{-1}): 3276, 3055, 2924, 1646, 1597, 1488, 1359, 1242, 1166, 1116, 1091, 1011, 971, 816, 709, 663, 593, 547. HRMS (ESI) for $C_{17}H_{18}BrN_2O_2S$ [M + H]$^+$ calcd 393.0267, found 393.0279.

*4-(3-Chlorophenyl)-2-methyl-1-tosyl-4,5-dihydro-1H-imidazole (**3f**).*[8d] Colorless oil; 58.0 mg, 83% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 2.40 (d, J = 1.3 Hz, 3H), 2.45 (s, 3H), 3.57 (dd, J = 9.9, 7.7 Hz, 1H), 4.18 (t, J = 10.2 Hz, 1H), 4.96 (t, J = 9.0 Hz, 1H), 6.94−6.97 (m, 2H), 7.20 (d, J = 5.2 Hz, 2H), 7.34 (d, J = 8.2 Hz, 2H), 7.73 (d, J = 8.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 16.8, 21.6, 55.2, 66.0, 124.6, 126.4, 127.1, 127.7, 129.9, 130.1, 134.5, 134.9, 143.7, 144.9, 156.9; IR (KBr, cm^{-1}): 3381, 3062, 2927, 1645, 1597, 1360, 1242, 1167, 1116, 1025, 787, 657, 593, 546. HRMS (ESI) for $C_{17}H_{18}ClN_2O_2S$ [M + H]$^+$ calcd 349.0772, found 349.0771.

*4-(2-Chlorophenyl)-2-methyl-1-tosyl-4,5-dihydro-1H-imidazole (**3g**).*[8d] Colorless oil; 61.1 mg, 88% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 2.43 (s, 6H), 3.53 (dd, J = 9.9, 7.7 Hz, 1H), 4.30 (t, J = 10.2 Hz, 1H), 5.31 (t, J = 8.6 Hz, 1H), 7.08−7.10 (m, 1H), 7.13−7.21 (m, 2H), 7.30−7.34 (m, 3H), 7.72 (d, J = 8.3 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 16.9, 21.5, 54.7, 63.7, 127.0, 127.1, 127.4, 128.6, 129.3, 130.0, 132.2, 135.2, 139.5, 144.7, 157.4; IR (KBr, cm^{-1}): 3379, 3064, 2924, 1648, 1473, 1437, 1360, 1244, 1186, 1118, 1092, 1030, 970, 815, 756, 731, 657, 593, 546. HRMS (ESI) for $C_{17}H_{18}ClN_2O_2S$ [M + H]$^+$ calcd 349.0772, found 349.0770.

*2-Methyl-4-(p-tolyl)-1-tosyl-4,5-dihydro-1H-imidazole (**3h**)..*[8b,d] Colorless oil; 56.7 mg, 86% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 2.94 (s, 3H), 2.37 (d, J = 1.5 Hz, 3H), 2.45 (s, 3H), 3.60 (dd, J = 9.8, 8.0 Hz, 1H), 4.14 (t, J = 10.0 Hz, 1H), 4.94 (t, J = 8.6 Hz, 1H), 6.92 (d, J = 8.0 Hz, 2H), 7.06 (d, J = 7.9 Hz, 2H), 7.34 (d, J = 8.1 Hz, 2H), 7.73 (d, J = 8.3 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 16.7, 21.0, 21.5, 55.5, 66.3, 126.2, 127.1, 129.2, 130.0, 135.1, 137.2, 138.5, 144.6, 156.1; IR (KBr, cm^{-1}): 3279, 3027, 2923, 1647, 1597, 1515, 1432, 1358, 1241, 1166, 1116, 1091, 1020, 969, 816, 711, 657, 594, 546. HRMS (ESI) for $C_{18}H_{21}N_2O_2S$ [M + H]$^+$ calcd 329.1318, found 329.1315.

*2-Methyl-4-(m-tolyl)-1-tosyl-4,5-dihydro-1H-imidazole (**3i**).*[8d] Colorless oil; 58.9 mg, 90% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 2.56 (s, 3H), 2.39 (s, 3H), 2.45 (s, 3H), 3.62 (dd, J = 9.6, 8.1

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J. Org. Chem. 2017, 82, 243−249

Hz, 1H), 4.16 (t, J = 10.1 Hz, 1H), 4.95 (t, J = 9.0 Hz, 1H), 6.80 (s, 1H), 6.84 (d, J = 7.6 Hz, 1H), 7.04 (d, J = 7.5 Hz, 1H), 7.14 (t, J = 7.5 Hz, 1H), 7.34 (d, J = 8.1 Hz, 2H), 7.75 (d, J = 8.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 16.8, 21.1, 21.5, 55.4, 66.5, 123.4, 126.9, 127.2, 128.3, 128.5, 130.0, 135.1, 138.3, 141.4, 144.6, 156.2; IR (KBr, cm^{-1}): 3281, 2923, 1646, 1597, 1431, 1359, 1167, 1114, 1092, 1024, 815, 788, 710, 659, 594, 546. HRMS (ESI) for $C_{18}H_{21}N_2O_2S$ [M + H]$^+$ calcd 329.1318, found 329.1326.

2-Methyl-4-(o-tolyl)-1-tosyl-4,5-dihydro-1H-imidazole (**3j**).[8d] Colorless oil; 64.0 mg, 97% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 2.27 (s, 3H), 2.42 (d, J = 1.4 Hz, 3H), 2.45 (s, 3H), 3.49 (dd, J = 9.4, 8.2 Hz, 1H), 4.22 (t, J = 10.0 Hz, 1H), 5.18 (t, J = 9.2 Hz, 1H), 6.92 (d, J = 7.5 Hz, 1H), 7.06−7.10 (m, 1H), 7.11−7.14 (m, 2H), 7.33 (d, J = 8.0 Hz, 2H), 7.33 (d, J = 8.3 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 16.9, 19.4, 21.6, 54.9, 63.5, 125.7, 126.4, 127.2, 127.3, 130.1, 130.3, 134.5, 135.3, 140.0, 144.7, 156.5; IR (KBr, cm^{-1}): 3278, 3060, 2928, 1649, 1597, 1510, 1461, 1359, 1271, 1167, 1117, 1091, 968, 815, 758, 711, 658, 594, 545. HRMS (ESI) for $C_{18}H_{21}N_2O_2S$ [M + H]$^+$ calcd 329.1318, found 329.1315.

4-(4-(tert-Butyl)phenyl)-2-methyl-1-tosyl-4,5-dihydro-1H-imidazole (**3k**). Colorless oil; 62.2 mg, 84% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 2.28 (s, 9H), 2.37 (d, J = 1.4 Hz, 3H), 2.45 (s, 3H), 3.65 (dd, J = 9.7, 8.0 Hz, 1H), 4.14 (t, J = 10.0 Hz, 1H), 4,95 (t, J = 8.7 Hz, 1H), 6.97 (d, J = 8.3 Hz, 2H), 7.27 (d, J = 8.3 Hz, 2H), 7.35 (d, J = 8.2 Hz, 2H), 7.75 (d, J = 8.3 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 16.7, 21.5, 31.2, 34.4, 55.3, 66.2, 125.5, 126.0, 127.2, 130.0, 135.2, 138.4, 144.6, 150.5, 156.0; IR (KBr, cm^{-1}): 3055, 2962, 1647, 1598, 1510, 1474, 1360, 1242, 1167, 1112, 1019, 971, 815, 656, 594, 546. HRMS (ESI) for $C_{21}H_{27}N_2O_2S$ [M + H]$^+$ calcd 371.1788, found 371.1786.

2-Methyl-1-tosyl-1,3a,8,8a-tetrahydroindeno[1,2-d]imidazole (**3l**). White solid; 52.1 mg, 80% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 2.25 (s, 3H), 2.46 (s, 3H), 3.43−3.57 (m, 2H), 4.83 (t, J = 6.7 Hz, 1H), 5.42 (d, J = 8.7 Hz, 1H), 7.24−7.27 (m, 3H), 7.37 (d, J = 8.0 Hz, 2H), 7.46 (d, J = 7.2 Hz, 1H), 7.78 (d, J = 8.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 16.8, 21.6, 41.2, 63.7, 74.7, 125.1, 125.3, 127.2, 127.4, 128.6, 130.1, 135.9, 139.8, 140.7, 144.8, 156.3; IR (KBr, cm^{-1}): 3028, 2927, 1716, 1646, 1598, 1434, 1358, 1235, 1165, 1092, 1014, 966, 913, 814, 754, 691, 595, 545. HRMS (ESI) for $C_{18}H_{19}N_2O_2S$ [M + H]$^+$ calcd 327.1162, found 327.1164.

2-Methyl-3-tosyl-3a,4,5,9b-tetrahydro-3H-naphtho[1,2-d]-imidazole (**3m**).[8b] Colorless oil; 57.8 mg, 85% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 2.00−2.15 (m, 2H), 2.29 (d, J = 1.8 Hz, 3H), 2.45 (s, 3H), 2.55−2.62 (m, 1H), 2.70−2.76 (m, 1H), 4.43−4.48 (m, 1H), 4,85 (d, J = 9.5 Hz, 1H), 7.08 (d, J = 7.3 Hz, 1H), 7.14−7.23 (m, 2H), 7.36 (d, J = 8.0 Hz, 2H), 7.43 (d, J = 7.4 Hz, 1H), 7.78 (d, J = 8.3 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 17.1, 21.5, 25.8, 28.5, 60.6, 65.6, 126.8, 126.9, 127.2, 128.1, 129.6, 130.1, 133.8, 136.6, 137.7, 144.5, 155.5; IR (KBr, cm^{-1}): 3278, 3062, 2929, 1647, 1598, 1494, 1454, 1356, 1244, 1164, 1096, 1021, 911, 814, 734, 664, 602, 546. HRMS (ESI) for $C_{19}H_{21}N_2O_2S$ [M + H]$^+$ calcd 341.1318, found 341.1322.

2,4-Dimethyl-4-phenyl-1-tosyl-4,5-dihydro-1H-imidazole (**3n**). Colorless oil; 58.3 mg, 89% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 1.46 (s, 3H), 2.37 (s, 3H), 2.41 (s, 3H), 3.79 (d, J = 9.6 Hz, 1H), 3.91 (d, J = 9.6 Hz, 1H), 7.20−7.30 (m, 7H), 7.70 (d, J = 8.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 16.8, 21.5, 29.5, 61.0, 69.5, 124.9, 126.8, 127.0, 128.4, 130.0, 135.3, 144.5, 146.3, 154.1; IR (KBr, cm^{-1}): 3282, 2969, 2926, 1647, 1447, 1358, 1167, 1090, 1041, 924, 815, 765, 717, 664, 593, 545. HRMS (ESI) for $C_{18}H_{21}N_2O_2S$ [M + H]$^+$ calcd 329.1318, found 329.1322.

4-(3-Chlorophenyl)-2,4-dimethyl-1-tosyl-4,5-dihydro-1H-imidazole (**3o**). Colorless oil; 66.6 mg, 92% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 1.45 (s, 3H), 2.38 (s, 3H), 2.42 (s, 3H), 3.74 (d, J = 9.8 Hz, 1H), 3.89 (d, J = 9.8 Hz, 1H), 7.09−7.11 (m, 1H), 7.16−7.22 (m, 3H), 7.30 (d, J = 8.1 Hz, 2H), 7.69 (d, J = 8.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 16.9, 21.5, 29.4, 60.9, 69.3, 123.2, 125.5, 127.0, 129.7, 130.0, 134.3, 135.1, 144.7, 148.5, 154.7; IR (KBr, cm^{-1}): 3367, 2973, 2926, 1736, 1648, 1597, 1475, 1359, 1283, 1168, 1118, 1041, 1010, 925, 814, 743, 696, 660, 594, 546. HRMS (ESI) for $C_{18}H_{20}ClN_2O_2S$ [M + H]$^+$ calcd 363.0929, found 363.0926.

2-Ethyl-4-phenyl-1-tosyl-4,5-dihydro-1H-imidazole (**3p**).[18] Colorless oil; 62.2 mg, 95% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 1.29 (t, J = 7.4 Hz, 3H), 2.44 (s, 3H), 2.77 (m, 2H), 3.61 (dd, J = 9.8, 7.8 Hz, 1H), 4.17 (t, J = 10.1 Hz, 1H), 4.99 (dd, J = 9.9, 8.0 Hz, 1H), 7.02−7.04 (m, 2H), 7.22−7.27 (m, 3H), 7.33 (d, J = 8.0 Hz, 2H), 7.73 (d, J = 8.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 11.0, 21.5, 23.3, 55.8, 66.6, 126.3, 127.2, 127.5, 128.6, 130.0, 135.3, 141.8, 144.6, 160.8; IR (KBr, cm^{-1}): 3296, 3030, 2925, 1736, 1637, 1598, 1494, 1471, 1401, 1359, 1244, 1162, 1097, 994, 815, 701, 660, 587, 567. HRMS (ESI) for $C_{18}H_{21}N_2O_2S$ [M + H]$^+$ calcd 329.1318, found 329.1315.

2-Cyclopropyl-4-phenyl-1-tosyl-4,5-dihydro-1H-imidazole (**3q**). Colorless oil; 58.0 mg, 85% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 0.97−1.12 (m, 4H), 2.42−2.50 (m, 4H), 3.60 (dd, J = 10.1, 7.4 Hz, 1H), 4.19 (t, J = 10.2 Hz, 1H), 4.89 (dd, J = 10.2, 7.5 Hz, 1H), 6.89−6.91 (m, 2H), 7.19−7.21 (m, 3H), 7.29 (d, J = 8.1 Hz, 2H), 7.79 (d, J = 8.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 8.6, 9.7, 10.4, 21.5, 56.4, 65.9, 126.2, 127.3, 127.4, 128.5, 129..9, 135.0, 142.1, 144.5, 161.7; IR (KBr, cm^{-1}): 3290, 2980, 2931, 1736, 1647, 1535, 1454, 1331, 1243, 1161, 1094, 913, 815, 701, 663, 590, 549. HRMS (ESI) for $C_{19}H_{21}N_2O_2S$ [M + H]$^+$ calcd 341.1318, found 341.1315.

2,4-Diphenyl-1-tosyl-4,5-dihydro-1H-imidazole (**3r**)..[8b,d] Colorless oil; 16.4 mg, 22% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 2.42 (s, 3H), 3.86 (dd, J = 11.4, 8.1 Hz, 1H), 4.43 (dd, J = 11.3, 10.2 Hz, 1H), 4.99 (dd, J = 9.8, 8.3 Hz, 1H), 6.96−6.98 (m, 2H), 7.18−7.22 (m, 5H), 7.39−7.45 (m, 4H), 7.52 (t, J = 7.4 Hz, 1H), 7.77 (d, J = 7.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 21.6, 56.9, 67.9, 126.3, 127.4, 127.6, 127.7, 128.6, 129.7, 129.8, 130.2, 131.1, 134.5, 141.5, 144.6, 159.8; IR (KBr, cm^{-1}): 3062, 2924, 1628, 1598, 1509, 1449, 1365, 1169, 1089, 1023, 911, 815, 732, 698, 663, 605, 546. HRMS (ESI) for $C_{22}H_{21}N_2O_2S$ [M + H]$^+$ calcd 377.1318, found 377.1316.

2,2-Dimethyl-5-phenyl-3-tosyloxazolidine (**4a**)..[8a,b] White solid; 58.3 mg, 88% yield; mp 89−91 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 1.68 (s, 3H), 1.74 (s, 3H), 2.42 (s, 3H), 3.14 (t, J = 9.1 Hz, 1H), 3.87 (dd, J = 8.7, 5.7 Hz, 1H), 5.10 (dd, J = 9.5, 5.7 Hz, 1H), 7.28−7.35 (m, 7H), 7.75 (d, J = 8.3 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 21.5, 27.0, 27.5, 54.0, 76.2, 97.3, 126.2, 127.3, 128.5, 128.5, 129.6, 137.3, 137.4, 143.4; IR (KBr, cm^{-1}): 3033, 2985, 2878, 1737, 1599, 1496, 1370, 1344, 1245, 1218, 1155, 1095, 1060, 1028, 955, 816, 759, 709, 659, 593, 548. HRMS (ESI) for $C_{18}H_{22}NO_3S$ [M + H]$^+$ calcd 332.1315, found 332.1314.

2,2-Dimethyl-5-(p-tolyl)-3-tosyloxazolidine (**4b**).[19] White solid; 38.6 mg, 56% yield; mp 120−122 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 1.68 (s, 3H), 1.73 (s, 3H), 2.33 (s, 3H), 2.42 (s, 3H), 3.14 (t, J = 9.1 Hz, 1H), 3.85 (dd, J = 8.7, 5.7 Hz, 1H), 5.08 (dd, J = 9.4, 5.6 Hz, 1H), 7.14 (d, J = 8.0 Hz, 2H), 7.20 (d, J = 8.1 Hz, 2H), 7.30 (d, J = 8.0 Hz, 2H), 7.76 (d, J = 8.1 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 21.1, 21.4, 27.1, 27.5, 54.0, 76.1, 97.2, 126.1, 127.3, 129.2, 129.6, 134.3, 137.3, 138.3, 143.3; IR (KBr, cm^{-1}): 3363, 2923, 2852, 1737, 1657, 1598, 1459, 1371, 1343, 1244, 1154, 1094, 954, 813, 687, 653, 592, 547. HRMS (ESI) for $C_{19}H_{24}NO_3S$ [M + H]$^+$ calcd 346.1471, found 346.1470.

5-(4-Isopropylphenyl)-2,2-dimethyl-3-tosyloxazolidine (**4c**). White solid; 43.4 mg, 58% yield; mp 102−103 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 1.21 (d, J = 6.9 Hz, 6H), 1.67 (s, 3H), 1.72 (s, 3H), 2.42 (s, 3H), 2,83−2.93 (m, 1H), 3.16 (t, J = 9.1 Hz, 1H), 3.84 (dd, J = 8.7, 5.7 Hz, 1H), 5.07 (dd, J = 9.5, 5.6 Hz, 1H), 7.17−7.24 (m, 4H), 7.29 (d, J = 8.0 Hz, 2H), 7.75 (d, J = 8.0 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 21.4, 23.8, 23.9, 27.0, 27.5, 33.8, 76.1, 97.2, 126.3, 126.6, 127.3, 129.6, 134.6, 137.3, 143.3, 149.4; IR (KBr, cm^{-1}): 2961, 2872, 1738, 1599, 1461, 1371, 1345, 1244, 1156, 1094, 956, 828, 687, 654, 593, 549. HRMS (ESI) for $C_{21}H_{28}NO_3S$ [M + H]$^+$ calcd 374.1784, found 374.1782.

5-(4-(tert-Butyl)phenyl)-2,2-dimethyl-3-tosyloxazolidine (**4d**). White solid; 51.2 mg, 66% yield; mp 99−100 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 1.29 (s, 9H), 1.67 (s, 3H), 1.73 (s, 3H), 2.42 (s, 3H), 3.16 (t, dd, J = 9.1 Hz, 1H), 3.85 (dd, J = 8.7, 5.6 Hz, 1H),

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J. Org. Chem. 2017, 82, 243−249

5.08 (dd, J = 9.5, 5.7 Hz, 1H), 7.24 (d, J = 8.3 Hz, 2H), 7.29 (d, J = 8.2 Hz, 2H), 7.35 (d, J = 8.3 Hz, 2H), 7.75 (d, J = 8.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 21.4, 27.0, 27.5, 31.2, 34.5, 53.8, 76.1, 97.2, 125.5, 126.0, 127.3, 129.6, 134.2, 137.3, 143.3, 151.6; IR (KBr, cm^{-1}): 2961, 1738, 1599, 1461, 1345, 1245, 1222, 1156, 1094, 1015, 956, 826, 711, 684, 653, 593, 549. HRMS (ESI) for $C_{22}H_{30}NO_3S$ $[M + H]^+$ calcd 388.1941, found 388.1940.

*5-(4-Fluorophenyl)-2,2-dimethyl-3-tosyloxazolidine (**4e**).* White solid; 42.0 mg, 60% yield; mp 91−93 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 1.68 (s, 3H), 1.72 (s, 3H), 2.42 (s, 3H), 3.11 (t, J = 9.1 Hz, 1H), 3.84 (dd, J = 8.7, 5.7 Hz, 1H), 5.07 (dd, J = 9.4, 5.7 Hz, 1H), 7.01 (t, J = 8.6 Hz, 2H), 7.26−7.31 (m, 4H), 7.75 (d, J = 8.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 21.5, 27.1, 27.4, 54.0, 75.6, 97.3, 115.5 (J = 21.7 Hz), 127.3, 128.0 (J = 8.1 Hz), 129.6, 133.2 (J = 3.0 Hz), 137.2, 143.5, 162.7 (J = 245.6 Hz); IR (KBr, cm^{-1}): 2985, 2929, 1606, 1513, 1459, 1370, 1342, 1225, 1153, 1093, 1014, 956, 835, 710, 688, 654, 592. HRMS (ESI) for $C_{18}H_{21}FNO_3S$ $[M + H]^+$ calcd 350.1221, found 350.1219.

*5-(4-Chlorophenyl)-2,2-dimethyl-3-tosyloxazolidine (**4f**).*[19] White solid; 34.2 mg, 47% yield; mp 112−114 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 1.67 (s, 3H), 1.72 (s, 3H), 2.42 (s, 3H), 3.09 (t, J = 9.1 Hz, 1H), 3.85 (dd, J = 8.8, 5.8 Hz, 1H), 5.07 (dd, J = 9.3, 5.7 Hz, 1H), 7.22−7.31 (m, 6H), 7.74 (d, J = 8.3 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 21.5, 27.0, 27.4, 53.5, 75.5, 97.4, 127.3, 127.5, 128.7, 129.6, 134.2, 136.1, 137.2, 143.5; IR (KBr, cm^{-1}): 3376, 2925, 1736, 1598, 1509, 1459, 1342, 1245, 1154, 1092, 1015, 955, 817, 682, 592. HRMS (ESI) for $C_{18}H_{21}ClNO_3S$ $[M + H]^+$ calcd 366.0925, found 366.0926.

*5-(4-Bromophenyl)-2,2-dimethyl-3-tosyloxazolidine (**4g**).* White solid; 40.0 mg, 49% yield; mp 100−102 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 1.67 (s, 3H), 1.72 (s, 3H), 2.42 (s, 3H), 3.09 (t, J = 9.0 Hz, 1H), 3.86 (dd, J = 8.7, 5.8 Hz, 1H), 5.06 (dd, J = 9.2, 5.7 Hz, 1H), 7.17 (d, J = 8.4 Hz, 2H), 7.30 (d, J = 8.0 Hz, 2H), 7.45 (d, J = 8.4 Hz, 2H), 7.74 (d, J = 8.3 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 21.5, 27.0, 27.4, 53.8, 75.5, 97.4, 122.3, 127.3, 127.8, 129.6, 131.7, 136.6, 137.1, 143.5; IR (KBr, cm^{-1}): 2984, 2931, 1736, 1597, 1489, 1343, 1246, 1217, 1156, 1093, 1012, 817, 676, 592, 551. HRMS (ESI) for $C_{18}H_{21}BrNO_3S$ $[M + H]^+$ calcd 410.0420, found 410.0418.

*2,2-Dimethyl-3-tosyl-3,3a,4,8b-tetrahydro-2H-indeno[2,1-d]-oxazole (**4h**).* White solid; 34.2 mg, 50% yield; mp 111−112 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 1.60 (s, 3H), 1.64 (s, 3H), 2.44 (s, 3H), 3.05−3.20 (m, 2H), 4.58−4.63 (m, 1H), 5.45 (d, J = 6.5 Hz, 1H), 7.15 (d, J = 7.5 Hz, 1H), 7.23 (d, J = 7.1 Hz, 1H), 7.28−7.33 (m, 3H), 7.39 (d, J = 7.4 Hz, 1H), 7.81 (d, J = 8.3 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 21.5, 25.8, 29.2, 39.7, 62.2, 83.1, 100.5, 125.2, 125.4, 127.2, 127.6, 129.6, 129.8, 138.2, 138.6, 142.3, 143.5; IR (KBr, cm^{-1}): 3370, 2984, 2927, 1737, 1599, 1462, 1370, 1338, 1245, 1210, 1152, 1093, 1014, 956, 753, 585, 552. HRMS (ESI) for $C_{19}H_{21}NO_3SNa$ $[M + Na]^+$ calcd 366.1134, found 366.1133.

*2,2-Dimethyl-3-tosyl-2,3,3a,4,5,9b-hexahydronaphtho[2,1-d]-oxazole (**4i**).* White solid; 33.0 mg, 46% yield; mp 121−123 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 1.67 (s, 3H), 1.73 (s, 3H), 1.78−1.88 (m, 1H), 2.02−2.09 (m, 1H), 2.43 (s, 3H), 2.58−2.67 (m, 1H), 2.76−2.82 (m, 1H), 3.95−4.00 (m, 1H), 4.83 (d, J = 5.5 Hz, 1H), 7.13 (d, J = 6.9 Hz, 1H), 7.18−7.25 (m, 2H), 7.30 (d, J = 8.2 Hz, 3H), 7.82 (d, J = 8.3 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 21.5, 25.5, 27.3, 27.7, 30.1, 58.5, 73.1, 97.0, 126.4, 127.4, 128.4, 128.5, 129.6, 130.2, 131.9, 137.8, 138.7, 143.3; IR (KBr, cm^{-1}): 3372, 2931, 2870, 1737, 1599, 1495, 1458, 1341, 1241, 1224, 1150, 1108, 1027, 984, 751, 664, 551. HRMS (ESI) for $C_{20}H_{23}NO_3SNa$ $[M + Na]^+$ calcd 380.1291, found 380.1289.

*2,2,5-Trimethyl-5-phenyl-3-tosyloxazolidine (**4j**).* Colorless oil; 57.3 mg, 83% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 1.46 (s, 3H), 1.54 (s, 3H), 1.74 (s, 3H), 2.39 (s, 3H), 3.65−3.71 (m, 2H), 7.22−7.31 (m, 7H), 7.67 (d, J = 8.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 21.4, 27.4, 28.8, 29.1, 57.6, 81.2, 97.2, 124.6, 127.1, 127.3, 128.2, 129.4, 137.1, 143.4, 145.0; IR (KBr, cm^{-1}): 3379, 2982, 2932, 1737, 1599, 1447, 1346, 1245, 1162, 1090, 1028, 701, 662, 594, 549. HRMS (ESI) for $C_{19}H_{23}NO_3SNa$ $[M + Na]^+$ calcd 368.1291, found 368.1290.

*5-(3-Chlorophenyl)-2,2,5-trimethyl-3-tosyloxazolidine (**4k**).* Colorless oil; 66.6 mg, 88% yield; ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 1.48 (s, 3H), 1.51 (s, 3H), 1.72 (s, 3H), 2.40 (s, 3H), 3.65 (s, 2H), 7.16−7.27 (m, 6H), 7.66 (d, J = 8.1 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 21.4, 27.3, 28.7, 28.9, 57.3, 80.7, 97.3, 122.9, 124.9, 127.3, 129.5, 129.6, 134.2, 136.9, 143.5, 147.2; IR (KBr, cm^{-1}): 2983, 2932, 1737, 1598, 1473, 1414, 1372, 1347, 1244, 1091, 996, 906, 790, 684, 594, 549. HRMS (ESI) for $C_{19}H_{22}ClNO_3SNa$ $[M + Na]^+$ calcd 402.0901, found 402.0902.

■ ASSOCIATED CONTENT

Supporting Information

. The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.joc.6b02377.

Devices for the photocatalytic reactions, NOE spectra of **3l**, **4h**, X-ray crystallographic data for **4i**, ^{1}H and ^{13}C NMR spectra of the products (PDF)

Crystallographic data (CIF)

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Notes

The authors declare no competing financial interest.

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