JOC The Journal of Organic Chemistry

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"On Water" C(sp^3)−H Functionalization/C−O/C−N Bonds Formations: Synthesis of Functionalized Oxazolidines and Imidazolidines

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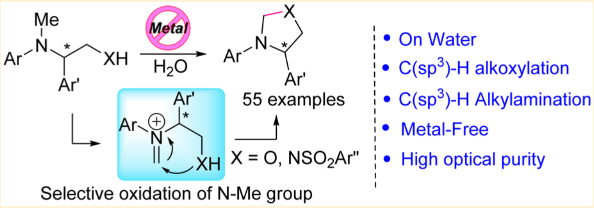
Supporting Information

ABSTRACT: On water oxidative C(sp^3)−H functionalization/C−O/C−N bonds formations using tetrabutylammonium iodide as the catalyst and *tert*-butyl hydroperoxide in water (T-Hydro) as the oxidant affords a potential route for the construction of functionalized oxazolidines and imidazolidines. The reaction is simple, regioselective, and effective at moderate temperature with broad substrate scope. In the case of optically active substrates, the oxidative cyclization can be accomplished with high optical purities.



■ INTRODUCTION

Water is highly abundant, inexpensive, nontoxic, and nonflammable. Thus, its use as a solvent for organic synthesis is attractive.[1,2] Oxazolidines[3,4] and imidazolidines[5,6] are privileged structural scaffolds that exist in many bioactive compounds (Figure 1). They are also widely employed as auxiliaries, catalysts, and ligands for transition-metal catalysts.[7,8] Generally, they are prepared by the condensation of aldehydes with the 1,2-amino alcohols[7c] and 1,2-diamines,[5a] respectively. The alternative methods for the synthesis of oxazolidines and imidazolidines include the cycloaddition,[3a,5b,9] conjugate addition,[3b] and aza-Wacker reaction.[3,10] The selective functionalization of C−H bonds is an efficient strategy for the conversion of prefunctionalized simple substrates into complex molecules with structural diversity.[11] Among them, the direct oxidative functionalization of a C−H bond next to nitrogen is attractive from both the synthetic and the enzymatic aspects.[12,13] More recently, the Rh-complex has been shown to catalyze the oxidative coupling of *N*,*N*-dimethylaniline with MeOH,[14] while the Ru-complex is employed for the amination of a benzylic C−H bond adjacent to nitrogen in the presence of visible light.[5d] Herein, we present a simple and efficient on water metal-free regioselective oxidative cross-coupling of C(sp^3)−H bonds adjacent to nitrogen with alkyl O−H and N−H bonds to furnish oxazolidines and imidazolidines from 1,2-amino alcohols and 1,2-diamines bearing *N*,*N*-arylalkyl and *N*,*N*-alkylsulfonyl substituents using tetrabutylammonium iodide (TBAI) in the presence of *tert*-butyl hydroperoxide in water (T-Hydro)[14] at moderate temperature under air. This reaction also allows the transformation of optically active substrates into the corresponding oxazolidines and imidazolidines without the loss of the optical purity that are broadly useful as chiral auxiliaries and chiral ligands for asymmetric synthesis.[7,8] The broad substrate scope, free from the contamination of metal salts, simplified product isolation, and use of water as the solvent are the significant practical features.

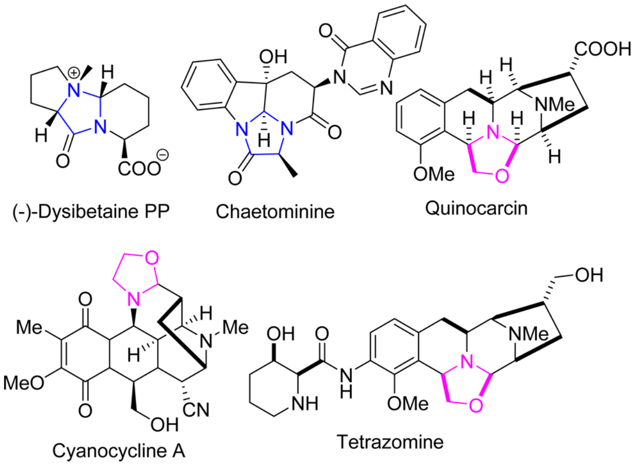


Figure 1. Examples of bioactive oxazolidine and imidazolidine containing natural products.

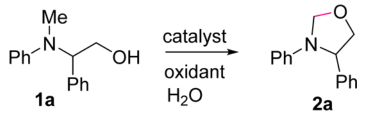
■ RESULTS AND DISCUSSION

First, the optimization of the reaction conditions was performed using amino alcohol **1a** as a model substrate in the presence of iodides and oxidants on water (Table 1). Gratifyingly, the reaction occurred to produce oxazolidine **2a** in 55% when **1a** was stirred with 15 mol % of TBAI and 2 equiv of T-Hydro at room temperature under air (entry 1). The use of 30% H_2O_2 and di-*tert*-butyl peroxide (DTBP) in place of T-hydro led to inferior results (entries 2 and 3). However, increasing the reaction temperature (60 °C) furnished the target product in 100% conversion and selectivity (entries 4 and 5). Similar results were observed using a N_2 atmosphere (entry 5). In a set of iodides screened, TBAI, NaI, KI, and I_2, the former furnished the best results (entries 6−8). Decreasing the

Received: July 30, 2016
Published: October 6, 2016

DOI: 10.1021/acs.joc.6b01850
J. Org. Chem. 2016, 81, 9792−9801

Table 1. Optimization of the Reaction Conditions



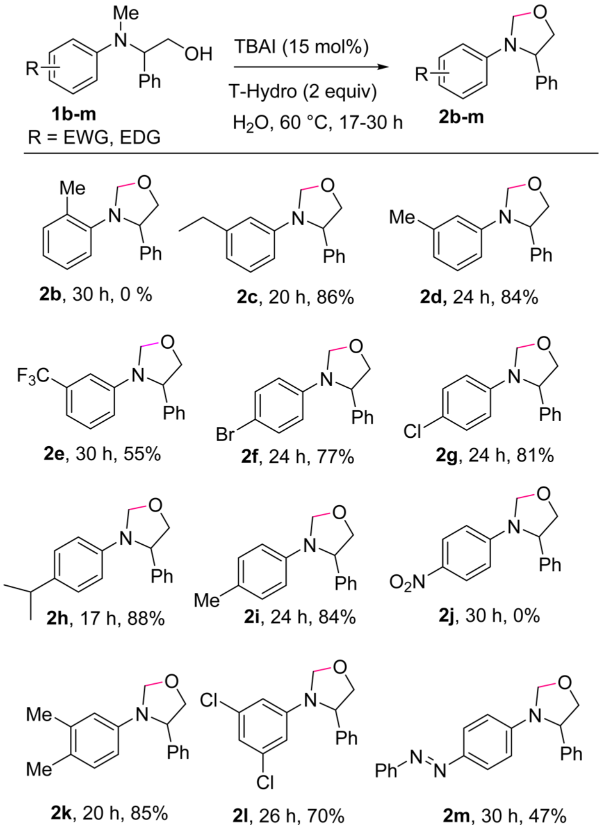
entry	catalyst (15 mol %)	oxidant (2 equiv)	temp (°C)	yield (%)[a,b]
1	TBAI	T-Hydro	rt	55
2	TBAI	30% H_2O_2	rt	5
3	TBAI	DTBP	rt	0
4	TBAI	T-Hydro	40	80
5	TBAI	T-Hydro	60	95(100)[c,d]
6	NaI	T-Hydro	60	0
7	KI	T-Hydro	60	0
8	I_2	T-Hydro	60	30
9	TBAI	T-Hydro	60	61[e], 65[f]
10		T-Hydro	60	0

[a] Amino alcohol **1a** (0.5 mmol), catalyst (15 mol %), oxidant (1 mmol), H_2O (1 mL), 24 h. [b] Isolated yield. [c] Determined by 400 MHz ^1H NMR. [d] Under a N_2 atmosphere. [e] TBAI (10 mol %) used. [f] T-Hydro (0.75 mmol) used.

amount of TBAI (10 mol %) or T-Hydro (1.5 equiv) led to the formation of **2a** in <65% yield (entry 9). A control experiment confirmed that the target heterocycle was not observed in the absence of TBAI (entry 10).

Having the optimal conditions, the reaction of amino alcohols **1b−m** having substitution in the *N*-aryl ring was investigated (Scheme 1). The substrate **1b** bearing substitution at the 2-position with a methyl group showed no product **2b** formation, which may be due to the steric hindrance of the methyl functionality. However, the reaction of **1c−e** containing substitution at the 3-position with ethyl, methyl, and trifluoromethyl groups gave **2c−e** in 55−86% yields. Similarly, the substrates **1f−i** having substitution at the 4-position with bromo, chloro, isopropyl, and methyl groups produced the corresponding oxazolidines **2f−i** in 77−88% yields. In contrast, **1j** with a strong electron-withdrawing nitro group failed to produce **2j**, which may be due to the delocalization of the nitrogen lone pair in the *N*-aryl ring toward the nitro group and not available for the single electron transfer (SET) reduction of iodine to I^- (Scheme 10, step (ii)). However, the reaction of **1k** and **1l** bearing 3,4-dimethyl and 3,5-dichloro functionalities furnished **2k** and **2l** in 85% and 70% yields, respectively, whereas **1m** containing a 4-azobenzene substituent afforded **2m** in good yield.

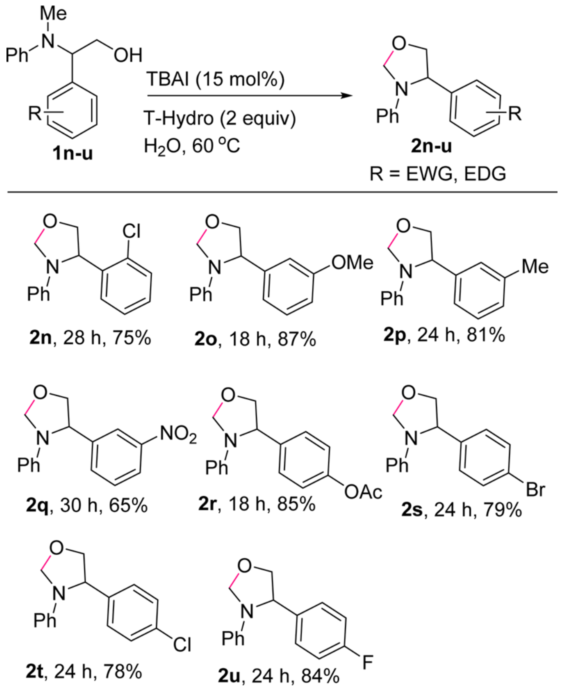
Scheme 1. Reaction of *N*-Aryl Substituted Substrates[a,b]



[a] Substrate **1b−m** (0.5 mmol), TBAI (15 mol %), T-Hydro (1 mmol), H_2O (1 mL), 60 °C. [b] Isolated yield.

Next, the reaction of the substrates bearing substitution at the 2-aryl ring was explored (Scheme 2). The substrate **1n** bearing a 2-chloro substituent underwent reaction to provide **2n** in 75% yield. The reaction of **1o−q** having substitution at the 3-position with methoxy, methyl, and nitro functionalities furnished the target oxazolidines **2o−q** in 65−87% yields, whereas **1r−u** containing substitution at the 4-position with acetoxy, bromo, chloro, and fluoro groups afforded the corresponding substituted oxazolidines **2r−u** in 78−85% yields.

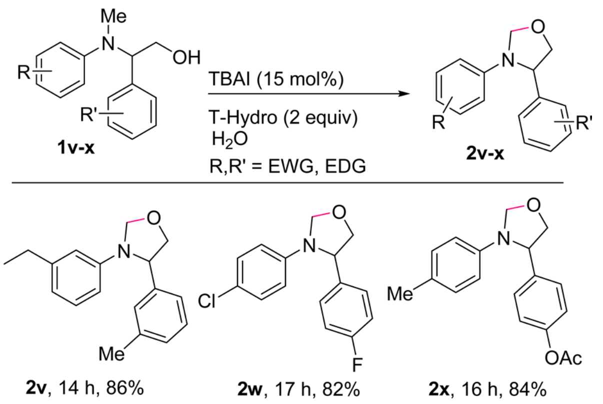
Scheme 2. Reaction of 2-Aryl Substituted Substrates[a,b]



[a] Substrate **1n−u** (0.5 mmol), TBAI (15 mol %), T-Hydro (1 mmol), H_2O (1 mL), 60 °C. [b] Isolated yield.

The reaction of the substrates bearing substitution in both aryl rings was further examined (Scheme 3). Substrate **1v** having substitution at the 3-position with methyl and ethyl groups underwent reaction to give **2v** in 86% yield. The reaction of **1w−x** having substitution at the 4-position with chloro, fluoro, methyl, and acetoxy functionalities afforded **2w** and **2x** in 82% and 84% yields, respectively. The reaction

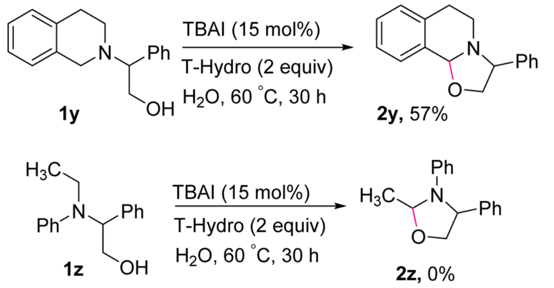
Scheme 3. Reaction of *N*- and 2-Aryl Substituted Substrates[a,b]



[a]Substrate **1v−x** (0.5 mmol), TBAI (15 mol %), T-Hydro (1 mmol), H_2O (1 mL), 60 °C. [b]Isolated yield.

condition is also compatible for the cyclization of tetrahydroisoquinoline derivative **1y**, affording the tricyclic oxazolidine **2y** in good yield (Scheme 4). Under these conditions, *N*-ethyl substrate **1z** showed no cyclization and the formation of **2z** was not observed.

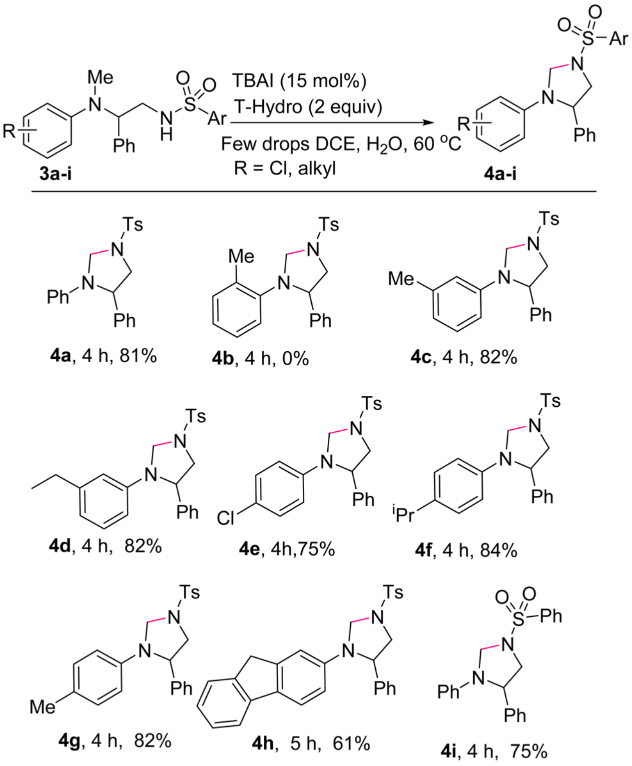
Scheme 4. Reaction of Tetrahydroisoquinoline and *N*-Ethyl Substrates



Having these results, we further studied the reaction of analogue diamines to produce imidazolidines (Scheme 5). The best results were observed in the presence of a few drops (200 *μ*L) of 1,2-dichloroethane (DCE), which assists the solid substrate to be gummy and float on the surface of water. The substrate **3a** underwent reaction to give imidazolidine **4a** in 81% yield. Next, the reaction of the substrates bearing substitution in the *N*-aryl ring was performed. As above, the reaction of **3b** containing a 2-methyl group showed no cyclization to furnish **4b**. However, the substrates **3c−d** having substitution at the 3-position with methyl and ethyl functionalities afforded imidazolidines **4c−d** in 82% yield. Similarly, the substrates **3e−g** bearing substitution at the 4-position with chloro, isopropyl, and methyl groups underwent cyclization to afford the corresponding imidazolidines **4e−g** in 75−84% yields, whereas **3h** with *N*-fluorene furnished **4h** in 61% yield. In addition, the substrate **3i** having an *N*-phenylsulfonyl substituent underwent reaction to furnish imidazolidine **4i** in 75% yield.

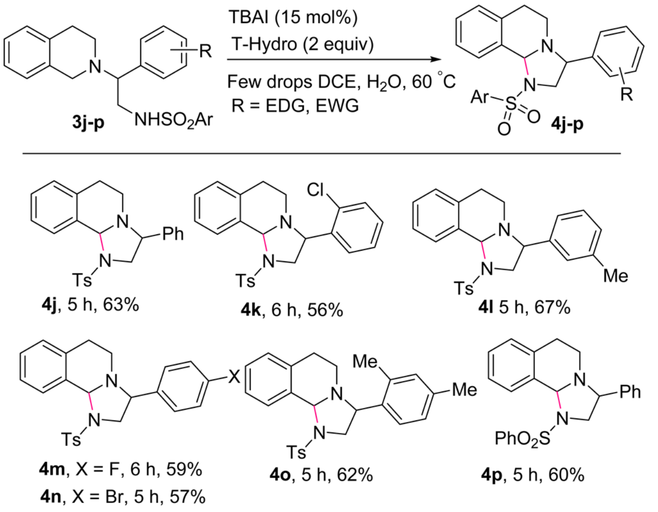
Next, the reaction of tetrahydroisoquinoline derivatives was investigated (Scheme 6). The substrate **3j** underwent reaction to provide imidazolidine **4j** in 63% yield. Similarly, the reaction of **3k** and **3l** bearing 2-chloro and 3-methyl groups in the 2-aryl ring afforded imidazolidines **4k** and **4l** in 56% and 67% yields, respectively, whereas **3m−n** bearing at the 4-position with bromo and fluoro groups produced imidazolidines **4m−n** in 57−59% yields. In addition, the substrate **3o** with 2,4-dimethyl groups underwent reaction to furnish **4o** in 62% yield, while **3p** with an *N*-phenylsulfonyl instead of an *N*-tosyl functionality cyclized to afford **4p** in 60% yield.

Scheme 5. Reaction of *N*-Aryl Substituted Substrates[a,b]



[a]Substrate **3a−i** (0.5 mmol), TBAI (15 mol %), T-Hydro (1 mmol), DCE (few drops), H_2O (1 mL), 60 °C. [b]Isolated yield.

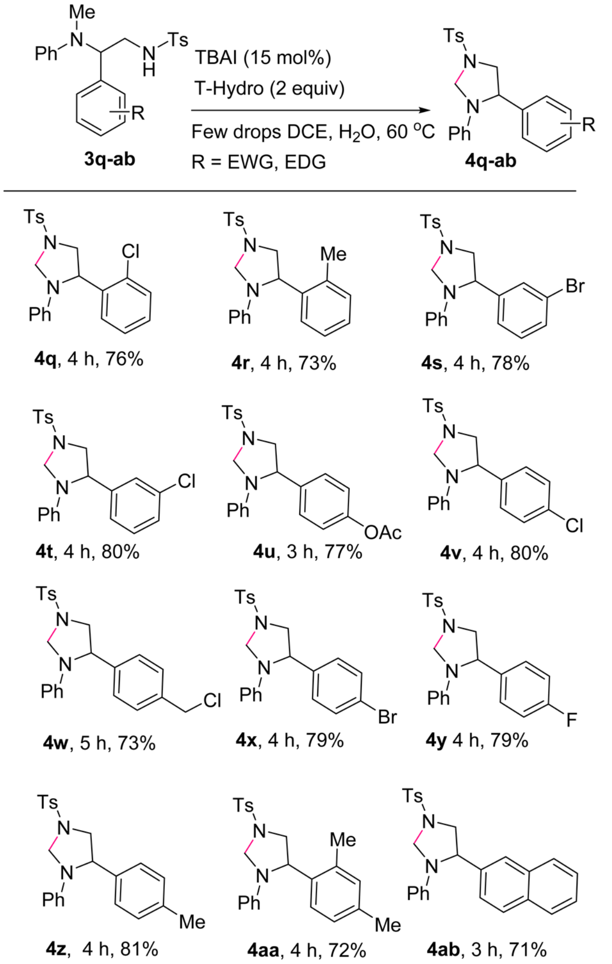
Scheme 6. Reaction of Tetrahydroisoquinoline Derivatives[a,b]



[a]Substrate **3j−p** (0.5 mmol), TBAI (15 mol %), T-Hydro (1 mmol), DCE (few drops), water (1 mL), 60 °C. [b]Isolated yield.

Finally, the reaction of the substrates with substitution in the 2-aryl ring was pursued (Scheme 7). The substrates **3q−r** bearing substitution at the 2-position with chloro and methyl

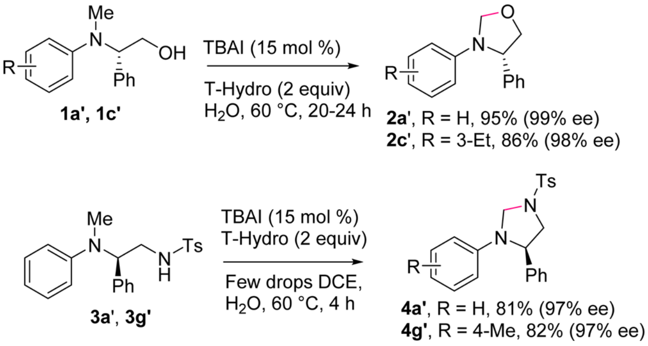
Scheme 7. Reaction of 2-Aryl Substituted Substrates[a,b]



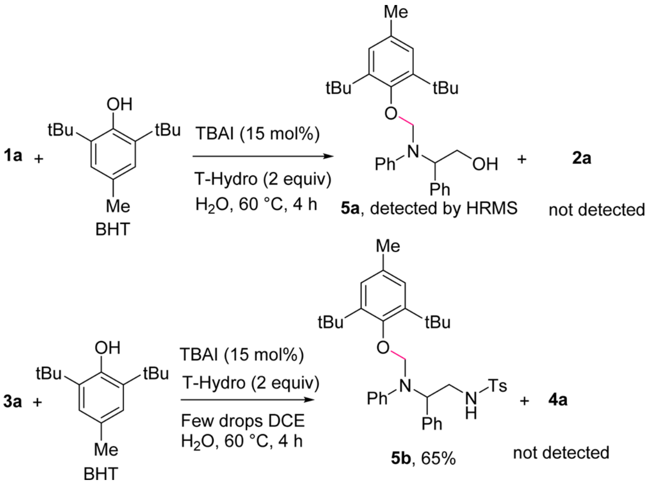
[a]Substrate **3q−ab** (0.5 mmol), TBAI (15 mol %), T-Hydro (1 mmol), DCE (few drops), water (1 mL), 60 °C. [b]Isolated yield.

groups produced imidazolidines **4q** and **4r** in 76% and 73% yields, respectively. The reaction of **3s−t** having substitution at the 3-position with bromo and chloro functionalities furnished imidazolidines **4s−t** in 78−80% yields. Similarly, the substrates **3u−z** containing substitution at the 4-position with acetoxy, chloro, bromo, fluoro, methyl, and chlormethyl groups underwent reaction to afford the corresponding imidazolidines **4u−z** in 73−81% yields. In addition, **3aa** with 2,4-dimethyl groups cross-coupled to give **4aa** in 72% yield, whereas **3ab** having a naphthyl functionality furnished imidazolidine **4ab** in 71% yield.

Scheme 8. Reaction of Optically Active Substrates



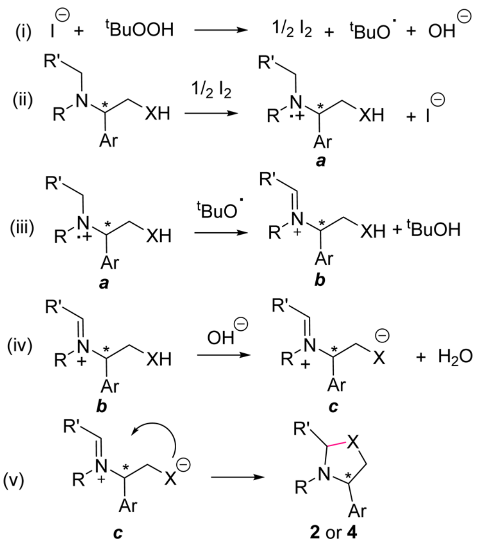
Scheme 9. Radical Scavenger Experiments



In the case of optically active substrates, the reaction proceeded with high optical purities (Scheme 8). The reaction of **1a**′, **1c**′, **3a**′, and **3g**′ was performed as the representative examples. The substrates **1a**′ and **1c**′ underwent reaction to produce **2a**′ and **2c**′ in 99% and 98% ee, respectively, while the reaction of **3a**′ and **3g**′ produced the target heterocycles **4a**′ and **4g**′ in 97% ee. These results suggest that the reaction provides a potential route to construct oxazolidines and imidazolidines with high optical purities.

To get insight into the catalytic pathway, the reaction of **1a** and **3a** was performed with BHT (Scheme 9).[15] HRMS analysis of the reaction mixtures revealed the formation of BHT adducts **5a** and **5b** as the sole products, which suggests that the reaction involves a radical intermediate (see the Supporting Information). Thus, the oxidation of TBAI by T-Hydro may give iodine, *tert*-butoxyl radical, and hydroxyl ion (Scheme 10, step (i)). Single electron transfer (SET) reduction of iodine may regenerate the catalyst with the formation of the radical cation ***a*** (Scheme 10, step (ii)).[16] Homolysis of the methyl C−H bond induced by *tert*-butoxyl radical may give the iminium ***b***, which may convert into the target heterocycles **2** and **4** via the

Scheme 10. Proposed Reaction Pathway



R = aryl; R′ = H, benzyl; X = O, N-SO$_2$Ar intermediate ***c*** (Scheme 10, steps (iv) and (v)). The tertiary benzylic C−H bond is intact, which may be due to its steric hindrance toward the *tert*-butoxyl radical compared to that of the methyl C−H bond. In these reactions, TBAI and TBHP are dissolved in water, while the substrates **1** and **3** are floated on the surface of the water, and the mixture is stirred. After completion, the products **2** and **4** are separated out as a colorless solid or liquid on the surface of water, which can be easily isolated. The reaction may take place in the interface of oil−water droplets.[1,2]

■ CONCLUSION

We described the oxidative cross-coupling of the *N*-alkyl C−H bond with alkyl O−H and N−H bonds for the construction of the functionalized oxazolidines and imidazolidines using TBAI in the presence of T-Hydro at moderate temperature. The use of water as the solvent, metal-free condition, regioselectivity, simplified product isolation, and broad substrate scope are the salient features. Optically active substrates can be converted enantiospecifically to the corresponding oxazolidines and imidazolidines.

■ EXPERIMENTAL SECTION

General Information. T-Hydro, DTBP (98%), I_2, 30% aq. H_2O_2, BHT (>99%), TBAI (99%), NaI (99%), and KI (99%) were purchased from a commercial source and used as received. All reactions were performed in pure water (>5 MΩ cm @ 25 °C, total organic content < 30 ppb). The reactions were monitored by analytical TLC on silica gel G/GF 254 plates. The column chromatography was performed with 60−120 mesh silica gel. NMR (^{1}H and ^{13}C) spectra were recorded on 600 and 400 MHz spectrometers using $CDCl_3$ as a solvent and TMS as an internal standard. The data are accounted as follows: chemical shifts (δ ppm) (multiplicity, coupling constant (Hz), integration). The abbreviations for multiplicity are as follows: s = singlet, d = doublet, t = triplet, q = quartet, m = multiplet, and dd = doublet of doublets. Melting points were determined with a melting point apparatus and are uncorrected. Optical rotation was determined using a polarimeter with a 50 mm path length cell at 589 nm at 23 °C. FT-IR spectra were recorded on an IR spectrometer. HRMS were analyzed with a Q-TOF instrument. HPLC analysis was carried out with Daicel Chiralcel OD and OJ columns.

General Procedure for the Synthesis of Oxazolidines 2a−y. To a mixture of the substituted 1,2-aminoalcohols **1a−y** (0.5 mmol), TBAI (15 mol %), and water (1 mL) was added T-Hydro (1.0 mmol) at room temperature under air. The substrate **1a−y** was floated on the surface of the water as an oil, and the resultant mixture was stirred at 60 °C for the appropriate time. The progress of the reaction was monitored by TLC using hexane and ethyl acetate as an eluent. After completion, the reaction mixture was cooled to room temperature and treated with saturated $Na_2S_2O_3$ (500 μL) and ethyl acetate (5 mL). The organic layer was separated, dried (Na_2SO_4), and evaporated to give a residue that was purified on a silica gel column chromatography using hexane and ethyl acetate as an eluent.

General Procedure for the Synthesis of Imidazolidines 4a−ab. To a mixture of substituted 1,2-diamines **3a−ab** (0.5 mml) and 1,2-dichloroethane (three drops, ∼200 μL) were added water (1 mL), TBAI (15 mol %), and T-Hydro (1.0 mmol) at room temperature under air. The substrate **3a−ab** with DCE was floated on the surface of the water, and the resultant mixture was stirred at 60 °C for the appropriate time. Monitoring of the reaction, workup, and purification were performed as described for **2a−y**.

3,4-Diphenyloxazolidine ***2a′***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.61; pale yellow liquid; yield 95% (107 mg); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.37−7.34 (m, 4H), 7.30−7.28 (m, 1H), 7.19 (t, J = 8.4 Hz, 2H), 6.75 (t, J = 7.2 Hz, 1H), 6.50 (d, J = 8.4 Hz, 2H) 5.33 (d, J = 1.8 Hz, 1H), 5.04 (d, J = 1.8 Hz, 1H), 4.71 (dd, J = 6.6, 4.2 Hz, 1H), 4.41 (t, J = 7.8 Hz, 1H), 3.99 (dd, J = 8.4, 4.2 Hz, 1H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 145.2, 141.6, 129.4, 129.0, 127.7, 126.4, 117.9, 112.9, 83.0, 75.9, 61.9; FT-IR (neat) 3061, 3028, 2928, 2864, 1600, 1508, 1495, 1346, 1166, 1090, 749, 692 cm^{-1}; HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{15}H_{15}NO$ 226.1226, found 226.1225; $[\alpha]_D^{20}$ = +159.0 (c = 0.2, $CHCl_3$); HPLC analysis: 99% ee [Daicel Chiralcel OD column, hexane/iPrOH = 90:10, flow rate: 1 mL/min, λ = 254 nm, t_R = 4.94 min (minor), 8.36 min (major)].

3-(3-Ethylphenyl)-4-phenyloxazolidine ***2c′***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.62; pale yellow liquid; yield 86% (109 mg); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.37−7.32 (m, 4H), 7.29−7.27 (m, 1H), 7.09 (t, J = 7.6 Hz, 1H), 6.62 (d, J = 7.6 Hz, 1H), 6.33−6.30 (m, 2H), 5.33 (d, J = 2 Hz, 1H), 5.03 (d, J = 2 Hz, 1H), 4.70 (dd, J = 6.8, 4.4 Hz, 1H), 3.39 (t, J = 7.6 Hz, 1H), 3.98 (dd, J = 8.4, 4.4 Hz, 1H), 2.56 (q, J = 7.6 Hz, 2H), 1.17 (t, J = 7.6 Hz, 3H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 145.6, 145.3, 141.8, 129.3, 129.0, 127.7, 126.4, 117.6, 112.5, 110.5, 83.0, 75.9, 61.9, 29.3, 15.7; FT-IR (neat) 3030, 2963, 2929, 2869, 1603, 1493, 1453, 1356, 1166, 1090, 759, 697 cm^{-1}; HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{17}H_{19}NO$ 254.1539, found 254.1539; $[\alpha]_D^{20}$ = +28.0 (c = 0.2, $CHCl_3$); HPLC analysis: 98% ee [Daicel Chiralcel OJ column, hexane/iPrOH = 85:15, flow rate: 1 mL/min, λ = 254 nm, t_R = 6.31 min (minor), 13.53 min (major)].

4-Phenyl-3-(m-tolyl)oxazolidine ***2d***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.62; pale yellow liquid; yield 84% (100 mg); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.36−7.33 (m, 4H), 7.29−7.27 (m, 1H), 7.06 (t, J = 7.8 Hz, 1H), 6.58 (d, J = 7.2 Hz, 1H), 6.32 (s, 1H), 6.30 (d, J = 8.4 Hz, 1H), 5.32 (d, J = 2.4 Hz, 1H), 5.02 (d, J = 1.8 Hz, 1H), 4.70 (dd, J = 6.6, 4.2 Hz, 1H), 4.38 (t, J = 8.4 Hz, 1H), 3.98 (dd, J = 7.8, 4.2 Hz, 1H), 2.26 (s, 3H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 145.3, 141.8, 139.2, 129.3, 129.0, 127.7, 126.4, 118.8, 113.6, 110.2, 83.0, 75.9, 61.8, 22.0; FT-IR (neat) 3063, 3032, 2921, 2863, 1605, 1493, 1453, 1356, 1170, 1089, 945, 840 cm^{-1}; HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{16}H_{17}NO$ 240.1383, found 240.1383.

4-Phenyl-3-(3-(trifluoromethyl)phenyl)oxazolidine ***2e***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.60; colorless liquid; yield 55% (81 mg); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.37−7.32 (m, 4H), 7.31−7.28 (m, 1H), 7.23 (t, J = 8.4 Hz, 1H), 6.97 (d, J = 7.8 Hz, 1H), 6.66 (s, 1H), 6.59 (dd, J = 8.4, 2.4 Hz, 1H), 5.33 (d, J = 2.4 Hz, 1H), 5.05 (d, J = 2.4 Hz, 1H), 4.72 (dd, J = 6.6, 4.2 Hz, 1H), 4.43 (dd, J = 8.4, 7.2 Hz, 1H), 4.00 (dd, J = 8.4, 4.2 Hz, 1H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 145.1, 140.7, 131.8 (q, $J_{C\text{-}F}$ = 31.5 Hz), 129.8, 128.1, 126.3, 124.7 (q, $J_{C\text{-}F}$ = 93.0 Hz), 115.9, 114.3 (q, $J_{C\text{-}F}$ = 4.5 Hz), 109.1 (q, $J_{C\text{-}F}$ = 3.0 Hz), 82.7, 76.0, 61.8; FT-IR (neat) 3061, 3031, 2923, 2868, 1615, 1508, 1493, 1459, 1372, 1166, 1121, 782, 697 cm^{-1}; HRMS (APCI) m/z [M + H]$^+$ calcd for $C_{16}H_{14}F_3NO$ 294.1100, found 294.1097.

3-(4-Bromophenyl)-4-phenyloxazolidine ***2f***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.59; pale yellow liquid; yield 77% (117 mg); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.36−7.27 (m, 5H), 7.24 (d, J = 8.4 Hz, 2H), 6.34 (d, J = 8.4 Hz, 2H), 5.27 (d, J = 2.4 Hz, 1H), 4.98 (d, J = 2.4 Hz, 1H), 4.66 (dd, J = 7.2, 4.8 Hz, 1H), 4.41 (t, J = 6.0 Hz, 1H), 3.97 (dd, J = 8.4, 4.8 Hz, 1H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 144.0, 140.9, 132.1, 129.1, 128.0, 126.3, 144.5, 109.9, 82.8, 76.0, 61.9; FT-IR (neat) 3061, 3028, 2923, 2853, 1595, 1505, 1489, 1360, 1165, 1089, 944, 807, 757 cm^{-1}; HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{15}H_{14}BrNO$ 304.0332, found 304.0328.

3-(4-Chlorophenyl)-4-phenyloxazolidine ***2g***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.59; pale yellow liquid; yield 81% (105 mg); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.36−7.27 (m, 5H), 7.11 (d, J = 9.0 Hz, 2H), 6.38 (d, J = 9.0 Hz, 2H), 5.28 (d, J = 2.4 Hz, 1H), 4.99 (d, J = 2.4 Hz, 1H), 4.66 (dd, J = 6.6, 4.2 Hz, 1H), 4.41 (dd, J = 8.4, 6.6 Hz, 1H), 3.96 (dd, J = 8.4, 4.2 Hz, 1H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 143.7, 141.0, 129.3, 129.1, 128.0, 126.3, 122.8, 114.0, 82.9, 76.0, 62.0; FT-IR (neat) 3063, 3028, 2924, 2854, 1601, 1504, 1493, 1358, 1166, 1095, 810 cm^{-1}; HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{15}H_{14}ClNO$ 260.0837, found 260.0836.

3-(4-Isopropylphenyl)-4-phenyloxazolidine ***2h***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.63; pale yellow liquid; yield 88% (117 mg); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.37−7.32 (m, 4H), 7.28 (d, J = 6.6, 1H), 7.05 (d, J = 7.8 Hz, 2H), 6.44 (d, J = 7.8 Hz, 2H), 5.31 (d, J = 2.4 Hz, 1H), 4.99 (d, J = 1.8 Hz, 1H), 4.66 (dd, J =

7.2, 4.8 Hz, 1H), 4.37 (t, J = 7.2 Hz, 1H), 3.95 (dd, J = 8.4, 4.8 Hz, 1H), 2.81−2.76 (m, 1H), 1.18 (d, J = 7.2 Hz, 6H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 143.6, 142.0, 138.4, 129.0, 127.7, 127.3, 126.5, 113.0, 83.3, 75.9, 62.3, 33.3, 24.4; FT-IR (neat) 3064, 3028, 2959, 2926, 2867, 1618, 1520, 1346, 1157, 1090, 816, 757, 704 cm^{-1}; HRMS (APCI) m/z $[M + H]^+$ calcd for $C_{18}H_{21}NO$ 268.1696, found 268.1697.

4-Phenyl-3-(p-tolyl)oxazolidine ***2i***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.62; colorless liquid; yield 84% (100 mg); ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.37−7.33 (m, 4H), 7.29−7.26 (m, 1H), 7.00 (d, J = 8.4 Hz, 2H), 6.42 (d, J = 8.4 Hz, 2H), 5.32 (d, J = 2.4 Hz, 1H), 5.00 (d, J = 2.4 Hz, 1H), 4.66 (dd, J = 6.6, 4.8 Hz, 1H), 4.40 (t, J = 8.4 Hz, 1H), 3.96 (dd, J = 8.4, 4.8 Hz, 1H), 2.23 (s, 3H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 143.2, 141.7, 129.9, 129.0, 127.7, 127.1, 126.4, 113.1, 83.3, 75.9, 62.1, 20.5; FT-IR (neat) 3063, 3027, 2918, 2861, 1621, 1522, 1452, 1340, 1170, 1090, 804 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{16}H_{17}NO$ 240.1383, found 240.1385.

3-(3,4-Dimethylphenyl)-4-phenyloxazolidine ***2k***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.63; brown liquid; yield 85% (108 mg); ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.36−7.32 (m, 4H), 7.27−7.25 (m, 1H), 6.92 (d, J = 7.8 Hz, 1H), 6.31 (d, J = 1.8 Hz, 1H), 6.24 (dd, J = 7.8, 2.4 Hz, 1H), 5.30 (d, J = 1.8 Hz, 1H), 4.98 (d, J = 2.4 Hz, 1H), 4.65 (dd, J = 6.6, 4.8 Hz, 1H), 4.37 (t, J = 7.8 Hz, 1H), 3.94 (dd, J = 8.4, 4.2 Hz, 1H), 2.16 (s, 3H), 2.13 (s, 3H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 143.7, 141.9, 137.6, 130.5, 129.0, 127.6, 126.4, 126.0, 114.6, 110.6, 83.4, 75.9, 62.0, 20.4, 18.9; FT-IR (neat) 3060, 3025, 2921, 2859, 1617, 1513, 1452, 1354, 1274, 1089, 800, 717, 700 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{17}H_{19}NO$ 254.1539, found 254.1539.

3-(3,5-Dichlorophenyl)-4-phenyloxazolidine ***2l***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.59; pale yellow liquid; yield 70% (103 mg); ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.36 (t, J = 7.2 Hz, 2H), 7.32−7.28 (m, 3H), 6.70 (t, J = 1.8 Hz, 1H), 6.31 (d, J = 1.2 Hz, 2H), 5.24 (d, J = 2.4 Hz, 1H), 4.97 (d, J = 2.4 Hz, 1H), 4.66 (dd, J = 6.6 Hz, 4.2 Hz, 1H), 4.39 (dd, J = 8.4, 6.6 Hz, 1H), 3.98 (dd, J = 8.4, 3.6 Hz, 1H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 146.3, 140.3, 135.7, 129.3, 128.2, 126.2, 117.6, 111.1, 82.4, 75.9, 61.6; FT-IR (neat) 3084, 3029, 2923, 2853, 1592, 1557, 1463, 1344, 1167, 1093, 820, 708 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{15}H_{13}Cl_2NO$ 294.0447, found 294.0448.

(E)-4-Phenyl-3-(4-(phenyldiazenyl)phenyl)oxazolidine ***2m***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.56; orange liquid; yield 47% (77 mg); ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.82 (d, J = 9.0 Hz, 4H), 7.46 (t, J = 7.2 Hz, 2H), 7.39−7.29 (m, 6H), 6.54 (d, J = 9.0 Hz, 2H), 5.38 (d, J = 2.4 Hz, 1H), 5.14 (d, J = 2.4 Hz, 1H), 4.84 (dd, J = 6.6, 4.2 Hz, 1H), 4.45 (dd, J = 8.4, 6.6 Hz, 1H), 4.04 (dd, J = 8.4, 4.2 Hz, 1H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 153.3, 147.0, 145.0, 140.8, 129.9, 129.2, 129.1, 128.1, 126.4, 125.2, 122.5, 112.7, 82.4, 76.0, 61.7; FT-IR (neat) 3061, 3025, 2959, 2922, 2851, 1602, 1515, 1497, 1389, 1373, 1140, 1088, 821, 766 cm^{-1}; HRMS (APCI) m/z $[M + H]^+$ calcd for $C_{21}H_{19}N_3O$ 330.1601, found 330.1601.

4-(2-Chlorophenyl)-3-phenyloxazolidine ***2n***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.58; pale yellow liquid; yield 75% (97 mg); ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.43 (d, J = 7.2 Hz, 1H), 7.34 (d, J = 6.0 Hz, 1H), 7.25−7.20 (m, 4H), 6.78 (t, J = 7.2 Hz, 1H), 6.44 (d, J = 7.8 Hz, 2H), 5.35 (s, 1H), 5.11 (d, J = 4.2 Hz, 1H), 5.01 (s, 1H), 4.47 (t, J = 7.8 Hz, 1H), 4.06 (dd, J = 8.4 Hz, 2.4 Hz, 1H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 144.7, 138.6, 132.4, 129.8, 129.6, 128.9, 127.9, 127.4, 118.1, 112.9, 82.6, 74.5, 59.2; FT-IR (neat) 3064, 3036, 2924, 2857, 1600, 1508, 1497, 1348, 1171, 1092, 744, 691 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{15}H_{14}ClNO$ 260.0837, found 260.0836.

4-(3-Methoxyphenyl)-3-phenyloxazolidine ***2o***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.41; colorless liquid; yield 87% (111 mg); ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.27 (t, J = 7.8 Hz, 1H), 7.18 (t, J = 8.4 Hz, 2H), 6.96 (d, J = 7.8 Hz, 1H), 6.91 (s, 1H), 6.83 (dd, J = 8.4, 2.4 Hz, 1H), 6.75 (t, J = 7.2 Hz, 1H), 6.50 (d, J = 8.4 Hz, 2H), 5.32 (d, J = 2.4 Hz, 1H), 5.02 (d, J = 2.4 Hz, 1H), 4.66 (dd, J = 6.6, 4.2 Hz, 1H), 4.39 (t, J = 8.4 Hz, 1H), 3.98 (dd, J = 8.4, 4.2 Hz, 1H), 3.79 (s, 3H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 160.3, 145.3, 143.5, 130.1, 129.4, 118.7, 117.9, 113.0, 112.9, 112.1, 83.0, 75.8, 61.9, 55.4; FT-IR (neat) 3040, 2997, 2935, 2866, 2835, 1600, 1508, 1495, 1346, 1284, 1146, 1091, 749, 692 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{16}H_{17}NO_2$ 256.1332, found 256.1337.

3-Phenyl-4-(m-tolyl)oxazolidine ***2p***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.62; pale yellow liquid; yield 81% (97 mg); ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.24 (t, J = 7.2 Hz, 1H), 7.20−7.15 (m, 4H), 7.10 (d, J = 7.2 Hz, 1H), 6.75 (t, J = 7.2 Hz, 1H), 6.50 (d, J = 7.8 Hz, 2H), 5.33 (d, J = 2.4 Hz, 1H), 5.02 (d, J = 2.4 Hz, 1H), 4.66 (dd, J = 6.6, 4.2 Hz, 1H), 4.39 (dd, J = 8.4, 7.2 Hz, 1H), 3.97 (dd, J = 8.4, 4.2 Hz, 1H), 2.35 (s, 3H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 145.3, 141.7, 138.7, 129.4, 128.9, 128.5, 127.0, 123.5, 117.8, 112.9, 83.0, 76.0, 62.0, 21.7; FT-IR (neat) 3051, 3025, 2922, 2853, 1600, 1506, 1384, 1343, 1277, 1087, 1033, 788 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{16}H_{17}NO$ 240.1383, found 240.1386.

4-(3-Nitrophenyl)-3-phenyloxazolidine ***2q***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.53; yellow liquid; yield 65% (88 mg); ^{1}H NMR (600 MHz, CDCl$_3$) δ 8.23 (t, J = 1.8 Hz, 1H), 8.16 (dd, J = 8.4, 1.2 Hz, 1H), 7.72 (d, J = 7.8 Hz, 1H), 7.53 (t, J = 7.8 Hz, 1H), 7.21 (dd, J = 9.0, 7.2 Hz, 2H), 6.79 (t, J = 7.2 Hz, 1H), 6.46 (d, J = 7.8 Hz, 2H), 5.38 (d, J = 2.4 Hz, 1H), 5.01 (d, J = 2.4 Hz, 1H), 4.79 (dd, J = 6.6, 3.6 Hz, 1H), 4.42 (dd, J = 8.4, 6.6 Hz, 1H), 4.00 (dd, J = 8.4, 3.6 Hz, 1H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 148.9, 144.6, 144.4, 132.6, 130.1, 129.7, 123.0, 121.5, 118.6, 113.0, 83.1, 75.5, 61.4; FT-IR (neat) 3031, 2964, 2928, 2867, 1603, 1494, 1454, 1355, 1172, 1091, 948, 848, 782 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{15}H_{14}N_2O_3$ 271.1077, found: 271.1077.

4-(3-Phenyloxazolidin-4-yl)phenyl Acetate ***2r***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.50; pale yellow liquid; yield 85% (120 mg); ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.37 (d, J = 8.4 Hz, 2H), 7.18 (t, J = 7.2 Hz, 2H), 7.07 (d, J = 8.4 Hz, 2H), 6.75 (t, J = 7.2 Hz, 1H), 6.47 (d, J = 9.0 Hz, 2H), 5.31 (d, J = 2.4 Hz, 1H), 5.00 (d, J = 2.4 Hz, 1H), 4.69 (dd, J = 7.2, 4.2 Hz, 1H), 4.37 (dd, J = 8.4, 6.6 Hz, 1H), 3.97 (dd, J = 8.4, 4.2 Hz, 1H), 2.29 (s, 3H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 169.7, 150.2, 145.1, 139.2, 129.5, 127.4, 122.1, 118.1, 113.0, 83.0, 75.8, 61.4, 21.4; FT-IR (neat) 3065, 3033, 2956, 2854, 1764, 1600, 1507, 1368, 1215, 1196, 1089, 749, 692 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{17}H_{17}NO_3$ 284.1281, found 284.1287.

4-(4-Bromophenyl)-3-phenyloxazolidine ***2s***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.58; pale yellow liquid; yield 79% (120 mg); ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.48 (d, J = 7.8 Hz, 2H), 7.25 (d, J = 8.4 Hz, 2H), 7.19 (t, J = 7.8 Hz, 2H), 6.77 (t, J = 7.2 Hz, 1H), 6.46 (d, J = 7.8 Hz, 2H), 5.31 (s, 1H), 5.00 (s, 1H), 4.65 (t, J = 6.0 Hz, 1H), 4.38 (t, J = 7.8 Hz, 1H), 3.95 (dd, J = 8.4 Hz, 4.2 Hz, 1H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 144.9, 140.8, 132.1, 129.5, 128.1, 121.5, 118.1, 112.9, 82.9, 75.7, 61.3; FT-IR (neat) 3041, 2986, 2866, 2828, 1599, 1507, 1487, 1345, 1170, 1010, 823, 749, 692 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{15}H_{14}BrNO$ 304.0332, found 304.0331.

4-(4-Chlorophenyl)-3-phenyloxazolidine ***2t***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.58; pale yellow liquid; yield 78% (101 mg); ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.33−7.29 (m, 4H), 7.18 (t, J = 7.8 Hz, 2H), 6.76 (t, J = 7.2, Hz 1H), 6.46 (d, J = 8.4 Hz, 2H), 5.31 (s, 1H), 5.00 (s, 1H), 4.66 (t, J = 5.4 Hz, 1H), 4.38 (t, J = 7.8 Hz, 1H), 3.94 (dd, J = 8.4 Hz, 3.6 Hz, 1H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 144.9, 140.3, 133.5, 129.5, 129.2, 127.8, 118.1, 112.9, 82.9, 75.7, 61.3; FT-IR (neat) 3063, 3040, 2924, 2854, 1600, 1508, 1495, 1346, 1167, 1090, 828, 747, 691 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{15}H_{14}ClNO$ 260.0837, found 260.0840.

4-(4-Fluorophenyl)-3-phenyloxazolidine ***2u***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.57; pale yellow liquid; yield 84% (102 mg); ^{1}H NMR (600 MHz, CDCl$_3$) 7.33 (t, J = 7.2 Hz, 2H), 7.20 (t, J = 7.2 Hz, 2H), 7.04 (t, J = 8.4 Hz, 2H), 6.77 (t, J = 7.2 Hz, 1H), 6.48 (d, J = 7.8 Hz, 2H), 5.33 (s, 1H), 5.01 (s, 1H), 4.68 (d, J = 3.6 Hz, 1H), 4.38 (t, J = 7.8 Hz, 1H), 3.96 (dd, J = 7.2, 3.0 Hz, 1H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 163.2 (d, $J_{C\text{-}F}$ = 244.5 Hz), 145.0, 137.4, 129.5, 128.0 (d, $J_{C\text{-}F}$ = 7.5 Hz), 118.0, 115.9 (d, $J_{C\text{-}F}$ = 21.0 Hz), 112.9, 82.9, 75.9, 61.2; FT-IR (neat) 3064, 3040, 2929, 2865, 1600, 1508, 1497, 1351, 1222, 1154, 1090, 836, 749, 692 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{15}H_{14}FNO$ 244.1132, found 244.1132.

3-(3-Ethylphenyl)-4-(m-tolyl)oxazolidine ***2v***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.63; colorless liquid; yield 86% (115 mg); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.24 (t, *J* = 7.2 Hz, 1H), 7.19−7.16 (m, 2H), 7.11−7.09 (m, 2H), 6.62 (d, *J* = 7.2 Hz, 1H), 6.35−6.32 (m, 2H), 5.33 (s, 1H), 5.02 (s, 1H), 4.66 (t, *J* = 5.4 Hz, 1H), 4.37 (t, *J* = 7.8 Hz, 1H), 3.98 (dd, *J* = 7.8, 3.6 Hz, 1H), 2.57 (q, *J* = 7.2 Hz, 2H), 2.35 (s, 3H), 1.19 (t, *J* = 7.8 Hz, 3H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 145.6, 145.5, 141.9, 138.6, 129.3, 128.8, 128.5, 127.0, 123.5, 117.5, 112.5, 110.5, 83.0, 75.9, 62.0, 29.3, 21.7, 15.7; FT-IR (neat) 3030, 2963, 2928, 2868, 1604, 1494, 1454, 1355, 1172, 1092, 948, 783, 695 cm^{-1}; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{18}H_{21}NO$ 268.1696, found 268.1709.

3-(4-Chlorophenyl)-4-(4-fluorophenyl)oxazolidine ***2w***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.57; colorless liquid; yield 82% (114 mg); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.30 (dd, *J* = 8.4, 5.4 Hz, 2H), 7.12 (d, *J* = 8.4 Hz, 2H), 7.03 (t, *J* = 8.4 Hz 2H), 6.36 (d, *J* = 9.0 Hz, 2H), 5.28 (d, *J* = 2.4 Hz, 1H), 4.96 (d, *J* = 1.8 Hz, 1H), 4.63 (dd, *J* = 6.6, 4.2 Hz, 1H), 4.39 (dd, *J* = 8.4, 7.2 Hz, 1H), 3.93 (dd, *J* = 8.4, 4.2 Hz, 1H); ^{13}C NMR (150 MHz, $CDCl_3$) 163.3 (d, $J_{C\text{-}F}$ = 244.5 Hz), 143.5, 136.8, 129.3, 127.9 (d, $J_{C\text{-}F}$ = 7.5 Hz), 123.0, 116.1 (d, $J_{C\text{-}F}$ = 22.5 Hz), 114.0, 82.9, 75.9, 61.3; FT-IR (neat) 3045, 2987, 2926, 2867, 1604, 1509, 1493, 1357, 1168, 1095, 809, 767 cm^{-1}; HRMS (APCI) *m/z* $[M + H]^+$ calcd for $C_{15}H_{13}ClFNO$ 278.0742, found 278.0738.

4-(3-(p-Tolyl)oxazolidin-4-yl)phenyl Acetate ***2x***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.51; colorless liquid; yield 84% (125 mg); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.37 (d, *J* = 8.4 Hz, 2H), 7.06 (d, *J* = 8.4 Hz, 2H), 6.99 (d, *J* = 8.4 Hz, 2H), 6.40 (d, *J* = 8.4 Hz, 2H), 5.30 (d, *J* = 2.4 Hz, 1H), 4.97 (d, *J* = 1.8 Hz, 1H), 4.65 (dd, *J* = 7.2, 5.4 Hz, 1H), 4.37 (dd, *J* = 8.4, 7.2 Hz, 1H), 3.95 (dd, *J* = 8.4, 4.2 Hz, 1H), 2.29 (s, 3H), 2.23 (s, 3H) ^{13}C NMR (150 MHz, $CDCl_3$) δ 169.7, 150.1, 143.1, 139.3, 130.0, 127.4, 122.1, 113.1, 83.3, 75.8, 61.6, 21.4, 20.5; FT-IR (neat) 3032, 3007, 2983, 2921, 2862, 1764, 1619, 1523, 1505, 1368, 1215, 1197, 1162, 1089, 803 cm^{-1}; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{18}H_{19}NO_3$ 298.1438, found 298.1437.

3-Phenyl-2,3,6,10b-tetrahydro-5H-oxazolo[2,3-a]isoquinoline ***2y***. Analytical TLC on silica gel, 1:19 ethyl acetate/hexane R_f = 0.48; colorless liquid; yield 57% (72 mg); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.46 (d, *J* = 7.6 Hz, 2H), 7.42−7.40 (m, 1H), 7.37 (t, *J* = 7.6 Hz, 2H), 7.30−7.26 (m, 3H), 7.19−7.17 (m, 1H), 5.45 (s, 1H), 4.47 (t, *J* = 8.0 Hz, 1H), 4.32 (t, *J* = 10.2 Hz, 1H), 3.89 (dd, 8, 6.4, 1H), 3.08−2.98 (m, 3H), 2.85−2.80 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 141.9, 135.7, 132.9, 128.8, 128.7, 128.3, 128.2, 127.4, 126.8, 126.6, 90.3, 71.4, 68.8, 46.9, 29.4; FT-IR (neat) 3062, 3027, 2924, 2852, 1604, 1494, 1395, 1384, 1126, 1029, 937, 746, 700; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{17}H_{17}NO$ 252.1383, found 252.1384.

3,4-Diphenyl-1-tosylimidazolidine ***4a′***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.55; colorless solid; yield 81% (153 mg); mp 126−127 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.68 (d, *J* = 8.4 Hz, 2H), 7.26−7.22 (m, 5H), 7.13−7.10 (m, 4H), 6.72 (t, *J* = 7.2 Hz, 1H), 6.37 (d, *J* = 8.4 Hz, 2H), 5.03 (d, *J* = 6.0 Hz, 1H), 4.76 (d, *J* = 6.0 Hz, 1H), 4.53 (t, *J* = 5.4 Hz, 1H), 3.92 (dd, *J* = 10.2, 7.2 Hz, 1H), 3.46 (dd, *J* = 10.8, 5.4 Hz, 1H), 2.40 (s, 3H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 145.0, 144.5, 140.6, 132.9, 130.1, 129.3, 129.1, 128.0, 127.9, 126.1, 118.4, 113.2, 66.2, 61.7, 55.7, 21.8; FT-IR (KBr) 3059, 3025, 2953, 2923, 2850, 1599, 1506, 1384, 1348, 1163, 1090, 1029, 814, 749 cm^{-1}; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{22}H_{22}N_2O_2S$ 379.1475, found 379.1473; $[\alpha]_D^{20}$ = −26.0 (*c* = 0.2, $CHCl_3$); HPLC analysis: 97% ee [Daicel Chiralcel OD column, hexane/*i*PrOH = 85:15, flow rate: 1 mL/min, λ = 215 nm, t_R = 9.49 min (major), 17.30 min (minor)].

4-Phenyl-3-(m-tolyl)-1-tosylimidazolidine ***4c***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.56; colorless solid; yield 82% (161 mg); mp 133−134 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.68 (d, *J* = 8.4 Hz, 2H), 7.25−7.22 (m, 4H), 7.11 (d, *J* = 6.0 Hz, 2H), 6.98 (t, *J* = 7.8 Hz, 1H), 6.56 (d, *J* = 7.8 Hz, 1H), 6.22 (s, 1H), 6.16 (d, *J* = 7.8 Hz, 2H), 5.06 (d, *J* = 5.4 Hz, 1H), 4.74 (d, *J* = 6.0 Hz, 1H), 4.54 (dd, *J* = 7.2, 4.8 Hz, 1H), 3.89 (dd, *J* = 10.2, 7.8 Hz, 1H), 3.45 (dd, *J* = 10.2, 4.8 Hz, 1H), 2.40 (s, 3H), 2.22 (s, 3H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 145.0, 144.4, 140.7, 139.1, 133.0, 130.0, 129.1, 129.0, 128.0, 127.8, 126.1, 119.4, 113.9, 110.5, 66.3, 61.6, 55.7, 21.9, 21.8; FT-IR (KBr) 3064, 3025, 2961, 2921, 2850, 1604, 1493, 1384, 1350, 1163, 1091, 1030, 813, 764 cm^{-1}; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{23}H_{24}N_2O_2S$ 393.1631, found 393.1630.

3-(3-Ethylphenyl)-4-phenyl-1-tosylimidazolidine ***4d***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.56; pale yellow liquid; yield 82% (166 mg); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.69 (d, *J* = 8.4 Hz, 2H), 7.25−7.21 (m, 5H), 7.11 (d, *J* = 6.0 Hz, 2H), 7.01 (t, *J* = 7.8 Hz, 1H), 6.59 (d, *J* = 7.2 Hz, 1H), 6.23 (s, 1H), 6.18 (dd, *J* = 7.8, 1.8 Hz, 1H), 5.04 (d, *J* = 6.0 Hz, 1H), 4.75 (d, *J* = 5.4 Hz, 1H), 4.54 (dd, *J* = 7.2, 4.8 Hz, 1H), 3.91 (dd, *J* = 10.8, 7.8 Hz, 1H), 3.46 (dd, *J* = 10.2, 4.8 Hz, 1H), 2.52 (q, *J* = 7.8 Hz, 2H), 2.40 (s, 3H), 1.12 (t, *J* = 7.8 Hz, 3H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 145.5, 145.1, 144.4, 140.8, 133.0, 130.0, 129.2, 129.0, 128.0, 127.8, 126.1, 118.2, 112.9, 110.8, 66.3, 61.8, 55.7, 29.3, 21.8, 15.7; FT-IR (neat) 3059, 3025, 2925, 2848, 1599, 1504, 1354, 1163, 1091, 816, 746 cm^{-1}; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{24}H_{26}N_2O_2S$ 407.1788, found 407.1791.

3-(4-Chlorophenyl)-4-phenyl-1-tosylimidazolidine ***4e***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.54; pale yellow liquid; yield 75% (155 mg); ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.68 (d, *J* = 8.4 Hz, 2H), 7.25−7.23 (m, 5H), 7.09 (d, *J* = 7.2 Hz, 2H), 7.05 (d, *J* = 9.0 Hz, 2H), 6.27 (d, *J* = 9.0 Hz, 2H), 4.98 (d, *J* = 6.0 Hz, 1H), 4.76 (d, *J* = 6.6 Hz, 1H), 4.45 (t, *J* = 6.6 Hz, 1H), 3.97 (dd, *J* = 10.8, 7.8 Hz, 1H), 3.43 (dd, *J* = 10.8, 5.4 Hz, 1H), 2.40 (s, 3H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 144.6, 140.0, 133.1, 130.1, 129.2, 129.1, 128.1, 128.0, 126.0, 123.4, 114.3, 66.3, 61.8, 55.8, 21.8; FT-IR (neat) 3067, 3025, 2925, 2889, 2820, 1601, 1507, 1346, 1162, 1024, 785, 748 cm^{-1}; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{22}H_{21}ClN_2O_2S$ 413.1085, found 413.1079.

3-(4-Isopropylphenyl)-4-phenyl-1-tosylimidazolidine ***4f***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.56; colorless solid; yield 84% (176 mg); mp 140−141 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.71 (d, *J* = 7.8 Hz, 2H), 7.26−7.23 (m, 5H), 7.15 (d, *J* = 6.6 Hz, 2H), 7.01 (d, *J* = 8.4 Hz, 2H), 6.37 (d, *J* = 8.4 Hz, 2H), 5.07 (d, *J* = 6.0 Hz, 1H), 4.72 (d, *J* = 6.0 Hz, 1H), 4.53 (t, *J* = 6.6 Hz, 1H), 3.91 (t, *J* = 8.4 Hz, 1H), 3.46 (dd, *J* = 10.8, 5.4 Hz, 1H), 2.80−2.76 (m, 1H), 2.41 (s, 3H), 1.19 (d, *J* = 6.0 Hz, 6H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 144.4, 143.2, 140.8, 138.9, 132.9, 130.0, 129.0, 127.8, 127.1, 126.1, 113.3, 66.5, 62.0, 55.7, 33.2, 24.3, 21.7; FT-IR (KBr) 3064, 3028, 2957, 2928, 2867, 1615, 1519, 1454, 1351, 1164, 1095, 814, 700 cm^{-1}; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{25}H_{28}N_2O_2S$ 421.1944, found 421.1945.

4-Phenyl-3-(p-tolyl)-1-tosylimidazolidine ***4g′***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.55; colorless solid; yield 82% (161 mg); mp 141−142 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.69 (d, *J* = 8.4 Hz, 2H), 7.24−7.20 (m, 5H), 7.10 (d, *J* = 6.0 Hz, 2H), 6.93 (d, *J* = 8.4 Hz, 2H), 6.31 (d, *J* = 8.4 Hz, 2H), 5.03 (d, *J* = 6.0 Hz, 1H), 4.70 (d, *J* = 6.0 Hz, 1H), 4.51 (t, *J* = 6.6 Hz, 1H), 3.91 (dd, *J* = 10.2, 7.2 Hz, 1H), 3.42 (dd, *J* = 10.2, 5.4 Hz, 1H), 2.40 (s, 3H), 2.19 (s, 3H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 144.4, 142.9, 140.7, 133.0, 130.0, 129.8, 129.0, 128.0, 127.8, 126.1, 113.5, 66.6, 61.9, 55.7, 21.8, 20.5; FT-IR (KBr) 3059, 3028, 2923, 2860, 1619, 1521, 1384, 1350, 1163, 1094, 1030, 804, 760 cm^{-1}; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{23}H_{24}N_2O_2S$ 393.1631, found 393.1633; $[\alpha]_D^{20}$ = −8.0 (*c* = 0.2, $CHCl_3$); HPLC analysis: 97% ee [Daicel Chiralcel OD column, hexane/*i*PrOH = 85:15, flow rate: 1 mL/min, λ = 215 nm, t_R = 8.25 min (major), 10.57 min (minor)].

3-(9H-Fluoren-2-yl)-4-phenyl-1-tosylimidazolidine ***4h***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.52; colorless solid; yield 61% (142 mg); mp 232−233 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.70 (d, *J* = 7.8 Hz, 2H), 7.59 (d, *J* = 7.8 Hz, 1H), 7.50 (d, *J* = 8.4 Hz, 1H), 7.44 (d, *J* = 7.8 Hz, 1H), 7.30−7.22 (m, 6H), 7.19−7.13 (m, 3H), 6.58 (s, 1H), 6.38 (d, *J* = 8.4 Hz, 1H), 5.10 (d, *J* = 5.4 Hz, 1H), 4.84 (d, *J* = 6.0 Hz, 1H), 4.59 (t, *J* = 6.0 Hz, 1H), 3.95 (t, *J* = 8.4 Hz, 1H), 3.74 (s, 2H), 3.48 (dd, *J* = 10.8, 5.4 Hz, 1H), 2.38 (s, 3H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 145.1, 144.5, 144.4, 142.4, 142.0, 140.6, 133.1, 132.8, 130.1, 129.1, 128.0, 127.9, 126.9, 126.1, 125.5, 124.9, 120.6, 118.9, 112.2, 110.0, 66.5, 61.8, 55.8, 37.2, 21.8; FT-IR (KBr) 3062, 3027, 2956, 2923, 2853, 1616, 1492, 1457, 1384, 1349, 1162, 1092, 1027, 812, 764 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{29}H_{26}N_2O_2S$ 467.1788, found 467.1790.

3,4-Diphenyl-1-(phenylsulfonyl)imidazolidine ***4i***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.54; colorless solid; yield 75% (137 mg); mp 125−126 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.81 (d, J = 7.8 Hz, 2H), 7.57 (t, J = 7.2 Hz, 1H), 7.45 (t, J = 7.2 Hz, 2H), 7.24−7.20 (m, 3H), 7.12−7.10 (m, 4H), 6.72 (t, J = 7.2 Hz, 1H), 6.38 (d, J = 7.8 Hz, 2H), 5.05 (d, J = 6.0 Hz, 1H), 4.78 (d, J = 6.0 Hz, 1H), 4.51 (t, J = 6.6 Hz, 1H), 3.92 (dd, J = 10.8, 7.8 Hz, 1H), 3.46 (dd, J = 10.2, 5.4 Hz, 1H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 144.9, 140.5, 136.0, 133.5, 129.4, 129.3, 129.1, 128.0, 127.9, 126.0, 118.5, 113.3, 66.2, 61.7, 55.7; FT-IR (KBr) 3062, 3028, 2928, 2857, 1600, 1506, 1446, 1351, 1166, 1095, 1030, 749 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{21}H_{20}N_2O_2S$ 365.1318, found 365.1319.

3-Phenyl-1-tosyl-1,2,3,5,6,10b-hexahydroimidazo[2,1-a]isoquinoline ***4j***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.50; colorless solid; yield 63% (127 mg); mp 137−138 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.97 (d, J = 7.8 Hz, 1H), 7.85 (d, J = 7.8 Hz, 2H), 7.41 (d, J = 7.8 Hz, 2H), 7.34 (t, J = 7.8 Hz, 1H), 7.26−7.20 (m, 4H), 7.07 (d, J = 7.8 Hz, 1H), 6.88 (d, J = 7.2 Hz, 2H), 5.94 (s, 1H), 4.10 (t, J = 8.4 Hz, 1H), 3.70 (t, J = 8.4 Hz, 1H), 3.18 (t, J = 9.6 Hz, 1H), 3.16−3.08 (m, 1H), 2.94−2.88 (m, 1H), 2.75−2.72 (m, 1H), 2.53 (s, 3H), 2.40 (d, J = 16.2 Hz, 1H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 144.0, 138.8, 134.9, 134.1, 129.9, 129.2, 128.8, 128.6, 128.3, 127.7, 127.6, 127.2, 76.5, 60.9, 55.7, 41.5, 21.9, 21.4; FT-IR (KBr) 3056, 3028, 2950, 2924, 2861, 1598, 1453, 1344, 1157, 1090, 1005, 817, 759, 665 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{24}H_{24}N_2O_2S$ 405.1631, found 405.1633.

3-(2-Chlorophenyl)-1-tosyl-1,2,3,5,6,10b-hexahydroimidazo[2,1-a]isoquinoline ***4k***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.49; colorless solid; yield 56% (123 mg); mp 145−146 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.91 (d, J = 7.8 Hz, 1H), 7.76 (d, J = 7.8 Hz, 2H), 7.35−7.31 (m, 3H), 7.30−7.24 (m, 2H), 7.13 (t, J = 7.8 Hz, 1H), 7.08 (d, J = 7.2 Hz, 1H), 6.99 (t, J = 7.8 Hz, 1H), 6.62 (d, J = 7.8 Hz, 1H), 6.01 (s, 1H), 4.59 (t, J = 7.8 Hz, 1H), 3.87 (t, J = 9.0 Hz, 1H), 3.17−3.13 (m, 2H), 2.99−2.92 (m, 1H), 2.78−2.75 (m, 1H), 2.50 (s, 3H), 2.41 (d, J = 16.8 Hz, 1H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 143.9, 137.4, 134.6, 134.5, 134.4, 134.0, 129.9, 129.5, 128.9, 128.7, 128.4, 127.9, 127.8, 127.2, 127.1, 76.4, 56.8, 53.8, 41.9, 21.8, 21.5; FT-IR (KBr) 3061, 3022, 2953, 2923, 2850, 1597, 1453, 1347, 1159, 1090, 999, 814, 752, 664 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{24}H_{23}ClN_2O_2S$ 439.1242, found 439.1243.

3-(m-Tolyl)-1-tosyl-1,2,3,5,6,10b-hexahydroimidazo[2,1-a]isoquinoline ***4l***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.51; colorless solid; yield 67% (140 mg); mp 151−152 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.95 (d, J = 7.8 Hz, 1H), 7.86 (d, J = 7.8 Hz, 2H), 7.41 (d, J = 7.8 Hz, 2H), 7.33 (t, J = 7.2 Hz, 1H) 7.25 (d, J = 7.2 Hz, 1H), 7.11−7.04 (m, 3H), 6.70 (d, J = 7.2 Hz, 1H), 6.60 (s, 1H), 5.94 (s, 1H), 4.06 (t, J = 8.4 Hz, 1H), 3.68 (t, J = 8.4 Hz, 1H), 3.16 (t, J = 9.6 Hz, 1H), 3.12−3.07 (m, 1H), 2.94−2.88 (m, 1H), 2.75−2.72 (m, 1H), 2.53 (s, 3H), 2.39 (d, J = 16.8 Hz, 1H), 2.23 (s, 3H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 144.0, 138.9, 138.5, 134.9, 134.2, 134.1, 129.9, 129.2, 129.1, 128.7, 128.6, 128.5, 128.1, 127.7, 127.2, 125.0, 76.6, 60.8, 55.7, 41.5, 21.9, 21.5, 21.4; FT-IR (KBr) 3056, 3025, 2959, 2923, 2851, 1598, 1492, 1453, 1345, 1159, 1091, 1004, 815, 751, 666 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{25}H_{26}N_2O_2S$ 419.1788, found 419.1789.

3-(4-Fluorophenyl)-1-tosyl-1,2,3,5,6,10b-hexahydroimidazo[2,1-a]isoquinoline ***4m***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.48; colorless solid; yield 59% (124 mg); mp 150−151 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.95 (d, J = 7.8 Hz, 1H), 7.84 (d, J = 7.2 Hz, 2H), 7.41 (d, J = 7.8 Hz, 2H), 7.33 (t, J = 7.8 Hz, 1H), 7.25 (d, J = 7.2 Hz, 1H), 7.07 (d, J = 7.8 Hz, 1H), 6.89 (t, J = 7.2 Hz, 2H), 6.82−6.80 (m, 2H), 5.93 (s, 1H), 4.08 (t, J = 7.2 Hz, 1H), 3.68 (t, J = 7.8 Hz, 1H), 3.13−3.08 (m, 2H), 2.92−2.86 (m, 1H), 2.72−2.68 (m, 1H), 2.53 (s, 3H), 2.41 (d, J = 16.8 Hz, 1H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 163.5 (d, $J_{C\text{-}F}$ = 244.5 Hz), 144.1, 134.8, 134.6, 134.2, 134.0, 129.9, 129.2 (d, $J_{C\text{-}F}$ = 9.0 Hz), 128.6, 128.5, 127.8, 127.3, 115.7 (d, $J_{C\text{-}F}$ = 21.0 Hz), 76.5, 60.2, 55.6, 41.5, 21.9, 21.4; FT-IR (KBr) 3062, 3024, 2928, 2901, 1601, 1509, 1453, 1344, 1224, 1159, 1013, 838, 816, 754, 673 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{24}H_{23}FN_2O_2S$ 423.1537, found 423.1538.

3-(4-Bromophenyl)-1-tosyl-1,2,3,5,6,10b-hexahydroimidazo[2,1-a]isoquinoline ***4n***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.48; colorless solid; yield 57% (137 mg); mp 148−149 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.93 (d, J = 7.8 Hz, 1H), 7.82 (d, J = 7.8 Hz, 2H), 7.40 (d, J = 7.8 Hz, 2H), 7.34−7.31 (m, 3H), 7.25 (d, J = 7.8 Hz, 1H), 7.06 (d, J = 7.8 Hz, 1H), 6.72 (d, J = 8.4 Hz, 2H), 5.93 (s, 1H), 4.06 (t, J = 7.8 Hz, 1H), 3.68 (t, J = 9.6 Hz, 1H), 3.12−3.09 (m, 2H), 2.89−2.86 (m, 1H), 2.71−2.68 (m, 1H), 2.53 (s, 3H), 2.41 (d, J = 16.8 Hz, 1H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 144.2, 138.1, 134.7, 134.1, 131.9, 129.9, 129.3, 129.2, 128.6, 128.5, 127.9, 127.3, 122.0, 76.5, 60.3, 55.4, 41.6, 21.9, 21.4; FT-IR (KBr) 3054, 3021, 2923, 2852, 1597, 1486, 1345, 1158, 1010, 818, 749, 667 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{24}H_{23}BrN_2O_2S$ 483.0736, found: 483.0737.

3-(2,4-Dimethylphenyl)-1-tosyl-1,2,3,5,6,10b-hexahydroimidazo-[2,1-a]isoquinoline ***4o***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.54; colorless solid; yield 62% (134 mg); mp 161−162 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.00 (d, J = 7.8 Hz, 1H), 7.83 (d, J = 7.8 Hz, 2H), 7.39−7.35 (m, 3H), 7.28 (d, J = 7.2 Hz, 1H), 7.09 (d, J = 7.8 Hz, 1H), 6.92 (s, 1H), 6.84 (d, J = 8.4 Hz, 1H), 6.61 (d, J = 7.8 Hz, 1H), 5.96 (s, 1H), 4.35 (t, J = 7.8 Hz, 1H), 3.75 (dd, J = 10.8, 8.4 Hz, 1H), 3.15−3.06 (m, 2H), 2.87−2.79 (m, 2H), 2.54 (s, 3H), 2.41 (d, J = 13.8, 1H), 2.29 (s, 3H), 2.15 (s, 3H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 144.0, 137.1, 136.3, 135.0, 134.2, 133.8, 131.3, 129.9, 129.2, 128.6, 128.5, 127.7, 127.3, 127.2, 126.5, 76.2, 56.5, 54.5, 41.6, 21.9, 21.8, 21.1, 19.4; FT-IR (KBr) 3062, 3021, 2922, 2853, 1598, 1452, 1347, 1159, 1010, 816, 748, 664 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{26}H_{28}N_2O_2S$ 433.1944, found 433.1941.

3-Phenyl-1-(phenylsulfonyl)-1,2,3,5,6,10b-hexahydroimidazo-[2,1-a]isoquinoline ***4p***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.50; colorless solid; yield 60% (117 mg); mp 140−141 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.97−7.95 (m, 3H), 7.24 (t, J = 7.2 Hz, 1H), 7.61 (t, J = 7.2 Hz, 2H), 7.34 (t, J = 7.2 Hz, 1H), 7.26−7.20 (m, 4H), 7.07 (d, J = 7.2 Hz, 1H), 6.86 (d, J = 7.2 Hz, 2H), 5.96 (s, 1H), 4.11 (t, J = 7.8 Hz, 1H), 3.71 (t, J = 7.8 Hz, 1H), 3.17 (t, J = 9.0 Hz, 1H), 3.12−3.08 (m, 1H), 2.93−2.88 (m, 1H), 2.75−2.71 (m, 1H), 2.40 (d, J = 16.8 Hz, 1H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 138.7, 137.0, 134.8, 134.1, 133.3, 129.3, 129.2, 128.8, 128.6, 128.5, 128.3, 127.8, 127.6, 127.3, 76.6, 60.8, 55.7, 41.5, 21.4; FT-IR (KBr) 3061, 3027, 2953, 2923, 1606, 1446, 1349, 1161, 1011, 754, 721, 603 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{23}H_{22}N_2O_2S$ 391.1475, found 391.1486.

4-(2-Chlorophenyl)-3-phenyl-1-tosylimidazolidine ***4q***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.54; colorless solid; yield 76% (157 mg); mp 120−121 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.67 (d, J = 7.8 Hz, 2H), 7.37 (d, J = 8.4 Hz, 1H), 7.20−7.16 (m, 3H), 7.13 (t, J = 7.2 Hz, 2H), 7.03 (t, J = 7.8 Hz, 1H), 7.00 (d, J = 9.0 Hz, 1H), 6.74 (t, J = 7.2 Hz, 1H), 6.28 (d, J = 8.4 Hz, 2H), 5.09 (d, J = 6.0 Hz, 1H), 4.84 (dd, J = 7.8, 4.2 Hz, 1H), 4.74 (d, J = 6.0 Hz, 1H), 4.00 (dd, J = 10.8, 7.2 Hz, 1H), 3.54 (dd, J = 10.8, 4.8 Hz, 1H), 2.38 (s, 3H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 144.5, 144.4, 137.6, 132.9, 132.3, 130.1, 129.9, 129.4, 129.0, 128.0, 127.5, 127.4, 118.6, 112.9, 66.0, 59.0, 54.1, 21.7; FT-IR (KBr) 3061, 2956, 2923, 2848, 1600, 1506, 1384, 1352, 1164, 1092, 1032, 814, 750, 664 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{22}H_{21}ClN_2O_2S$ 413.1085, found 413.1083.

3-Phenyl-4-(o-tolyl)-1-tosylimidazolidine ***4r***. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.56; colorless solid; yield 73% (143 mg); mp 144−145 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.70 (dd, J = 8.4, 1.8 Hz, 2H), 7.23 (d, J = 8.4 Hz, 2H), 7.17−7.11 (m, 4H), 7.00−6.96 (m, 2H), 6.73 (t, J = 7.2 Hz, 1H), 6.29 (d, J = 8.4 Hz, 2H), 5.07 (d, J = 6.0 Hz, 1H), 4.62 (d, J = 6.0 Hz, 1H), 4.61 (t, J = 7.2 Hz, 1H), 4.01 (t, J = 9.6 Hz, 1H), 3.39 (dd, J = 10.2, 5.4 Hz, 1H), 2.40 (s, 3H), 2.36 (s, 3H); ^{13}C NMR (150 MHz, $CDCl_3$) δ 144.9, 144.5, 137.9, 134.2, 133.0, 130.9, 130.0, 129.2, 127.9, 127.5, 126.8, 125.4, 118.3, 113.0, 66.2, 58.8, 54.3, 21.7, 19.4; FT-IR (KBr) 3061, 3025, 2925, 2856, 1599, 1506, 1384, 1350, 1164, 1094, 1032, 814, 750 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{23}H_{24}N_2O_2S$ 393.1631, found 393.1631.

4-(3-Bromophenyl)-3-phenyl-1-tosylimidazolidine **4s**. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.53; colorless solid; yield 78% (178 mg); mp 130−131 °C; ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.68 (d, J = 8.4 Hz, 2H), 7.37 (d, J = 7.8 Hz, 1H), 7.24−7.23 (m, 3H), 7.15−7.11 (m, 3H), 7.07 (d, J = 7.8 Hz, 1H), 6.76 (t, J = 7.2 Hz, 1H), 6.37 (d, J = 8.4 Hz, 2H), 5.05 (d, J = 6.0 Hz, 1H), 4.75 (d, J = 6.0 Hz, 1H), 4.49 (t, J = 6.0 Hz, 1H), 3.90 (t, J = 9.6 Hz, 1H), 3.47 (dd, J = 10.8, 4.8 Hz, 1H), 2.40 (s, 3H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 144.7, 144.6, 143.2, 132.9, 131.1, 130.7, 130.1, 129.4, 129.1, 127.9, 124.8, 123.2, 118.8, 113.2, 66.2, 61.2, 55.5, 21.8; FT-IR (KBr) 3070, 3022, 2920, 2817, 1600, 1507, 1385, 1346, 1161, 1088, 1024, 847, 747, 665 cm^{-1}; HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{22}H_{21}BrN_2O_2S$ 457.0580, found 457.0584.

4-(3-Chlorophenyl)-3-phenyl-1-tosylimidazolidine **4t**. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.54; colorless solid; yield 80% (165 mg); mp 140−141 °C; ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.69 (d, J = 7.8 Hz, 2H), 7.24−7.17 (m, 4H), 7.16−7.13 (m, 2H), 7.07 (s, 1H), 7.04 (d, J = 7.2 Hz, 1H), 6.77 (t, J = 7.2 Hz, 1H), 6.38 (d, J = 8.4 Hz, 2H), 5.07 (d, J = 6.0 Hz, 1H), 4.75 (d, J = 6.0 Hz, 1H), 4.51 (t, J = 6.6 Hz, 1H), 3.93 (t, J = 10.2 Hz, 1H), 3.48 (dd, J = 10.8, 4.8 Hz, 1H), 2.40 (s, 3H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 144.7, 144.6, 142.9, 134.9, 132.9, 130.3, 130.0, 129.3, 128.0, 127.9, 126.1, 124.3, 118.7, 113.2, 66.1, 61.2, 55.4, 21.7; FT-IR (KBr) 3061, 3028, 2925, 2889, 2820, 1601, 1508, 1346, 1162, 1027, 849, 758, 667 cm^{-1}; HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{22}H_{21}ClN_2O_2S$ 413.1085, found 413.1084.

4-(3-Phenyl-1-tosylimidazolidin-4-yl)phenyl Acetate **4u**. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.41; pale yellow liquid; yield 77% (168 mg); ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.67 (d, J = 7.2 Hz, 2H), 7.24 (d, J = 7.8 Hz, 2H), 7.13−7.11 (m, 4H), 6.97 (d, J = 7.2 Hz, 2H), 6.74 (t, J = 6.6 Hz, 1H), 6.37 (d, J = 7.2 Hz, 2H), 5.02 (d, J = 5.4 Hz, 1H), 4.74 (d, J = 4.8 Hz, 1H), 4.54 (t, J = 4.8 Hz, 1H), 3.91 (t, J = 8.4 Hz, 1H), 3.44 (dd, J = 10.8, 5.4 Hz, 1H), 2.39 (s, 3H), 2.28 (s, 3H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 169.5, 150.2, 144.9, 144.6, 138.0, 132.9, 130.1, 129.4, 128.0, 127.1, 122.2, 118.6, 113.3, 66.2, 61.2, 55.6, 21.7, 21.3; FT-IR (neat) 3028, 2923, 2854, 1758, 1600, 1504, 1351, 1197, 1163, 1015, 912, 752, 666, 599 cm^{-1}; HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{24}H_{24}N_2O_4S$ 437.1530, found 437.1532.

4-(4-Chlorophenyl)-3-phenyl-1-tosylimidazolidine **4v**. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.53; colorless solid; yield 80% (165 mg); mp 170−171 °C; ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.66 (d, J = 7.8 Hz, 2H), 7.23−7.19 (m, 4H), 7.13 (t, J = 7.8 Hz, 2H), 7.03 (d, J = 8.4 Hz, 2H), 6.75 (t, J = 7.2 Hz, 1H), 6.35 (d, J = 8.4 Hz, 2H), 5.03 (d, J = 6.0 Hz, 1H), 4.73 (d, J = 6.0 Hz, 1H), 4.53 (dd, J = 7.2, 4.8 Hz, 1H), 3.90 (dd, J = 10.2, 7.2 Hz, 1H), 3.46 (dd, J = 10.8, 4.8 Hz, 1H), 2.41 (s, 3H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 144.7, 144.6, 139.2, 133.6, 133.1, 130.1, 129.4, 129.2, 128.0, 127.5, 118.7, 113.2, 66.1, 61.1, 55.6, 21.8; FT-IR (KBr) 3065, 3035, 2922, 2852, 1599, 1506, 1350, 1163, 1091, 813, 778, 664 cm^{-1}; HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{22}H_{21}ClN_2O_2S$ 413.1085, found 413.1085.

4-(4-(Chloromethyl)phenyl)-3-phenyl-1-tosylimidazolidine **4w**. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.46; colorless solid; yield 73% (156 mg); mp 173−174 °C; ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.67 (d, J = 7.2 Hz, 2H), 7.27−7.23 (m, 4H), 7.13−7.10 (m, 4H), 6.74 (t, J = 6.6 Hz, 1H), 6.36 (d, J = 7.2 Hz, 2H), 5.03 (d, J = 4.8 Hz, 1H), 4.74 (d, J = 5.4 Hz, 1H), 4.54 (s, 3H), 3.91 (t, J = 7.8 Hz, 1H), 3.45 (dd, J = 10.8, 4.8 Hz, 1H), 2.40 (s, 3H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 144.8, 144.5, 141.0, 137.1, 133.0, 130.1, 129.4, 128.0, 126.5, 118.6, 113.2, 66.2, 61.4, 55.6, 46.0, 21.8; FT-IR (KBr) 3053, 2965, 2921, 2822, 1600, 1508, 1386, 1347, 1161, 1092, 1028, 816, 744, 667 cm^{-1}; HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{23}H_{23}ClN_2O_2S$ 427.1242, found 427.1240.

4-(4-Bromophenyl)-3-phenyl-1-tosylimidazolidine **4x**. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.53; colorless solid; yield 79% (180 mg); mp 179−180 °C; ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.66 (d, J = 7.8 Hz, 2H), 7.35 (d, J = 8.4 Hz, 2H), 7.23 (d, J = 7.8 Hz, 2H), 7.13 (t, J = 7.8 Hz, 2H), 6.97 (d, J = 7.8 Hz, 2H), 6.75 (t, J = 7.2 Hz, 1H), 6.36 (d, J = 7.8 Hz, 2H), 5.04 (d, J = 6.0 Hz, 1H), 4.73 (d, J = 5.4 Hz, 1H), 4.52 (t, J = 4.8 Hz, 1H), 3.91 (dd, J = 10.8, 7.8 Hz, 1H), 3.47 (dd, J = 10.2, 4.8 Hz, 1H), 2.41 (s, 3H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 144.6, 142.0, 139.7, 133.1, 132.1, 130.0, 129.4, 127.9, 127.8, 121.6, 118.7, 113.2, 66.1, 61.1, 55.5, 21.8; FT-IR (KBr) 3075, 3019, 2897, 2812, 1600, 1504, 1381, 1343, 1158, 1029, 816, 748, 667 cm^{-1}; HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{22}H_{21}BrN_2O_2S$ 457.0580, found 457.0583.

4-(4-Fluorophenyl)-3-phenyl-1-tosylimidazolidine **4y**. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.51; colorless solid; yield 79% (156 mg); mp 181−182 °C; ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.69 (d, J = 7.2 Hz, 2H), 7.24 (d, J = 7.8 Hz, 2H), 7.13 (t, J = 7.2 Hz, 2H), 7.08 (t, J = 7.2 Hz, 2H), 6.93 (t, J = 7.8 Hz, 2H), 6.75 (t, J = 7.2 Hz, 1H), 6.37 (d, J = 7.8 Hz, 2H), 5.05 (d, J = 5.4 Hz, 1H), 4.73 (d, J = 5.4 Hz, 1H), 4.54 (t, J = 6.6 Hz, 1H), 3.89 (t, J = 9.6 Hz, 1H), 3.45 (dd, J = 10.2, 4.2 Hz, 1H), 2.41 (s, 3H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 163.1 (d, $J_{C\text{-}F}$ = 244.5 Hz), 144.7, 144.6, 136.3, 133.0, 130.0, 129.3, 128.0, 127.7 (d, $J_{C\text{-}F}$ = 9.0 Hz), 118.6, 116.0 (d, $J_{C\text{-}F}$ = 22.5), 113.2, 66.1, 61.0, 55.7, 21.7; FT-IR (KBr) 3058, 2923, 2892, 2821, 1601, 1508, 1384, 1346, 1161, 1028, 829, 750, 666 cm^{-1}; HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{22}H_{21}FN_2O_2S$ 397.1381, found 397.1384.

3-Phenyl-4-(p-tolyl)-1-tosylimidazolidine **4z**. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.56; colorless solid; yield 81% (159 mg); mp 147−148 °C; ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.68 (d, J = 8.4 Hz, 2H), 7.23 (d, J = 8.4 Hz, 2H), 7.11 (t, J = 7.2 Hz, 2H), 7.05 (d, J = 7.8 Hz, 2H), 7.00 (d, J = 7.8 Hz, 2H), 6.72 (t, J = 7.2 Hz, 1H), 6.37 (d, J = 7.8 Hz, 2H), 5.01 (d, J = 6.0 Hz, 1H), 4.75 (d, J = 6.0 Hz, 1H), 4.49 (t, J = 6.0 Hz, 1H), 3.90 (dd, J = 10.2, 7.2 Hz, 1H), 3.43 (dd, J = 10.2, 4.8 Hz, 1H), 2.40 (s, 3H), 2.30 (s, 3H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 145.0, 144.4, 137.5, 133.0, 130.0, 129.7, 129.3, 128.0, 126.0, 118.4, 113.2, 66.2, 61.4, 55.9, 21.8, 21.3; FT-IR (KBr) 3060, 3029, 2922, 2855, 1599, 1506, 1350, 1163, 1093, 814, 749, 664 cm^{-1} HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{23}H_{24}N_2O_2S$ 393.1631, found 393.1633.

4-(2,4-Dimethylphenyl)-3-phenyl-1-tosylimidazolidine **4aa**. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.56; colorless solid; yield 72% (146 mg); mp 162−163 °C; ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.67 (d, J = 7.8 Hz, 2H), 7.21 (d, J = 8.4 Hz, 2H), 7.10 (t, J = 8.4 Hz, 2H), 6.97 (s, 1H), 6.84 (d, J = 7.8 Hz, 1H), 6.79 (d, J = 7.8 Hz, 1H), 6.71 (t, J = 7.2 Hz, 1H), 6.27 (d, J = 8.4 Hz, 2H), 5.02 (d, J = 6.0 Hz, 1H), 4.82 (d, J = 6.0 Hz, 1H), 4.56 (t, J = 6.6 Hz, 1H), 3.97 (dd, J = 10.2, 7.2 Hz, 1H), 3.34 (dd, J = 10.8, 6.0 Hz, 1H), 2.39 (s, 3H), 2.29 (s, 3H), 2.26 (s, 3H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 145.0, 144.5, 137.2, 134.9, 134.1, 133.1, 131.7, 130.0, 129.3, 128.0, 127.5, 125.5, 118.2, 113.0, 66.2, 58.8, 54.5, 21.7, 21.2, 19.4; FT-IR (KBr) 3065, 3041, 2921, 2855, 1600, 1506, 1384, 1350, 1164, 1092, 1032, 814, 748, 664 cm^{-1}; HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{24}H_{26}N_2O_2S$ 407.1788, found 407.1782.

4-(Naphthalen-2-yl)-3-phenyl-1-tosylimidazolidine **4ab**. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.56; colorless solid; yield 71% (152 mg); mp 166−167 °C; ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.80−7.79 (m, 1H), 7.75 (d, J = 8.4 Hz, 1H), 7.69−7.65 (m, 3H), 7.51 (s, 1H), 7.46−7.45 (m, 2H), 7.23 (d, J = 8.4 Hz, 1H), 7.15 (d, J = 7.8 Hz, 2H), 7.11 (t, J = 7.8 Hz, 2H), 6.72 (t, J = 7.2 Hz, 1H), 6.44 (d, J = 7.8 Hz, 2H), 5.12 (d, J = 6.0 Hz, 1H), 4.83 (d, J = 6.0 Hz, 1H), 4.71 (t, J = 5.4 Hz, 1H), 4.02 (dd, J = 10.2, 7.8 Hz, 1H), 3.58 (dd, J = 10.8, 5.4 Hz, 1H), 2.31 (s, 3H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 145.0, 144.5, 138.0, 133.4, 133.4, 133.1, 129.9, 129.3, 129.1, 128.0, 127.9, 127.8, 126.5, 126.2, 125.0, 123.9, 118.5, 113.2, 66.2, 61.8, 55.6, 21.7; FT-IR (KBr) 3058, 3047, 2928, 2810, 1599, 1506, 1343, 1158, 1030, 815, 745, 666 cm^{-1}; HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{26}H_{24}N_2O_2S$ 429.1631, found 429.1628.

N-(2-(((2,6-Di-tert-butyl-4-methylphenoxy)methyl)(phenyl)amino)-2-phenylethyl)-4-methylbenzenesulfonamide **5b**. Analytical TLC on silica gel, 1:9 ethyl acetate/hexane R_f = 0.58; colorless liquid; yield 65% (194 mg); ^{1}H NMR (600 MHz, CDCl$_3$) δ 7.71 (d, J = 8.4 Hz, 2H), 7.26−7.20 (m, 5 H), 7.17 (t, J = 7.8 Hz, 2H), 7.05 (d, J = 6.6 Hz, 2H), 6.87 (s, 2H), 6.71 (t, J = 7.2 Hz, 1H), 6.61 (d, J = 7.8 Hz, 2H), 5.17 (s, 1H), 5.10 (t, J = 7.2 Hz, 1H), 4.48 (d, J = 15.0 Hz, 1H), 4.10 (d, J = 15.6 Hz, 1H), 3.93 (dd, J = 14.4, 6.6 Hz, 1H), 3.59 (dd, J = 14.4, 8.4 Hz, 1H), 2.51 (s, 3H), 2.40 (s, 3H), 1.30 (s, 18 H); ^{13}C NMR (150 MHz, CDCl$_3$) δ 153.6, 150.1, 143.5, 138.5, 137.1, 136.2, 130.0, 129.4, 128.5, 127.7, 127.5, 126.3, 125.2, 117.4, 113.7, 60.7, 53.0, 48.2, 34.4, 32.4, 30.3, 21.7; FT-IR (neat) 3061, 3028, 2956, 2917, 2870, 1597, 1503, 1433, 1339, 1159, 1090, 926, 748 cm^{-1}; HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{37}H_{46}N_2O_3S$ 599.3302, found 599.3303.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.joc.6b01850.

NMR (^1H and ^{13}C) spectra, HRMS of the reaction mixture of **5a−b**, and HPLC of **2a′**, **2c′**, **4a′**, and **4g′** (PDF)

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Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

We thank the Science and Engineering Research Board (EMR-2015-43) and the Council of Scientific and Industrial Research (02(0255)/16/EMR-II) for the financial support. V.S. thanks the Council of Scientific and Industrial Research and M.S thanks the University Grants Commission for Research Fellowships. We are grateful to the Central Instrumentation Facility, Indian Institute of Technology Guwahati, for NMR and Mass facilities.

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