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# Chemo- and Regioselective Palladium(II)-Catalyzed Aminoarylation of *N*-Allylureas Providing 4-Arylmethyl Imidazolidinones

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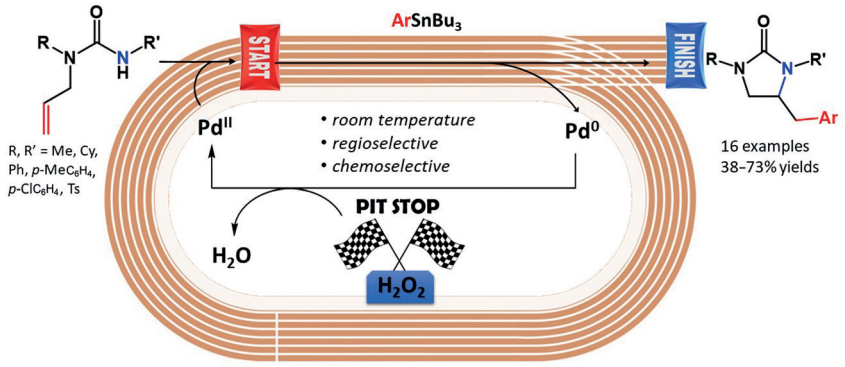


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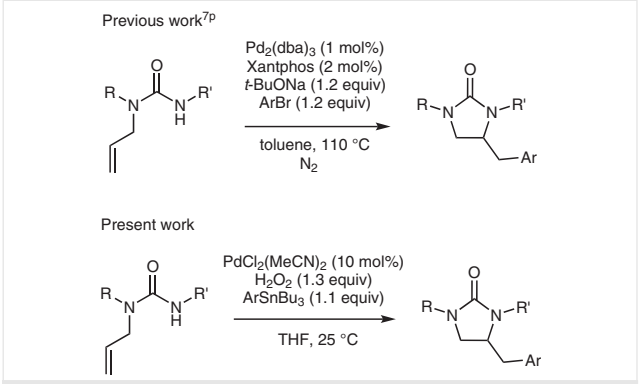
**Abstract** The aminoarylation reaction of *N*-allylureas under oxidative palladium catalysis, in the absence of ligands and by using inexpensive $H_2O_2$ as the sole oxidant, occurs in a chemo- and regioselective fashion. This intramolecular process takes place via a 5-*exo*-trig cyclization, and represents an easy approach to 4-arylmethyl imidazolidinones.

**Key words** palladium catalysis, aminoarylation, hydrogen peroxide, imidazolidinones, ureas, domino reaction

The synthesis of complex molecules through the formation of more than one bond in a single step represents a powerful tool for organic chemists.[1] Among the transition-metal-catalyzed reactions, the double functionalization of unsaturated systems under palladium catalysis represents a rapid and economical method to obtain functionalized substrates. In particular, when the domino process involved the formation of an intramolecular C-N bond on alkenes, alkynes or allenes combined to a new C-C, C-O or C-N bond formation, the result was the synthesis of functionalized (poly)heterocyclic systems.[2] Synthetic strategies involving the palladium catalyst under oxidative conditions[3] represent a complementary reactivity to the Pd(0)-catalyzed reactions of aryl(alkyl) halides, offering the possibility of alternative regioselectivity during bond formation. In this context, the use of hydrogen peroxide as the sole oxidant is quite rare.[4]

In continuation of our interest in the difunctionalization reactions of alkenes, we have applied palladium-catalyzed reactions in arylation/halogenation, arylation/esterification, aminohalogenation and diamination processes.[5] Recently we focused our attention on oxyarylation studies and reported the formation of oxazepanes through a selective 7-*endo*-cyclization process.[6]

Based on literature data, aminoarylation reactions are of great interest, as intra- or intermolecular processes, providing heterocyclic systems.[7] In particular, 4-arylmethyl imidazolidinones have been prepared from *N*-allylureas and aryl bromides. The reactions are performed under Pd(0) catalysis in the presence of a ligand and a base in toluene at 110 °C under $N_2$ (Scheme 1).

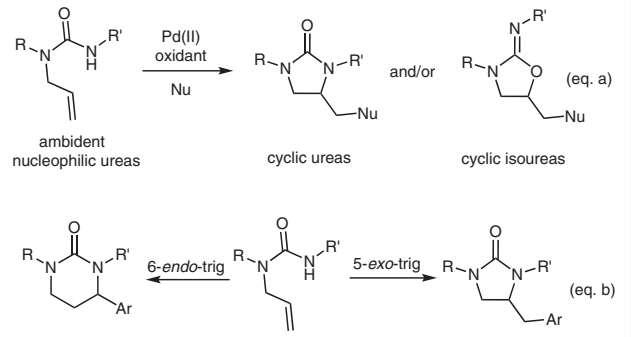



**Scheme 1** Aminoarylation of secondary *N*-allylureas

In the present work, we were interested in extending the arylation process combined with the formation of new bonds, with the purpose of: (a) searching for new conditions for the difunctionalization of alkenes and heterocyclization, (b) investigating in-depth the chemo- and regioselectivity of the process, and (c) achieving mild reaction conditions.

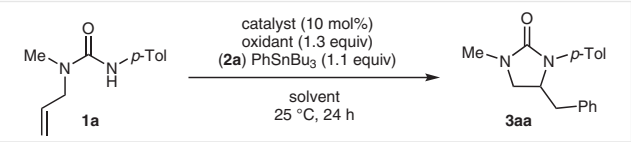
Toward this aim, we envisaged the reactivity of *N*-allylureas as ambident nucleophiles, showing the possibility of C–N vs C–O bond formation and allowing the construction of cyclic ureas or isoureas.[8] The presence of a second nucleophile among the reagents paved the way for a difunctionalization process (Scheme 2, eq. a). Moreover, these sub-

strates offered the possibility of regioselective 5-*exo*-trig/6-*endo*-trig cyclization in the amination step, with the presence of an aryl nucleophile resulting in the aminoarylation reaction involving the formation of C–N and C–C bonds (Scheme 2, eq. b).




**Scheme 2** Chemo- and regioselectivity of secondary *N*-allylureas

Substrate **1a**, prepared in quantitative yield from the simple one-step reaction between *p*-tolylisocyanate and methylallylamine at room temperature in acetonitrile, was selected for this study. Initially we tested the use of a catalytic amount of $PdCl_2(MeCN)_2$ (10 mol%) in the presence of a slight excess of both $CuCl_2$ as oxidant and phenyltributylstannane (**2a**) as nucleophile. The complete conversion of the substrate afforded the aminoarylation product **3aa** (39% yield) along with 4-chloromethyl-imidazolidinone **4** (35% yield), arising from an aminohalogenation process due to the double action of the copper salt as an oxidant and a nucleophile (Scheme 3).




**Scheme 3** Aminoarylation reaction of the substrate **1a** with **2a**

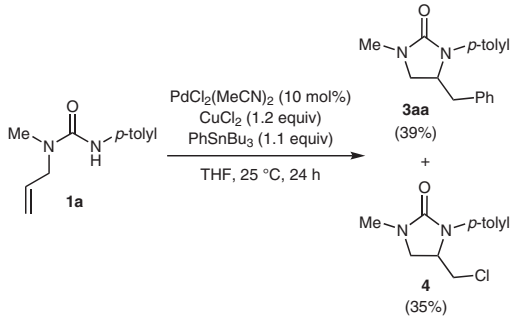
The observed cyclization was totally chemoselective due to the addition of the nitrogen to the double bond, resulting in formation of the cyclic urea. Moreover, the reaction gave exclusive 5-*exo*-trig cyclization, showing total regioselectivity with the formation of 4-substituted imidazolidinones. The selective formation of the imidazolidinone scaffold is a valuable goal because of the presence of this heterocycle in numerous natural products[9a] and in a wide range of pharmaceuticals including anti-inflammatory,[9b] anti-infective,[9c] antibacterial[9d] and antitumor agents.[9e–g]

It is worth noting the *exo*-cyclization process. In fact, when *exo*-cyclizations on protected aminoalkenes have been reported,[10] generally, palladium-catalyzed difunctionalization of 4-pentenyl-amides under oxidative conditions afforded hexatomic six-membered rings, a result which was never observed in our experiments (see Table 1).[4a,b,7q,10a,11] Moreover, palladium-catalyzed oxidative oxyarylation reactions highlighted the preference for the *endo*-cyclization.[6]

To improve the yield of the reaction, and to exclude the formation of the 4-chloromethy-limidazolidinone by-product, we tested different reaction conditions by changing the oxidant and the solvent (Table 1). The use of $Cu(OAc)_2$ as the oxidant was unfruitful (entry 2), as were a silver salt (entry 3) and 1,4-benzoquinone (BQ) (entry 4). Positive results were obtained with inexpensive $H_2O_2$ as the sole oxidant (entry 5). The palladium catalyst was essential for the outcome of the reaction (entry 6), whereas solvents other than THF did not result in formation of the desired product (entries 7 and 8).

The use of different aryl nucleophiles such as aryl boronic acids, Grignard reagents and arylzinc bromide didn't provide any addition. To exploit the central role of the arylstannane reagent, 4-benzyloxyphenyltributylstannane (**2b**) was also tested with various *N*-allylureas (**1b–h**). Using the optimized conditions (see Table 1, entry 5), the reactions proceeded smoothly providing exclusive formation of the 4-substituted imidazolidinones **3**, confirming the selective 5-*exo* cyclization (Table 2). Only in two cases (the reactions

**Table 1** Optimization of the Reaction Conditions for the Aminoarylation of *N*-Allylurea **1a**[a]

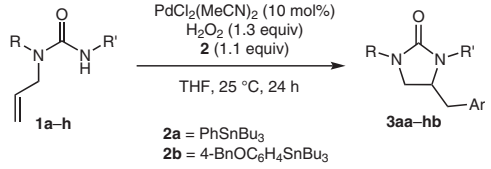



| Entry | Catalyst | Oxidant | Solvent | **3aa** (%)[b] |
|---|---|---|---|---|
| 1 | $PdCl_2(MeCN)_2$ | $CuCl_2$ | THF | 39[c] |
| 2 | $PdCl_2(MeCN)_2$ | $Cu(OAc)_2$ | THF | – |
| 3 | $PdCl_2(MeCN)_2$ | $AgCO_3$ | THF | traces |
| 4 | $PdCl_2(MeCN)_2$ | BQ | THF | traces |
| 5 | $PdCl_2(MeCN)_2$ | $H_2O_2$ | THF | 59 |
| 6 | – | $H_2O_2$ | THF | –[d] |
| 7 | $PdCl_2(MeCN)_2$ | $H_2O_2$ | MeCN | traces |
| 8 | $PdCl_2(MeCN)_2$ | $H_2O_2$ | DMF | traces |

[a] Reaction conditions: **1a** (0.25 mmol), catalyst (10 mol%), oxidant (1.3 equiv), arylating agent (1.1 equiv), solvent (5 mL).
[b] Yield of isolated product.
[c] Compound **4** was also isolated in 35% yield.
[d] Starting material was recovered.

of substrates **1a** and **1g** with arylstannanes **2b** and **2a**, respectively) were the corresponding products not isolated from the reaction mixture.

**Table 2** Aminoarylation of *N*-Allylureas **1a–h**




| Substrate | R | R′ | $ArSnBu_3$ | Product | Yield (%)[a] |
|---|---|---|---|---|---|
| **1a** | Me | *p*-tolyl | **2a** | **3aa** | 73 |
| **1a** | Me | *p*-tolyl | **2b** | **3ab** | – |
| **1b** | Me | Ts | **2a** | **3ba** | 59 |
| **1b** | Me | Ts | **2b** | **3bb** | 48 |
| **1c** | Me | $4\text{-ClC}_6H_4$ | **2a** | **3ca** | 66 |
| **1c** | Me | $4\text{-ClC}_6H_4$ | **2b** | **3cb** | 51 |
| **1d** | cyclohexyl | Ts | **2a** | **3da** | 62 |
| **1d** | cyclohexyl | Ts | **2b** | **3db** | 71 |
| **1e** | cyclohexyl | $4\text{-ClC}_6H_4$ | **2a** | **3ea** | 58 |
| **1e** | cyclohexyl | $4\text{-ClC}_6H_4$ | **2b** | **3eb** | 50 |
| **1f** | Ph | $4\text{-ClC}_6H_4$ | **2a** | **3fa** | 38 |
| **1f** | Ph | $4\text{-ClC}_6H_4$ | **2b** | **3fb** | 56 |
| **1g** | Ph | Ts | **2a** | **3ga** | – |
| **1g** | Ph | Ts | **2b** | **3gb** | 55 |
| **1h** | Ph | $4\text{-MeC}_6H_4$ | **2a** | **3ha** | 59 |
| **1h** | Ph | $4\text{-MeC}_6H_4$ | **2b** | **3hb** | 54 |

[a] Yield of isolated product.

The ring structure was unambiguously confirmed by single-crystal X-ray diffraction analysis of the product **3ea** (Figure 1).[12]

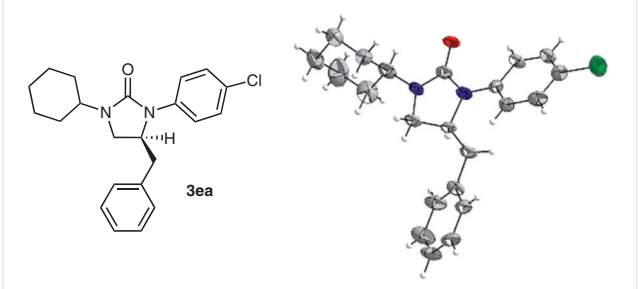
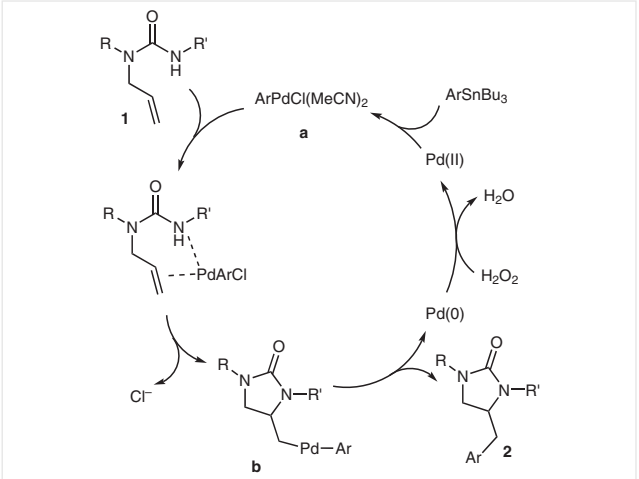


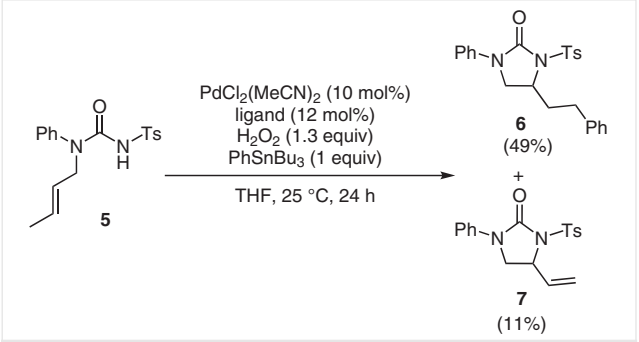

**Figure 1** Left: molecular structure of the *S*-enantiomer of compound **3ea**, as determined by single-crystal X-ray diffraction. Right: ORTEP representation showing the corresponding in-crystal molecular conformation of (*S*)-**3ea** at room temperature. Thermal ellipsoids are drawn at the 25% probability level. C: gray; H: white; O: red; N: blue; Cl: green.

On the basis of the previous results reported on the aminoarylation of alkenes under Pd(II) catalysis,[7e,i,r,s] a plausible mechanism is shown in Scheme 4. Arylpalladium(II) species (**a**) was initially formed by transmetalation of the arylstannane, followed by insertion of the double bond generating the σ-alkyl-Pd(II) intermediate (**b**). Reductive elimination of Pd(0) and oxidation to Pd(II) by $H_2O_2$ completed the catalytic cycle.



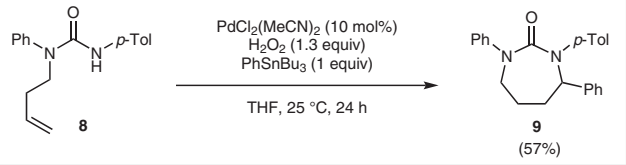

**Scheme 4** Proposed mechanism for the 5-*exo*-trig cyclization of substrates **1**

The aminoarylation process was also applied to a nonterminal alkene. *N*-2-Butenyl-*N*-phenyl-*N*′-tosyl-urea **5** afforded 4-(2-phenylethyl)-imidazolidinone **6** as the aminoarylation product (Scheme 5). In this case the process was improved by using the Pd complex in the presence of (*S*)-4-*tert*-butyl-2-(2-pyridyl)oxazoline as the ligand. The structure of the obtained product was justified through the isolation of 4-vinyl-imidazolidinone **7**, which resulted from the amination step only. The subsequent aryl insertion step with reverse regiochemistry afforded the aminoarylation product **6**.




**Scheme 5** Aminoarylation reaction of substrate **5**

We also applied the developed procedure for the aminoarylation of *N*-homoallylic substituted urea **8** with phenyltributylstannane under the same reaction conditions. The corresponding product, 4-phenyl-1,3-diazepan-2-one **9**, was obtained in 57% yield via a 7-*endo*-trig cyclization process (Scheme 6). This result supported a mechanism similar to that suggested in Scheme 4.[6] Initially, the double bond of the substrate **8** underwent the arylation step (through the insertion of PhPdCl arising from transmetalation) with the formation of the σ-alkyl-Pd(II) intermediate. β-Hydride elimination and a subsequent amination step, with reverse regiochemistry involving the benzylic position, provided the 4-phenyl-1,3-diazepane **9**.




**Scheme 6** Aminoarylation reaction of substrate **8**

In summary, we have developed a process for the aminoarylation of *N*-allylureas in the presence of aryltributylstannanes. The reactions are performed under ligand-free Pd catalysis using $H_2O_2$ as the oxidant. The reaction is chemo- and regioselective, affording the corresponding imidazolidinones as the exclusive cyclic products.

Thin-layer chromatographic separations were performed on precoated Merck silica gel 60-$F_{254}$. Preparative separations were performed by flash chromatography using Merck silica gel (0.035–0.070 mm). Melting points were determined by the capillary method with a Büchi B-540 apparatus and are uncorrected. IR spectra were measured with a Jasco FT/IR 5300 spectrophotometer. For oil compounds were used NaCl disks and for solid compounds KBr pellets. $^1$H NMR and $^{13}$C NMR spectra were recorded with an AVANCE 400 Bruker spectrometer at 400 and 100 MHz, a Varian Oxford 300 MHz spectrometer at 300 and 75 MHz and an AVANCE 500 Bruker spectrometer at 500 and 125 MHz, respectively. Chemical shifts are given as δ values in ppm relative to residual solvent peaks ($CHCl_3$) as the internal reference. $^{13}$C NMR spectra are $^1$H-decoupled and the determination of the multiplicities was achieved from the APT pulse sequence. Mass spectra were recorded using electrospray ionization (ESI) technique on a LCQ Advantage Thermo Finningan spectrometer. Elemental analyses were executed on a Perkin-Elmer CHN Analyzer Series II 2400.

### *N*-Protected Ureas 1, 5 and 8; General Procedure

To a solution of the appropriate amine (4 mmol) in MeCN (40 mL) was added dropwise the isocyanate (4 mmol) at 0 °C under an inert atmosphere. The reaction mixture was allowed to warm to room temperature and stirred for 24 h. The solvent was evaporated under reduced pressure and the crude product was used without any further purification.

### *N*-Allyl-*N*-methyl-*N*′-tolyl-urea (1a)[13]

White solid; yield: 620 mg (76%); mp 110 °C (hexane/$Et_2O$).

$^1$H NMR (300 MHz, $CDCl_3$): δ = 7.35–7.19 (m, 2 H), 7.09 (d, *J* = 8.2 Hz, 2 H), 6.73 (br s, 1 H), 6.10–5.66 (m, 1 H), 5.43–5.15 (m, 2 H), 3.96 (d, *J* = 5.4 Hz, 2 H), 3.00 (s, 3 H), 2.29 (s, 3 H).

### *N*-Allyl-*N*-methyl-*N*′-tosyl-urea (1b)[14]

Colorless oil; yield: 1040 mg (97%).

$^1$H NMR (300 MHz, $CDCl_3$): δ = 7.93 (d, *J* = 8.3 Hz, 2 H), 7.38–7.27 (m, 3 H), 5.72 (ddt, *J* = 15.8, 10.5, 5.3 Hz, 1 H), 5.17 (dd, *J* = 20.5, 13.7 Hz, 2 H), 3.83 (d, *J* = 5.3 Hz, 2 H), 2.86 (s, 3 H), 2.43 (s, 3 H).

### *N*-Allyl-*N*-methyl-*N*′-(4-chlorophenyl)urea (1c)[13]

White solid; yield: 842 mg (94%); mp 115 °C (hexane/$Et_2O$).

$^1$H NMR (400 MHz, $CDCl_3$): δ = 7.32 (d, *J* = 8.8 Hz, 2 H), 7.23 (d, *J* = 8.8 Hz, 2 H), 6.45 (br s, 1 H), 5.76–5.94 (m, 1 H), 5.25–5.30 (m, 2 H), 3.97 (d, *J* = 5.2 Hz, 2 H), 3.01 (s, 3 H).

### *N*-Allyl-*N*-cyclohexyl-*N*′-tosyl-urea (1d)

Yellow oil; yield: 1317 mg (98%).

IR: 1658 $cm^{-1}$.

$^1$H NMR (400 MHz, $CDCl_3$): δ = 7.79 (d, *J* = 8.1 Hz, 2 H), 7.26 (d, *J* = 8.5 Hz, 2 H), 5.99–5.84 (m, 1 H), 5.35–5.18 (m, 2 H), 3.52 (d, *J* = 6.7 Hz, 2 H), 2.91–2.72 (m, 1 H), 2.40 (s, 3 H), 1.98–0.96 (m, 10 H).

$^{13}$C NMR (101 MHz, $CDCl_3$): δ = 155.3 (s), 143.4 (s), 142.5 (s), 139.3 (s), 129.9 (d), 129.6 (d), 126.4 (d), 121.7 (t), 55.7 (d), 47.0 (t), 29.5 (t), 25.9 (t), 25.1 (t), 24.6 (t), 21.5 (q).

MS: *m*/*z* = 337.26 $[M + H]^+$.

### *N*-Allyl-*N*-cyclohexyl-*N*′-(4-chlorophenyl)urea (1e)

White solid; yield: 1113 mg (95%); mp 122 °C (hexane/$Et_2O$).

IR: 1663 $cm^{-1}$.

$^1$H NMR (300 MHz, $CDCl_3$): δ = 7.31–7.23 (m, 2 H), 7.23–7.15 (m, 2 H), 6.52 (br s, 1 H), 5.90 (ddt, *J* = 17.3, 10.1, 5.0 Hz, 1 H), 5.54–5.14 (m, 2 H), 4.23 (tt, *J* = 11.5, 3.4 Hz, 1 H), 3.83 (dt, *J* = 4.8, 1.8 Hz, 2 H), 1.95–0.90 (m, 10 H).

$^{13}$C NMR (75 MHz, $CDCl_3$): δ = 155.4 (s), 138.1 (s), 136.2 (d), 128.7 (d), 127.5 (s), 120.6 (d), 117.2 (s), 54.2 (d), 45.1 (t), 31.0 (t), 25.8 (t), 25.5 (t).

MS: *m*/*z* = 293.75 $[M + H]^+$.

Anal. Calcd for $C_{16}H_{21}ClN_2O$: C, 65.63; H, 7.23; N, 9.57. Found: C, 65.55; H, 7.29; N, 9.69.

### *N*-Allyl-*N*-phenyl-*N*′-(4-chlorophenyl)urea (1f)

White solid; yield: 1098 mg (96%); mp 75–76 °C (hexane/$Et_2O$).

IR: 1670 $cm^{-1}$.

$^1$H NMR (300 MHz, $CDCl_3$): δ = 7.62–7.07 (m, 9 H), 6.19 (br s, 1 H), 6.08–5.76 (m, 1 H), 5.29–5.05 (m, 2 H), 4.35 (d, *J* = 6.1 Hz, 2 H).

$^{13}$C NMR (75 MHz, $CDCl_3$): δ = 153.8 (s), 141.1 (s), 137.4 (s), 133.9 (d), 130.3 (d), 128.7 (d), 128.5 (d), 128.3 (d), 127.8 (s), 120.4 (d), 117.6 (t), 52.3 (t).

MS: *m*/*z* = 287.54 $[M + H]^+$.

Anal. Calcd for $C_{16}H_{15}ClN_2O$: C, 67.02; H, 5.27; N, 9.77. Found: C, 66.96; H, 5.36; N, 9.81.

**N-Allyl-N-phenyl-N′-tosyl-urea (1g)**

Orange solid; yield: 1293 mg (98%); mp 74–76 °C (hexane/$Et_2O$).

IR: 1696 $cm^{-1}$.

$^1H$ NMR (400 MHz, $CDCl_3$): δ = 7.93 (d, *J* = 8.3 Hz, 2 H), 7.53–7.38 (m, 3 H), 7.38–7.26 (m, 2 H), 7.19 (d, *J* = 7.2 Hz, 2 H), 7.09 (br s, 1 H), 5.79 (ddt, *J* = 16.7, 10.2, 6.4 Hz, 1 H), 5.13–4.97 (m, 2 H), 4.18 (d, *J* = 6.3 Hz, 2 H), 2.46 (s, 3 H).

$^{13}C$ NMR (101 MHz, $CDCl_3$): δ = 150.1 (s), 144.6 (s), 139.3 (s), 136.1 (s), 132.4 (d), 130.6 (d), 129.5 (d), 129.1 (d), 128.4 (d), 128.3 (d), 118.7 (t), 52.4 (t), 21.7 (q).

MS (ESI): *m*/*z* = 353.67 $[M + Na]^+$.

Anal. Calcd for $C_{17}H_{18}N_2O_3S$: C, 61.80; H, 5.49; N, 8.48. Found: C, 61.66; H, 5.67; N, 8.31.

**N-Allyl-N-phenyl-N′-tolyl-urea (1h)**

Pale-yellow solid; yield: 1000 mg (94%); mp 83–84 °C (hexane/$Et_2O$).

IR: 1660 $cm^{-1}$.

$^1H$ NMR (300 MHz, $CDCl_3$): δ = 7.69–7.24 (m, 5 H), 7.24–7.16 (m, 2 H), 7.06 (d, *J* = 8.3 Hz, 2 H), 6.15 (br s, 1 H), 5.97 (ddt, *J* = 17.4, 9.8, 6.1 Hz, 1 H), 5.27–4.96 (m, 2 H), 4.37 (dt, *J* = 6.1, 1.2 Hz, 2 H), 2.29 (s, 3 H).

$^{13}C$ NMR (75 MHz, $CDCl_3$): δ = 154.1 (s), 141.5 (s), 136.2 (s), 134.2 (d), 132.5 (s), 130.2 (d), 129.3 (d), 128.5 (d), 128.0 (d), 119.4 (d), 117.4 (t), 52.3 (t), 20.7 (q).

MS: *m*/*z* = 267.27 $[M + H]^+$, 289.29 $[M + Na]^+$.

Anal. Calcd for $C_{17}H_{18}N_2O$: C, 76.66; H, 6.81; N, 10.52. Found: C, 76.54; H, 6.90; N, 10.57.

**(E)-N-But-2-en-1-yl-N-phenyl-N′-tosyl-urea (5)**

White solid; yield: 1087 mg (79%); mp 104–107 °C (dec.).

IR: 1690 $cm^{-1}$.

$^1H$ NMR (300 MHz, $CDCl_3$): δ = 8.01–7.85 (m, 2 H), 7.50–7.26 (m, 5 H), 7.19–7.11 (m, 2 H), 6.93 (br s, 1 H), 5.46–5.36 (m, 2 H), 4.12–4.01 (m, 2 H), 2.44 (s, 3 H), 1.58 (ddd, *J* = 4.0, 2.0, 1.0 Hz, 3 H).

$^{13}C$ NMR (75 MHz, $CDCl_3$): δ = 149.9 (s), 144.5 (s), 139.4 (s), 136.2 (s), 130.4 (d), 130.2 (d), 129.4 (d), 129.0 (d), 128.4 (d), 128.3 (d), 125.1 (d), 51.7 (t), 21.6 (q), 17.6 (q).

MS: *m*/*z* = 345.62 $[M + H]^+$.

Anal. Calcd for $C_{18}H_{20}N_2O_3S$: C, 62.77; H, 5.85; N, 8.13. Found: C, 62.86; H, 5.79; N, 8.04.

**1-(But-3-en-1-yl)-1-phenyl-3-(p-tolyl)urea (8)**

Pale-yellow oil; yield: 1001 mg (89%).

IR: 1678 $cm^{-1}$.

$^1H$ NMR (300 MHz, $CDCl_3$): δ = 7.49 (t, *J* = 7.4 Hz, 2 H), 7.38 (dd, *J* = 15.1, 7.7 Hz, 1 H), 7.32 (d, *J* = 7.2 Hz, 2 H), 7.15 (d, *J* = 8.4 Hz, 2 H), 7.03 (d, *J* = 8.3 Hz, 2 H), 6.03 (s, 1 H), 5.80 (ddt, *J* = 17.0, 10.2, 6.8 Hz, 1 H), 5.06 (dd, *J* = 21.0, 5.3 Hz, 2 H), 3.92–3.73 (m, 2 H), 2.39–2.28 (m, 2 H), 2.26 (s, 3 H).

$^{13}C$ NMR (75 MHz, $CDCl_3$): δ = 154.2 (s), 141.4 (s), 136.3 (s), 135.4 (d), 132.4 (s), 130.3 (d), 129.2 (d), 128.8 (d), 128.1 (d), 119.4 (d), 116.6 (t), 48.7 (t), 32.9 (t), 20.7 (q).

MS: *m*/*z* = 281.18 $[M + H]^+$.

Anal. Calcd for $C_{18}H_{20}N_2O$: C, 77.11; H, 7.19; N, 9.99. Found: C, 77.33; H, 7.12; N, 9.90.

**Aminoarylation of Ureas; General Procedure**

A mixture of urea **1**, **5** or **8** (0.25 mmol), $PdCl_2(MeCN)_2$ (10 mol%), $H_2O_2$ (0.33 mmol) and arylstannane **2a** or **2b** (0.25 mmol) in THF (0.2 M) was stirred at room temperature. In the case of substrate **5**, the ligand (*S*)-4-*tert*-butyl-2-(2-pyridyl)oxazoline (12 mol%) was also added to the reaction mixture. After 24 h, the solvent was evaporated under reduced pressure and $H_2O$ (10 mL) was added. The aqueous layer was extracted with $CH_2Cl_2$ (3 × 10 mL) and the combined organic layer dried was over $Na_2SO_4$ and concentrated under reduced pressure. The crude residue was purified by silica gel column chromatography to afford the corresponding imidazolidinones **3**, **4**, **6** and **7** or diazepane **9**.

**4-Benzyl-1-methyl-3-(p-tolyl)imidazolidin-2-one (3aa)**

Yellow oil; yield: 51 mg (73%); $R_f$ = 0.37 (hexane/EtOAc, 3:1).

IR: 1680 $cm^{-1}$.

$^1H$ NMR (400 MHz, $CDCl_3$): δ = 7.34 (d, *J* = 8.3 Hz, 2 H), 7.30–7.03 (m, 7 H), 4.40–4.29 (m, 1 H), 3.27 (t, *J* = 8.7 Hz, 1 H), 3.14–3.00 (m, 2 H), 2.72 (s, 3 H), 2.59 (dd, *J* = 13.6, 9.7 Hz, 1 H), 2.27 (s, 3 H).

$^{13}C$ NMR (101 MHz, $CDCl_3$): δ = 136.7 (s), 136.2 (s), 133.4 (s), 129.6 (d), 129.2 (d), 128.7 (d), 126.8 (d), 121.4 (d), 54.8 (d), 49.4 (t), 38.1 (t), 31.1 (q), 20.8 (q).

MS: *m*/*z* = 281.51 $[M + H]^+$.

Anal. Calcd for $C_{18}H_{20}N_2O$: C, 77.11; H, 7.19; N, 9.99. Found: C, 77.22; H, 7.12; N, 9.91.

**4-(Chloromethyl)-1-methyl-3-(p-tolyl)imidazolidin-2-one (4)**

Yellow oil; yield: 54 mg (35%); $R_f$ = 0.39 (hexane/EtOAc, 3:1).

IR: 1696 $cm^{-1}$.

$^1H$ NMR (400 MHz, $CDCl_3$): δ = 7.32 (d, *J* = 8.2 Hz, 2 H), 7.17 (d, *J* = 8.0 Hz, 2 H), 4.60–4.27 (m, 1 H), 3.70–3.59 (m, 2 H), 3.56–3.38 (m, 2 H), 2.90 (s, 3 H), 2.34 (s, 3 H).

$^{13}C$ NMR (101 MHz, $CDCl_3$): δ = 158.1 (s), 135.5 (s), 134.2 (s), 129.8 (d), 121.7 (d), 54.5 (d), 48.4 (t), 43.8 (t), 31.0 (q), 20.8 (q).

MS: *m*/*z* = 239.34 $[M + H]^+$.

Anal. Calcd for $C_{12}H_{15}ClN_2O$: C, 60.38; H, 6.33; N, 11.74. Found: C, 60.49; H, 6.30; N, 11.68.

**4-Benzyl-1-methyl-3-tosylimidazolidin-2-one (3ba)**

Yellow oil; yield: 52 mg (59%); $R_f$ = 0.43 (hexane/EtOAc, 3:1).

IR: 1690 $cm^{-1}$.

$^1H$ NMR (400 MHz, $CDCl_3$): δ = 7.91 (d, *J* = 8.2 Hz, 2 H), 7.29–7.07 (m, 7 H), 4.54–4.34 (m, 1 H), 3.43 (dd, *J* = 13.0, 3.6 Hz, 1 H), 3.19 (t, *J* = 9.0 Hz, 1 H), 2.98 (dd, *J* = 9.2, 3.4 Hz, 1 H), 2.71 (dd, *J* = 13.2, 9.9 Hz, 1 H), 2.58 (s, 3 H), 2.35 (s, 3 H).

$^{13}C$ NMR (101 MHz, $CDCl_3$): δ = 153.9 (s), 144.6 (s), 136.6 (s), 135.7 (s), 129.6 (d), 129.5 (d), 128.8 (d), 128.2 (d), 127.2 (d), 115.2 (d), 54.9 (d), 48.7 (t), 40.8 (t), 30.5 (q), 21.7 (q).

MS: *m*/*z* = 345.32 $[M + H]^+$.

Anal. Calcd for $C_{18}H_{20}N_2O_3S$: C, 62.77; H, 5.85; N, 8.13. Found: C, 62.83; H, 5.80; N, 8.17.

**4-(4-Benzyloxyphenyl)methyl-1-methyl-3-tosylimidazolidin-2-one (3bb)**

Yellow oil; yield: 54 mg (48%); $R_f$ = 0.39 (hexane/EtOAc, 3:1).

IR: 1696 $cm^{-1}$.

$^1$H NMR (400 MHz, $CDCl_3$): δ = 7.89 (dd, *J* = 18.8, 8.3 Hz, 2 H), 7.43–7.20 (m, 7 H), 7.04 (d, *J* = 8.4 Hz, 2 H), 6.85 (d, *J* = 8.4 Hz, 2 H), 4.97 (s, 2 H), 4.46–4.26 (m, 1 H), 3.34 (dd, *J* = 13.5, 3.3 Hz, 1 H), 3.19 (t, *J* = 9.0 Hz, 1 H), 2.97 (dd, *J* = 9.4, 3.2 Hz, 1 H), 2.66 (dd, *J* = 13.4, 9.7 Hz, 1 H), 2.57 (s, 3 H), 2.35 (s, 3 H).

$^{13}$C NMR (101 MHz, $CDCl_3$): δ = 158.0 (s), 153.9 (s), 144.5 (s), 136.9 (s), 136.6 (s), 130.5 (d), 129.6 (d), 128.6 (d), 128.2 (d), 128.0 (d), 127.9 (s), 127.5 (d), 70.1 (t), 55.0 (d), 48.7 (t), 39.9 (t), 30.5 (q), 21.7 (q).

MS: *m*/*z* = 451.39 $[M + H]^+$.

Anal. Calcd for $C_{25}H_{26}N_2O_4S$: C, 66.65; H, 5.82; N, 6.22. Found: C, 66.51; H, 5.89; N, 6.20.

### 4-Benzyl-1-methyl-3-(4-chlorophenyl)imidazolidin-2-one (3ca)

White solid; yield: 50 mg (66%); mp 90 °C (hexane/$Et_2O$); $R_f$ = 0.41 (hexane/EtOAc, 3:1).

IR: 1651 $cm^{-1}$.

$^1$H NMR (400 MHz, $CDCl_3$): δ = 7.53 (d, *J* = 8.8 Hz, 2 H), 7.42–7.21 (m, 5 H), 7.16 (d, *J* = 7.2 Hz, 2 H), 4.45 (ddd, *J* = 12.9, 8.6, 4.1 Hz, 1 H), 3.40 (t, *J* = 8.8 Hz, 1 H), 3.23 (dd, *J* = 8.9, 4.7 Hz, 1 H), 3.12 (dd, *J* = 13.8, 3.1 Hz, 1 H), 2.82 (s, 3 H), 2.72 (dd, *J* = 13.7, 9.4 Hz, 1 H).

$^{13}$C NMR (101 MHz, $CDCl_3$): δ = 137.5 (s), 136.2 (s), 131.3 (s), 129.2 (d), 129.0 (d), 128.8 (d), 128.5 (s), 127.0 (d), 121.7 (d), 54.3 (d), 49.1 (t), 37.9 (t), 31.0 (q).

MS: *m*/*z* = 301.79 $[M + H]^+$.

Anal. Calcd for $C_{17}H_{17}ClN_2O$: C, 67.88; H, 5.70; N, 9.31. Found: C, 67.97; H, 5.62; N, 9.36.

### 4-(4-Benzyloxyphenyl)methyl-1-methyl-3-(4-chlorophenyl)imidazolidin-2-one (3cb)

Yellow oil; yield: 52 mg (51%); $R_f$ = 0.38 (hexane/EtOAc, 3:1).

IR: 1646 $cm^{-1}$.

$^1$H NMR (400 MHz, $CDCl_3$): δ = 7.59–7.28 (m, 9 H), 7.03 (d, *J* = 8.3 Hz, 2 H), 6.91 (d, *J* = 8.3 Hz, 2 H), 5.04 (s, 2 H), 4.43–4.28 (m, 1 H), 3.37 (t, *J* = 8.7 Hz, 1 H), 3.18 (dd, *J* = 8.5, 4.4 Hz, 1 H), 3.00 (d, *J* = 13.9 Hz, 1 H), 2.78 (s, 3 H), 2.65 (dd, *J* = 13.7, 9.2 Hz, 1 H).

$^{13}$C NMR (101 MHz, $CDCl_3$): δ = 158.0 (s), 157.9 (s), 137.5 (s), 136.9 (s), 130.2 (d), 129.1 (d), 129.0 (d), 128.6 (d), 128.4 (s), 128.0 (d), 127.5 (d), 121.8 (d), 115.2 (d), 70.1 (t), 54.4 (d), 49.1 (t), 37.00 (t), 31.0 (q).

MS: *m*/*z* = 407.85 $[M + H]^+$.

Anal. Calcd for $C_{24}H_{23}ClN_2O_2$: C, 70.84; H, 5.70; N, 6.88. Found: C, 70.95; H, 5.63; N, 6.79.

### 4-Benzyl-1-cyclohexyl-3-tosylimidazolidin-2-one (3da)

Yellow oil; yield: 64 mg (62%); $R_f$ = 0.44 (hexane/EtOAc, 3:1).

IR: 1658 $cm^{-1}$.

$^1$H NMR (400 MHz, $CDCl_3$): δ = 7.93 (d, *J* = 8.1 Hz, 2 H), 7.31–7.16 (m, 5 H), 7.10 (d, *J* = 7.2 Hz, 2 H), 4.54–4.37 (m, 1 H), 3.58–3.42 (m, 1 H), 3.31 (dd, *J* = 13.4, 3.4 Hz, 1 H), 3.17 (t, *J* = 9.0 Hz, 1 H), 2.95 (dd, *J* = 9.3, 2.6 Hz, 1 H), 2.70 (dd, *J* = 13.3, 9.4 Hz, 1 H), 2.35 (s, 3 H), 1.79–0.62 (m, 10 H).

$^{13}$C NMR (101 MHz, $CDCl_3$): δ = 152.9 (s), 144.5 (s), 136.7 (s), 135.7 (s), 129.6 (d), 129.5 (d), 128.8 (d), 128.2 (d), 127.1 (d), 55.0 (d), 51.4 (d), 42.1 (t), 40.4 (t), 30.0 (t), 29.7 (t), 25.3 (t), 25.2 (t), 21.7 (q).

MS: *m*/*z* = 413.26 $[M + H]^+$.

Anal. Calcd for $C_{23}H_{28}N_2O_3S$: C, 66.96; H, 6.84; N, 6.79. Found: C, 66.89; H, 6.92; N, 6.69.

### 4-(4-Benzyloxyphenyl)methyl-1-cyclohexyl-3-tosylimidazolidin-2-one (3db)

Yellow oil; yield: 92 mg (71%); $R_f$ = 0.36 (hexane/EtOAc, 2:1).

IR: 1692 $cm^{-1}$.

$^1$H NMR (400 MHz, $CDCl_3$): δ = 8.01 (d, *J* = 8.2 Hz, 2 H), 7.47–7.28 (m, 7 H), 7.11 (d, *J* = 8.4 Hz, 2 H), 6.94 (d, *J* = 8.4 Hz, 2 H), 5.06 (s, 2 H), 4.61–4.44 (m, 1 H), 3.69–3.52 (m, 1 H), 3.46–3.23 (m, 2 H), 3.04 (dd, *J* = 9.2, 2.6 Hz, 1 H), 2.75 (dd, *J* = 13.5, 9.3 Hz, 1 H), 2.44 (s, 3 H), 1.92–0.87 (m, 10 H).

$^{13}$C NMR (101 MHz, $CDCl_3$): δ = 158.0 (s), 152.9 (s), 144.4 (s), 137.0 (s), 136.8 (s), 130.6 (d), 129.5 (d), 128.6 (d), 128.3 (d), 128.1 (d), 128.0 (d), 127.9 (s), 127.4 (d), 115.2 (d), 70.1 (t), 55.1 (d), 51.4 (d), 42.2 (t), 39.5 (t), 30.1 (t), 29.7 (t), 25.3 (t), 25.2 (t), 21.7 (q).

MS: *m*/*z* = 519.67 $[M + H]^+$.

Anal. Calcd for $C_{30}H_{34}N_2O_4S$: C, 69.47; H, 6.61; N, 5.40. Found: C, 69.55; H, 6.69; N, 5.34.

### 4-Benzyl-1-cyclohexyl-3-(4-chlorophenyl)imidazolidin-2-one (3ea)

White solid; yield: 54 mg (58%); mp 76 °C, $R_f$ = 0.43 (hexane/EtOAc, 3:1).

IR: 1698 $cm^{-1}$.

$^1$H NMR (300 MHz, $CDCl_3$): δ = 7.55 (d, *J* = 8.7 Hz, 2 H), 7.42–7.23 (m, 5 H), 7.13 (d, *J* = 6.8 Hz, 2 H), 4.51–4.32 (m, 1 H), 3.81–3.62 (m, 1 H), 3.38 (t, *J* = 8.7 Hz, 1 H), 3.17 (dd, *J* = 8.8, 3.9 Hz, 1 H), 3.04 (dd, *J* = 14.0, 2.7 Hz, 1 H), 2.71 (dd, *J* = 13.8, 8.8 Hz, 1 H), 1.88–0.98 (m, 10 H).

$^{13}$C NMR (75 MHz, $CDCl_3$): δ = 156.7 (s), 137.7 (s), 136.2 (s), 129.2 (d), 129.0 (d), 128.7 (d), 128.0 (s), 126.9 (d), 121.3 (d), 54.3 (d), 51.2 (d), 42.1 (t), 37.6 (t), 30.2 (t), 29.9 (t), 25.5 (t), 25.5 (t).

MS: *m*/*z* = 369.45 $[M + H]^+$.

Anal. Calcd for $C_{22}H_{25}ClN_2O$: C, 71.63; H, 6.83; N, 7.59. Found: C, 71.58; H, 6.80; N, 7.66.

### Single-Crystal X-ray Diffraction Analysis

The data collection was carried out on a Bruker AXS Smart APEX 3-circle diffractometer with graphite-monochromated Mo Kα radiation (λ = 0.71073 Å) at a nominal power of the source of 50 kV × 30 mA. A 100% complete full sphere of reflections was measured up to a resolution of sinθ/λ = 0.45 $Å^{-1}$ by means of 5 ω scans of the reciprocal lattice. The Saint+[15] and SADABS[16] programs were employed to account for systematic errors, including absorption and beam anisotropy corrections. The compound crystallizes in a $P2_1/c$ centric lattice [*a* = 11.1136(19) Å, *b* = 12.867(2) Å, *c* = 29.058(5) Å, β = 94.558(11)°] as a 1:1 racemate, with two molecules with inverse handedness per asymmetric unit and a total of 8 molecules per cell. The molecular structure was solved through the charge flipping method[17] and least-squares refined within the Independent Atom Model approximation implemented in SHELX.[18] A total of 23311 individual structure factor amplitudes, corresponding to 3255 independent observations, entered the fitting, giving a final agreement factor *R*1(*F*) of 0.1064 for 1758 $F_o > 4\sigma(F_o)$ in conjunction with a goodness-of-fit of 1.040 and largest Fourier residuals of +0.40/–0.31 e/$Å^3$, both close to the chlorine heavy atom. See the Supporting Information for more details.

### 4-(4-Benzyloxyphenyl)methyl-1-cyclohexyl-3-(4-chlorophenyl)imidazolidin-2-one (3eb)

Pale-yellow oil; yield: 59 mg (50%); $R_f$ = 0.32 (hexane/EtOAc, 3:1).

IR: 1698 $cm^{-1}$.

$^1$H NMR (300 MHz, $CDCl_3$): δ = 7.60–7.51 (m, 2 H), 7.49–7.30 (m, 7 H), 7.10–7.00 (m, 2 H), 6.98–6.89 (m, 2 H), 5.07 (s, 2 H), 4.40 (ddd, *J* = 12.5, 8.6, 3.9 Hz, 1 H), 3.74 (tt, *J* = 12.1, 3.9 Hz, 1 H), 3.39 (t, *J* = 8.8 Hz, 1 H), 3.17 (dd, *J* = 8.9, 4.3 Hz, 1 H), 2.98 (dd, *J* = 13.9, 3.3 Hz, 1 H), 2.68 (dd, *J* = 13.9, 8.7 Hz, 1 H), 1.93–0.95 (m, 10 H).

$^{13}$C NMR (75 MHz, $CDCl_3$): δ = 157.9 (s), 156.7 (s), 137.8 (s), 137.0 (s), 130.3 (d), 129.0 (d), 128.6 (d), 128.5 (s), 128.0 (d), 127.4 (d), 121.3 (d), 115.1 (d), 70.1 (t), 54.4 (d), 51.2 (d), 42.1 (t), 36.8 (t), 30.3 (t), 29.9 (t), 25.5 (t), 25.5 (t).

MS: *m*/*z* = 475.56 $[M + H]^+$, 498.03 $[M + Na]^+$.

Anal. Calcd for $C_{29}H_{31}ClN_2O_2$: C, 73.33; H, 6.58; N, 5.90. Found: C, 73.46; H, 6.50; N, 5.84.

**4-benzyl-3-(4-chlorophenyl)-1-phenylimidazolidin-2-one (3fa)**

Colorless oil; yield: 34 mg (38%); $R_f$ = 0.31 (hexane/EtOAc, 4:1).

IR: 1690 $cm^{-1}$.

$^1$H NMR (300 MHz, $CDCl_3$): δ = 7.61–7.53 (m, 2 H), 7.51–7.44 (m, 2 H), 7.43–7.13 (m, 8 H), 7.11–7.02 (m, 2 H), 4.67–4.51 (m, 1 H), 3.90 (t, *J* = 9.0 Hz, 1 H), 3.67 (dd, *J* = 9.3, 4.8 Hz, 1 H), 3.18 (dd, *J* = 13.8, 3.4 Hz, 1 H), 2.77 (dd, *J* = 13.8, 9.4 Hz, 1 H).

$^{13}$C NMR (75 MHz, $CDCl_3$): δ = 154.8 (s), 139.7 (s), 136.8 (s), 135.7 (s), 129.4 (s), 129.1 (d), 128.9 (d), 128.8 (d), 127.2 (d), 123.2 (d), 122.7 (d), 118.2 (d), 53.9 (d), 47.0 (t), 38.3 (t).

MS: *m*/*z* = 363.17 $[M + H]^+$.

Anal. Calcd for $C_{22}H_{19}ClN_2O$: C, 72.82; H, 5.28; N, 7.72. Found: C, 72.76; H, 5.25; N, 7.79.

**4-(4-Benzyloxyphenyl)methyl-1-phenyl-3-(4-chlorophenyl)imidazolidin-2-one (3fb)**

Yellow oil; yield: 66 mg (56%); $R_f$ = 0.34 (hexane/EtOAc, 4:1).

IR: 1688 $cm^{-1}$.

$^1$H NMR (300 MHz, $CDCl_3$): δ = 7.62–7.28 (m, 13 H), 7.15–7.03 (m, 3 H), 6.97–6.84 (m, 2 H), 5.04 (s, 2 H), 4.62–4.46 (m, 1 H), 3.90 (t, *J* = 9.0 Hz, 1 H), 3.66 (dd, *J* = 9.2, 4.9 Hz, 1 H), 3.10 (dd, *J* = 13.9, 3.4 Hz, 1 H), 2.73 (dd, *J* = 13.9, 9.1 Hz, 1 H).

$^{13}$C NMR (75 MHz, $CDCl_3$): δ = 158.0 (s), 154.9 (s), 139.7 (s), 136.9 (s), 130.2 (d), 129.4 (s), 129.1 (d), 128.8 (d), 128.6 (d), 128.0 (d), 127.9 (s), 127.4 (d), 123.1 (d), 122.6 (d), 118.2 (d), 115.3 (d), 70.1 (t), 53.9 (d), 47.0 (t), 37.4 (t).

MS: *m*/*z* = 469.57 $[M + H]^+$.

Anal. Calcd for $C_{29}H_{25}ClN_2O_2$: C, 74.27; H, 5.37; N, 5.97. Found: C, 74.36; H, 5.31; N, 5.94.

**4-(4-Benzyloxyphenyl)methyl-1-phenyl-3-tosylimidazolidin-2-one (3gb)**

Yellow oil; yield: 71 mg (55%); $R_f$ = 0.36 (hexane/EtOAc, 3:1).

IR: 1695 $cm^{-1}$.

$^1$H NMR (400 MHz, $CDCl_3$): δ = 7.96 (d, *J* = 8.2 Hz, 2 H), 7.41–7.15 (m, 11 H), 7.08 (d, *J* = 8.4 Hz, 2 H), 7.00 (t, *J* = 7.1 Hz, 1 H), 6.86 (d, *J* = 8.5 Hz, 2 H), 4.97 (s, 2 H), 4.65–4.41 (m, 1 H), 3.72 (t, *J* = 9.1 Hz, 1 H), 3.50–3.37 (m, 2 H), 2.76 (dd, *J* = 13.4, 9.8 Hz, 1 H), 2.36 (s, 3 H).

$^{13}$C NMR (101 MHz, $CDCl_3$): δ = 158.1 (s), 151.4 (s), 144.9 (s), 138.4 (s), 136.9 (s), 136.3 (s), 130.6 (d), 129.6 (d), 128.9 (d), 128.6 (d), 128.4 (d), 128.0 (d), 127.7 (s), 127.5 (d), 124.3 (d), 118.8 (d), 115.3 (d), 70.1 (t), 54.5 (d), 47.2 (t), 40.0 (t), 21.7 (q).

MS: *m*/*z* = 513.43 $[M + H]^+$.

Anal. Calcd for $C_{30}H_{28}N_2O_4S$: C, 70.29; H, 5.51; N, 5.46. Found: C, 70.38; H, 5.43; N, 5.43.

**4-Benzyl-1-phenyl-3-(*p*-tolyl)imidazolidin-2-one (3ha)**

Colorless oil; yield: 50 mg (59%); $R_f$ = 0.38 (hexane/EtOAc, 4:1).

IR: 2921, 1705, 1404, 1292 $cm^{-1}$.

$^1$H NMR (300 MHz, $CDCl_3$): δ = 7.55–7.44 (m, 4 H), 7.37–7.13 (m, 9 H), 7.10–7.01 (m, 1 H), 4.64–4.51 (m, 1 H), 3.85 (t, *J* = 9.0 Hz, 1 H), 3.65 (dd, *J* = 9.2, 5.3 Hz, 1 H), 3.20 (dd, *J* = 13.7, 3.3 Hz, 1 H), 2.74 (dd, *J* = 13.7, 9.6 Hz, 1 H), 2.36 (d, *J* = 8.2 Hz, 3 H).

$^{13}$C NMR (75 MHz, $CDCl_3$): δ = 155.3 (s), 140.1 (s), 136.1 (s), 135.5 (s), 134.3 (s), 129.7 (d), 129.4 (d), 129.2 (d), 128.8 (d), 128.8 (d), 127.0 (d), 122.8 (d), 122.3 (d), 118.0 (d), 54.3 (d), 47.1 (t), 38.5 (t), 20.9 (q).

MS: *m*/*z* = 343.60 $[M + H]^+$.

Anal. Calcd for $C_{23}H_{22}N_2O$: C, 80.67; H, 6.48; N, 8.18. Found: C, 80.75; H, 6.54; N, 8.11.

**4-(4-Benzyloxyphenyl)methyl-1-phenyl-3-(*p*-tolyl)imidazolidin-2-one (3hb)**

Colorless oil; yield: 61 mg (54%); $R_f$ = 0.40 (hexane/EtOAc, 4:1).

IR: 1690 $cm^{-1}$.

$^1$H NMR (300 MHz, $CDCl_3$): δ = 7.57–7.19 (m, 13 H), 7.12–7.04 (m, 3 H), 6.97–6.88 (m, 2 H), 5.04 (s, 2 H), 4.59–4.45 (m, 1 H), 3.86 (t, *J* = 8.9 Hz, 1 H), 3.64 (dd, *J* = 9.2, 5.3 Hz, 1 H), 3.12 (dd, *J* = 13.8, 3.3 Hz, 1 H), 2.69 (dd, *J* = 13.8, 9.5 Hz, 1 H), 2.37 (s, 3 H).

$^{13}$C NMR (75 MHz, $CDCl_3$): δ = 157.9 (s), 155.3 (s), 140.1 (s), 136.9 (s), 135.5 (s), 134.2 (s), 130.2 (d), 129.7 (d), 128.7 (d), 128.6 (d), 128.3 (s), 128.0 (d), 127.4 (d), 122.7 (d), 122.2 (d), 118.0 (d), 115.2 (d), 70.1 (t), 54.3 (d), 47.1 (t), 37.5 (t), 20.9 (q).

MS: *m*/*z* = 449.48 $[M + H]^+$, 471.52 $[M + Na]^+$.

Anal. Calcd for $C_{30}H_{28}N_2O_2$: C, 80.33; H, 6.29; N, 6.25. Found: C, 80.26; H, 6.28; N, 6.28.

**4-Phenethyl-1-phenyl-3-tosylimidazolidin-2-one (6)**

Colorless oil; yield: 52 mg (49%); $R_f$ = 0.29 (hexane/EtOAc, 4:1).

IR: 1695 $cm^{-1}$.

$^1$H NMR (300 MHz, $CDCl_3$): δ = 8.00 (d, *J* = 8.3 Hz, 2 H), 7.50–7.08 (m, 12 H), 4.57–4.46 (m, 1 H), 4.03 (t, *J* = 9.1 Hz, 1 H), 3.56 (dd, *J* = 9.2, 3.4 Hz, 1 H), 2.77–2.64 (m, 2 H), 2.50–2.37 (m, 4 H), 2.28–2.15 (m, 1 H).

$^{13}$C NMR (75 MHz, $CDCl_3$): δ = 144.8 (s), 140.0 (s), 138.3 (s), 136.1 (s), 129.6 (d), 129.0 (d), 128.6 (d), 128.3 (d), 128.3 (d), 126.4 (d), 124.4 (d), 119.5 (s), 118.7 (d), 53.1 (d), 48.1 (t), 36.3 (t), 30.3 (t), 21.7 (q).

MS: *m*/*z* = 420.90 $[M + H]^+$.

Anal. Calcd for $C_{24}H_{24}N_2O_3S$: C, 68.55; H, 5.75; N, 6.66. Found: C, 68.63; H, 5.71; N, 6.60.

**4-Vinyl-1-phenyl-3-tosylimidazolidin-2-one (7)**

Colorless oil; yield: 10 mg (11%); $R_f$ = 0.19 (hexane/EtOAc, 4:1).

IR: 1689 $cm^{-1}$.

$^1$H NMR (500 MHz, $CDCl_3$): δ = 8.02–7.97 (m, 2 H), 7.48–7.43 (m, 2 H), 7.38–7.31 (m, 4 H), 7.14 (d, *J* = 7.4 Hz, 1 H), 5.95 (ddd, *J* = 17.0, 10.1, 8.0 Hz, 1 H), 5.53 (dd, *J* = 17.0 Hz, 1 H), 5.38 (d, *J* = 10.2 Hz, 1 H), 4.94 (td, *J* = 8.8, 3.4 Hz, 1 H), 4.15 (t, *J* = 9.1 Hz, 1 H), 3.58 (dd, *J* = 9.3, 3.4 Hz, 1 H), 2.45 (s, 3 H).

$^{13}$C NMR (126 MHz, $CDCl_3$): δ = 151.3 (s), 144.7 (s), 138.4 (s), 136.2 (s), 135.2 (d), 129.5 (d), 129.0 (d), 128.6 (d), 124.4 (d), 119.4 (t), 118.7 (d), 55.8 (d), 49.2 (t), 21.7 (q).

MS: *m*/*z* = 365.22 $[M + Na]^+$.

**1,4-Diphenyl-3-(*p*-tolyl)-1,3-diazepan-2-one (9)**

Colorless oil; yield: 51 mg (57%); $R_f$ = 0.37 (hexane/EtOAc, 3:1).

IR: 1694 $cm^{-1}$.

$^1$H NMR (300 MHz, $CDCl_3$): δ = 7.62–7.11 (m, 12 H), 7.05 (d, *J* = 8.4 Hz, 2 H), 4.93 (dd, *J* = 8.1, 6.2 Hz, 1 H), 3.82 (td, *J* = 7.3, 2.0 Hz, 2 H), 2.28 (s, 3 H), 2.24–2.02 (m, 2 H), 1.90–1.49 (m, 2 H).

$^{13}$C NMR (75 MHz, $CDCl_3$): δ = 154.4 (s), 141.7 (s), 141.1 (s), 136.2 (s), 132.5 (s), 130.4 (d), 129.3 (d), 128.7 (d), 128.6 (d), 128.3 (d), 128.2 (d), 126.9 (d), 119.5 (d), 63.4 (d), 48.3 (t), 37.1 (t), 26.0 (t), 20.7 (q).

MS: *m*/*z* = 379.81 $[M + Na]^+$.

Anal. Calcd $C_{24}H_{24}N_2O$: C, 80.86; H, 6.79; N, 7.86. Found: C, 81.07; H, 6.70; N, 7.79.

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## Supporting Information

Supporting information for this article is available online at https://doi.org/10.1055/s-0037-1611539.

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