Notes

N-Boc-*N*-(benzotriazol-1-ylmethyl)benzylamine as a 1,1-Dipole Equivalent in Stereoselective Synthesis of 4,5-Disubstituted Imidazolidin-2-ones

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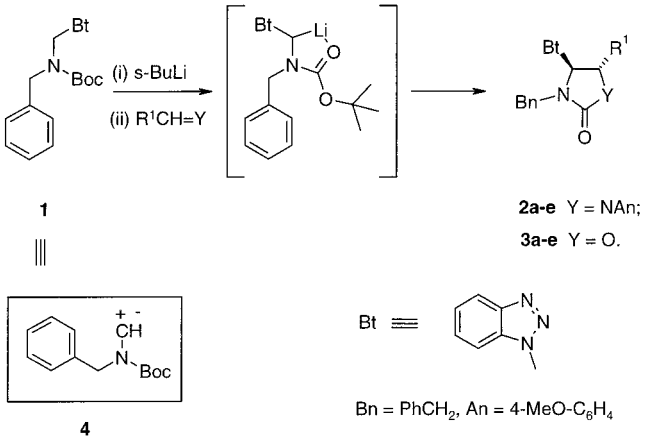
Introduction

The formation and reactions of dipole-stabilized carbanions adjacent to nitrogen atoms have been well documented.[1a−g] Benzyl or allyl activation can supplement dipole stabilization in the formation of a carbanion intermediate,[1a] but heteroaromatic ring activation sufficient to generate a carbanion between two nitrogen atoms, to the best of our knowledge, has not been well studied. Since benzotriazole activates an α-CH toward proton loss more than phenyl or vinyl,[2] *N*-Boc-*N*-(benzotriazol-1-ylmethyl)benzylamine (**1**) should be lithiated selectively at the carbon adjacent to the benzotriazole residue. Benzotriazole α to a heteroatom can act as a good leaving group to form a carbocation. We now report that *N*-Boc-*N*-(benzotriazol-1-ylmethyl)benzylamine (**1**) can behave as the 1,1-dipole equivalent **4** (cf. Scheme 1) for the stereoselective synthesis of 4,5-disubstituted imidazolidin-2-ones **5a**−**k** via benzotriazole intermediates of type **2**.

4,5-Disubstituted imidazolidin-2-ones are protected precursors of vicinal diamines. Vicinal diamine structural units are very important as structural fragments of biologically active natural products, in medicinal chemistry, and in the asymmetric synthesis as chiral ligands and chiral auxiliaries.[3a−c] Thus, it is still of considerable interest to explore novel synthetic routes for this type of compounds although a number of methods have been reported. Previous methods for the stereoselective generation of 1,2-diamines comprise the following: (i) transformations of diols, diazides,[4] aziridines,[5a,b] and α-bromo oxime ethers;[6] (ii) reduction of diimines derived from diketones or dialdehydes;[7a,b] (iii) reduction of 2-aminonitroalkanes;[8] (iv) conversion of enantiopure amino acids;[9] (v) addition of Grignard reagents to α-amino nitrones;[10] (vi) addition of α-nitrogen carbanions to imines;[1d,e] (vii) aza-Michael additions to nitroalkenes;[11] (viii) nitro-Mannich reactions;[12] (ix) addition of organozinc reagents to 1,4-diazadienes.[13] Although reaction of stabilized α-nitrogen carbanions with imines affords *trans*-4,5-disubstituted imidazolidin-2-ones in good yields and with good stereoselectivities, the examples published[1d,e] are limited to the preparation of 4-aryl and 4,5-diaryl derivatives. The present method using *N*-Boc-*N*-(benzotriazol-1-ylmethyl)benzylamine as a 1,1-dipole synthon enables the efficient introduction of a variety of substituents at the 4 position of 5-aryl- and 5-heteroaryl-4,5-imidazolidin-2-ones stereospecifically and in good yields.

Scheme 1



Results and Discussion

Preparation of Benzotriazolylimidazolidinone Derivatives 2a−e. Starting material **1** is easily synthesized by a two-step procedure. Protection of benzylamine with $(Boc)_2O$ afforded *N*-Boc-benzylamine,[1c] which re-

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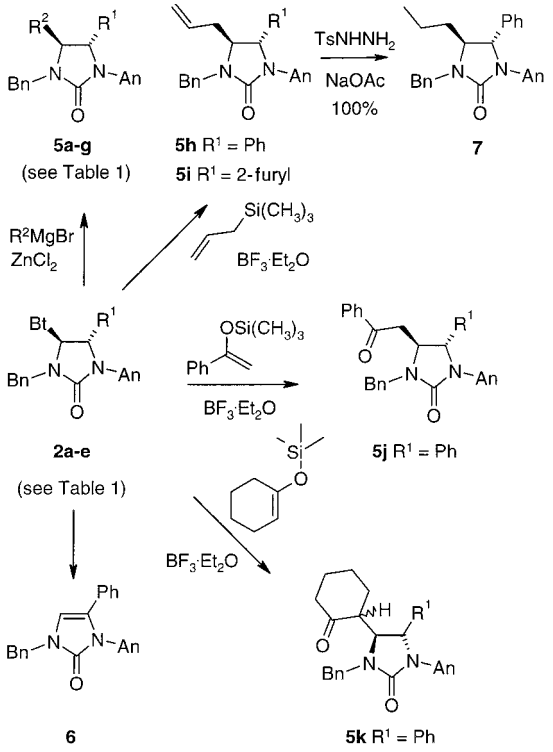
Table 1. Preparation of Benzotriazole Derivatives 2a−e and 3a−e

entry	R^1	product	yield (%)	product	yield (%)
1	Ph	**2a**	82	**3a**	82
2	4-Me-C_6H_4	**2b**	74	**3b**	76
3	4-MeO-C_6H_4	**2c**	80	**3c**	72
4	4-F-C_6H_4	**2d**	61	**3d**	70
5	2-furyl	**2e**	70	**3e**	66

Scheme 2



acted further (without purification) with formaldehyde and benzotriazole in the presence of a catalytic amount of TsOH using a Dean−Stark apparatus to remove the water azeotropically to give *N*-Boc-*N*-(benzotriazol-1-ylmethyl)benzylamine (**1**) in 70% overall yield. Treatment of **1** with 1 equiv of *s*-BuLi in THF at −78 °C for 0.5 h, followed by the addition of imines as electrophiles, produced the benzotriazolyl-substituted imidazolidinones **2a**−**e** (Scheme 1). When imines derived from aromatic aldehydes (electron-donating substituent, entry 3; electron-withdrawing substituent, entry 4) or a heteroaromatic aldehyde (entry 5) were used, the corresponding benzotriazole derivatives **2** were obtained in good yields (Table 1) and stereospecifically (trans/cis >99:1). According to ^{1}H NMR and ^{13}C NMR spectra, only one isomer was observed and isolated from the reaction mixture in each case.

Reactions of Synthons 2 with Diverse Zinc Reagents. Treatment of **2a**−**e** with organozinc reagents gave compounds **5a**−**g** (Scheme 2) in good yields and stereospecifically (trans/cis >99:1 according to NMR of the reaction mixtures) (Table 2). Arylzinc reagents (entries 1−5**)**, an alkynylzinc reagent (entry 6), and an alkenylzinc reagent (entry 7) all afforded the corresponding desired products. Various 5-substituted imidazolidinones **2a**−**e** can be used for these substitution reactions (entries 1−5). However, alkylzinc reagents produced no desired products but *β*-H elimination occurred to give compounds such as **6** together with other products still under investigation.

Table 2. Substitution of the Benzotriazole Moiety in 2a−e by Carbon Nucleophiles

entry	starting material	R^1	R^2	product	yield (%)
1	**2a**	Ph	Ph	**5a**	68
2	**2b**	4-Me-C_6H_4	Ph	**5b**	50
3	**2c**	4-MeO-C_6H_4	Ph	**5c**	65
4	**2d**	4-F-C_6H_4	Ph	**5d**	60
5	**2e**	2-furyl	Ph	**5e**	66
6	**2a**	Ph	Ph-C≡C	**5f**	74
7	**2a**	Ph	CH_2=CH	**5g**	68
8	**2a**	Ph	CH_2=CH-CH_2	**5h**	67
9	**2e**	2-furyl	CH_2=CH-CH_2	**5i**	40
10	**2a**	Ph	PhC(=O)CH_2	**5j**	64
11	**2a**	Ph	*c*-C_4H_8CHCO	**5k**	70

Reactions of Synthons 2 with Allyltrimethylsilane and Vinyloxysilanes. Treatment of **2a** with $SnCl_4$ at −78 °C for 30 min followed by the addition of allyltrimethylsilane afforded *trans*-4,5-disubstituted imidazolidinone (**5h**) (20%) along with the *β*-H elimination product **6** (50%) (Scheme 2). When BF_3•Et_2O was used, exclusively **5h** was produced in 67% yield (Table 2) and with high trans selectivity (trans/cis >99:1 according to NMR of the reaction mixture). Under the same reaction conditions, **5i** was prepared in 40% yield.

Reactions of trimethyl[(1-phenylvinyl)oxy]silane or (1-cyclohexen-1-yloxy)trimethylsilane with **2a** gave products **5j**−**k** in good yields and specifically (trans/cis >99:1 according to NMR of the reaction mixtures). In the case of **5k**, no stereochemical control at the carbon α to the carbonyl group in the cyclohexanone substituent was observed. The H4 was shown as a doublet in ^{1}H NMR and the coupling constant between H4 and H5 was 4.0 Hz in **5k**, indicating that H4 and H5 were in trans configuration.

Compound **7** was easily obtained from the reduction of **5h**[14] (Scheme 2), which made it possible for the indirect stereospecific introduction of an alkyl group in the 4 position of 4,5-disubstituted imidazolidinones.

The trans configuration for compounds **2a**−**e**, **5a**−**k**, and **7** was assigned from the coupling constant $J_{4,5}$ between H4 and H5 in the heterocyclic five-membered rings based on literature reports[15,16] and X-ray crystallography. It is generally accepted that the trans coupling constant is smaller than that of the corresponding cis coupling in a five-membered heterocyclic ring system. When a vicinal coupling constant is smaller than 4.0 Hz in a five-membered heterocyclic ring system, the geometry is assigned as trans.[15] The coupling constants in 4,5-disubstituted imidazolidin-2-ones **2a**−**e** were around 2.5 Hz, so these compounds were assigned the trans configuration. The full structure and stereochemistry of **2a** was further confirmed by a single-crystal X-ray structure determination.[17] The coupling constant $J_{4,5}$ (around 6.0 Hz) between H4 and H5 in **5a** is more ambiguous in terms of stereochemical assignment but is consistent with similar compounds which have trans configurations.[1d−e] The structure of **5a** was unambiguously determined by X-ray crystallography,[17] which confirmed the trans ster-

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(17) See the Supporting Information for X-ray crystal structures and data of **2a** and **5a**.

eochemistry. Similarly, the other compounds were also assigned as having trans geometry.

The high trans selectivity for **2a**−**e** probably derives from the formation of the products under thermodynamic control; however, products **5a**−**k** are all formed under kinetic control indicating that both thermodynamic and kinetic control highly favor trans products. In the presence of a Lewis acid (such as in situ generated $MgBr_2$ or BF_3•Et_2O), the benzotriazole moiety in **2a**−**e** can be removed to form acyliminium salts, which are attacked by nucleophiles exclusively from the opposite face of the R^1 group affording trans products. With basic nucleophiles (such as alkyl in contrast to aryl zinc reagents) or a strong Lewis acid (such as $SnCl_4$ relative to BF_3•Et_2O[18]), β-H elimination occurs to give products such as **6**.

Under similar reaction conditions as for imines, aldehydes were used as electrophiles for lithiated **1** to give analogous oxazolidinones **3a**−**e** (Scheme 1). All the reactions produced the trans isomers selectively in good yields when arylaldehydes were used (Table 1). The trans conformation assignment is based on the coupling constant ($J_{4,5}$ = 2.5 Hz), which is also consistent with the literature report.[16]

Attempts to substitute the benzotriazole residue in oxazolidinones **3a**−**e** failed. When organozinc reagents, organocerium reagents, organocopper reagents, Grignard reagents and allyltrimethylsilanes were used as nucleophiles, either no reactions occurred or only elimination products were isolated. Probably in these 4,5-disubstituted oxazolidin-2-ones **3a**−**e**, the single nitrogen atom cannot assist the removal of the benzotriazole moiety due to its relatively electron deficient character compared to that of the analogous nitrogen atom in 4,5-disubstituted imidazolidin-2-ones **2**.

Conclusion

A general method using *N*-Boc-*N*-(benzotriazol-1-ylmethyl)benzylamine as a 1,1-dipole equivalent to synthesize various trans-4,5-disubstituted imidazolidinones was reported. A number of trans-4,5-disubstituted imidazolidinones were obtained in good yields and with excellent stereoselectivity. Imidazolidin-2-ones can be converted directly into the corresponding secondary diamines under basic conditions (NaOH) without affecting the two N-protecting groups.[19a,b] Alternatively, removal of one or both of the N-protecting groups[1d,20a] followed by hydrolysis under acidic conditions (HCl or HBr) gives the corresponding primary diamines.[20b−d] Although benzotriazole derivatives **3a**−**e** of 4,5-disubstituted oxazolidin-2-ones could be obtained, attempted displacement of the benzotriazole moiety was not successful.

Experimental Section

General Comments. Melting points were determined on a hot-stage apparatus and are uncorrected. ^{1}H (300 MHz) and ^{13}C (75 MHz) NMR spectra were recorded in $CDCl_3$ with TMS as the internal reference. THF was distilled from sodium/benzophenone and CH_2Cl_2 was distilled from CaH_2 under nitrogen immediately prior to use. All reactions with air-sensitive compounds were carried out under argon atmosphere.

Synthesis of *N*-Boc-*N*-(benzotriazol-1-ylmethyl)benzylamine (1). Benzylamine (8.56 g, 0.08 mol) and triethylamine (11.7 mL) were mixed in methylene chloride (300 mL) at 0 °C, and then di-*tert*-butyl dicarbonate (17.44 g, 0.08 mol) in methylene chloride (100 mL) was added. The solution was stirred overnight, and methylene chloride was removed under reduced pressure. The crude product obtained can be used in the next step without purification.

The crude product, prepared as shown above, benzotriazole (9.52 g, 0.08 mol), paraformaldehyde (2.4 g, 0.08 mol) and a catalytic amount of *p*-toluenesulfonic acid were mixed in toluene (400 mL) and refluxed overnight with a Dean−Stark trap to remove the water formed. After toluene was removed under reduced pressure, the crude product was subjected directly to silica gel column chromatography, and product **1** was obtained as white prisms (from methylene chloride and hexanes): mp 126−127 °C; yield 70%; ^{1}H NMR δ 1.50 (s, 9H), 4.43 (s, 2H), 6.10 (s, 2H), 7.26−7.41 (m, 7H), 7.51 (t, J = 7.6 Hz, 1H), 8.05 (d, J = 8.1 Hz, 1H); ^{13}C NMR δ 28.2, 48.7, 57.3, 81.6, 111.0, 119.6, 124.2, 127.6, 127.8, 128.6, 132.5, 136.7, 146.2, 155.2. Anal. Calcd for $C_{19}H_{22}N_4O_2$: C, 67.44; H, 6.55; N, 16.56. Found: C, 67.26; H, 6.70; N, 16.60.

General Procedure for the Synthesis of Compounds 2a−e and 3a−e. At −78 °C, *s*-BuLi (2 mmol, 1.3 M in hexane) was added slowly to a solution of *N*-Boc-*N*-(benzotriazol-1-ylmethyl)benzylamine (**1**) (2 mmol) in THF (25 mL), and the reaction mixture was stirred for 0.5 h at this temperature. Then an imine or an aldehyde (2 mmol) in THF (5 mL) was added slowly, and the reaction solution was further stirred for 4 h. Saturated NH_4Cl aqueous solution (20 mL) was added to quench the reaction. Ethyl acetate (50 mL) was added, and the organic layer was separated. The aqueous layer was extracted with ethyl acetate (2 × 25 mL). The combined organic layer was washed with brine, dried over Na_2SO_4, and evaporated under reduced pressure. The residue was subjected to silica gel chromatography giving the pure products **2a**−**e** and **3a**−**e**, respectively.

***trans*-4-(1*H*-1,2,3-Benzotriazol-1-yl)-3-benzyl-5-phenyl-1-(4-methoxyphenyl)imidazolidin-2-one (2a):** white needles (from methylene chloride and hexanes); mp 155−156 °C; yield 82%; ^{1}H NMR δ 3.73 (s, 3H), 3.84 (d, J = 15.1 Hz, 1H), 4.86 (d, J = 15.1 Hz, 1H), 5.29 (d, J = 2.1 Hz, 1H), 6.20 (d, J = 2.1 Hz, 1H), 6.80 (d, J = 9.0 Hz, 2H), 7.08−7.19 (m, 7H), 7.30−7.40 (m, 3H), 7.43−7.45 (m, 5H), 8.08 (d, J = 8.6 Hz, 1H); ^{13}C NMR δ 45.4, 55.3, 64.5, 75.5, 109.7, 114.3, 116.3, 120.5, 122.3, 124.6, 125.9, 127.8, 128.2, 128.4, 128.5, 129.1, 129.4, 130.7, 135.1, 137.7, 146.9, 156.2, 156.4. Anal. Calcd for $C_{29}H_{25}N_5O_2$: C, 73.25; H, 5.30; N, 14.73. Found: C, 73.24; H, 5.46; N, 14.69.

Crystal data for 2a: $C_{29}H_{25}N_5O_2$, MW 475.54, orthorhombic, $P2_12_12_1$, a = 8.419(5) Å, b = 15.655(9) Å, c = 18.395(11) Å, V = 2420(2) Å^3, Z = 4, T = −105 °C, $F(000)$ = 1000, μ (Mo Kα) = 0.085 mm^{-1}, D_{calcd} = 1.305 g·cm^{-3}, $2\theta_{max}$ 53° (CCD area detector, 99.4% completeness), wR(F^2) = 0.071 (all 4851 data), R = 0.037 (3510 data with $I > 2\sigma I$).

***trans*-4-(1*H*-1,2,3-Benzotriazol-1-yl)-3-benzyl-5-phenyl-1,3-oxazolan-2-one (3a):** white prisms (from methylene chloride and hexanes); mp 110−112 °C; yield 82%; ^{1}H NMR δ 3.86 (d, J = 15.0 Hz, 1H), 4.70 (d, J = 15.0 Hz, 1H), 5.71 (d, J = 2.8 Hz, 1H), 6.37 (d, J = 2.8 Hz, 1H), 7.03−7.06 (m, 2H), 7.11−7.15 (m, 2H), 7.24−7.27 (m, 2H), 7.35−7.53 (m, 7H), 8.07 (d, J = 8.4 Hz, 1H); ^{13}C NMR δ 46.1, 76.2, 78.9, 109.3, 120.5, 124.8, 125.0, 128.1, 128.3, 128.6, 128.7, 129.2, 129.5, 130.4, 133.6, 136.0, 146.8, 156.0; HRMS calcd for $C_{22}H_{19}N_4O_2$ 371.1508 (M + 1), found 371.1514 (M + 1).

General Procedure for the Synthesis of Compounds 5a−g and 6. Grignard reagent R^2MgBr (2 mmol, 1.0 M in THF) was added to $ZnCl_2$ (2 mmol, 1.0 M in diethyl ether) in THF (10 mL) at 0 °C, and the resulting mixture was further stirred at room temperature for 20 min. Then **2a**−**e** (0.4 mmol) in THF (3 mL) was added, and the reaction solution was heated under reflux for 12 h. After the solution cooled to room temperature, saturated NH_4Cl aqueous solution (20 mL) was added and the resulting solution was extracted with ethyl acetate (3 × 30 mL). The combined organic layers were washed with brine, dried over

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Na_2SO_4, and evaporated under reduced pressure. The residue was then subjected to silica gel chromatography, and the pure products **5a**−**g** and **6** were obtained, respectively.

***trans*-1-Benzyl-3-(4-methoxyphenyl)-4,5-diphenylimidazolidin-2-one (5a):** white prisms (from methylene chloride and hexanes); mp 113−114 °C; yield 68%; ^{1}H NMR δ 3.69 (s, 3H), 3.70 (d, J = 14.9 Hz, 1H), 4.14 (d, J = 6.6 Hz, 1H), 4.86 (d, J = 6.6 Hz, 1H), 5.07 (d, J = 15.0 Hz, 1H), 6.75 (d, J = 9.0 Hz, 2H), 7.06−7.38 (m, 17H); ^{13}C NMR δ 45.6, 55.3, 65.7, 67.7, 113.8, 122.6, 126.4, 127.3, 127.4, 128.1, 128.5, 128.6, 128.8, 129.0, 132.2, 136.4, 138.3, 139.2, 155.8, 158.4. Anal. Calcd for $C_{29}H_{26}N_2O_2$: C, 80.16; H, 6.03; N, 6.45. Found: C, 79.95; H, 6.18; N, 6.44.

Crystal data for 5a: $C_{29}H_{26}N_2O_2$, MW 434.52, orthorhombic, *Pbca*, a = 10.311(3) Å, b = 10.693(3) Å, c = 41.759(11) Å, V = 4602(2) Å^3, Z = 8, T = −105 °C, F(000) = 1840, μ (Mo Kα) = 0.079 mm^{-1}, D_{calcd} = 1.254 g·cm^{-3}, $2\theta_{max}$ 53° (CCD area detector, 99.2% completeness), wR(F^2) = 0.115 (all 4700 data), R = 0.044 (3347 data with $I > 2\sigma I$).

***trans*-1-Benzyl-3-(4-methoxyphenyl)-4-phenyl-5-(2-phenylethynyl)imidazolidin-2-one (5f):** yellow oil; yield 74%; ^{1}H NMR δ 3.71 (s, 3H), 4.14 (d, J = 6.6 Hz, 1H), 4.30 (d, J = 15.0 Hz, 1H), 5.04 (d, J = 15.0 Hz, 1H), 6.77 (d, J = 6.6 Hz, 1H), 6.76−6.79 (m, 2H), 7.41−7.45 (m, 17H); ^{13}C NMR δ 46.2, 54.1, 55.3, 64.4, 84.3, 86.9, 113.9, 121.8, 122.9, 126.5, 127.5, 128.4, 128.5, 128.6, 128.9, 129.0, 131.8, 136.4, 138.4, 147.2, 156.1, 157.6. Anal. Calcd for $C_{31}H_{26}N_2O_2$: N, 6.11. Found: N, 6.12.

***trans*-1-Benzyl-3-(4-methoxyphenyl)-4-phenyl-5-vinylimidazolidin-2-one (5g):** white prisms (from methylene chloride and hexanes); mp 107−108 °C; yield 68%; ^{1}H NMR δ 3.64 (dd, J = 7.0, 8.6 Hz, 1H), 3.70 (s, 3H), 4.05 (d, J = 15.1 Hz, 1H), 4.73 (d, J = 7.0 Hz, 1H), 4.91 (d, J = 15.1 Hz, 1H), 5.06 (d, J = 17.0 Hz, 1H), 5.30 (d, J = 10.7 Hz, 1H), 5.82 (ddd, J = 8.6, 10.7, 17.0 Hz, 1H), 6.75 (m, 2H), 7.16−7.29 (m, 12H); ^{13}C NMR δ 45.7, 55.3, 64.9, 65.4, 113.8, 120.8, 122.6, 126.6, 127.3, 128.0, 128.4, 128.5, 128.7, 132.2, 135.1, 136.9, 138.8, 155.8, 158.2. Anal. Calcd for $C_{25}H_{24}N_2O_2$: C, 78.10; H, 6.29; N, 7.29. Found: C, 77.59; H, 6.60; N, 7.21.

1-Benzyl-3-(4-methoxyphenyl)-4-phenyl-1,3-dihydro-2*H*-imidazol-2-one (6): white needles (from methylene chloride and hexanes); mp 95−96 °C; yield 50%; ^{1}H NMR δ 3.79 (s, 3H), 4.90 (s, 2H), 6.32 (s, 1H), 6.86 (d, J = 8.9 Hz, 2H), 6.99−7.02 (m, 2H), 7.13−7.17 (m, 5H), 7.34−7.38 (m, 5H); ^{13}C NMR δ 47.3, 55.4, 108.8, 114.2, 124.2, 126.9, 127.2, 127.9, 128.2, 128.3, 128.4, 128.8, 129.4, 136.7, 153.4, 158.4; HRMS calcd for $C_{23}H_{21}N_2O_2$ 357.1603 (M + 1), found 357.1601 (M + 1).

General Procedure for the Synthesis of Compounds 5h−k. BF_3•Et_2O (2 mmol) in CH_2Cl_2 (2 mL) was added to **2a** or **2e** (0.4 mmol) in CH_2Cl_2 (10 mL) at −78 °C. After the reaction mixture was stirred for 30 min, allyltrimethylsilane or a vinyloxytrimethylsilane (2 mmol) was added, and the reaction mixture was further stirred for 4 h at this temperature. Then the reaction solution was warmed to room temperature and stirred overnight. Saturated NH_4Cl aqueous solution (20 mL) was added and extracted by CH_2Cl_2 (3 × 15 mL). Combined organic layers were washed with brine, dried over Na_2SO_4, and evaporated under reduced pressure. The residue was then subjected to silica gel chromatography affording the pure products **5h**−**k**, respectively.

***trans*-4-Allyl-3-benzyl-1-(4-methoxyphenyl)-5-phenylimidazolidin-2-one (5h):** white needles (from methylene chloride and hexanes); mp 74−75 °C; yield 67%; ^{1}H NMR δ 2.39−2.43 (m, 2H), 3.32−3.37 (m, 1H), 3.71 (s, 3H), 4.15 (d, J = 15.3 Hz, 1H), 4.76 (d, J = 5.1 Hz, 1H), 4.98 (d, J = 15.3 Hz, 1H), 5.20−5.26 (m, 2H), 5.66−5.75 (m, 1H), 6.73−6.78 (m, 2H), 7.17−7.33 (m, 12H); ^{13}C NMR δ 35.8, 45.6, 55.4, 60.4, 62.7, 113.9, 119.6, 122.1, 126.4, 127.4, 127.9, 128.0, 128.6, 128.9, 132.3, 136.9, 140.3, 155.6, 158.0; HRMS Calcd for $C_{26}H_{27}N_2O_2$ 399.2073 (M + 1), found 399.2071 (M + 1).

***trans*-1-Benzyl-5-(2-oxo-2-phenylethyl)-3-(4-methoxyphenyl)-4-phenylimidazolidin-2-one (5j):** white needles (from methylene chloride and hexanes); mp 118−119 °C; yield 63%; ^{1}H NMR δ 3.21 (dd, J = 8.5, 17.4 Hz, 1H), 3.36 (dd, J = 4.2, 17.4 Hz, 1H), 3.70 (s, 3H), 3.94−3.99 (m, 1H), 4.31 (d, J = 15.4 Hz, 1H), 4.79 (d, J = 15.4 Hz, 1H), 4.85 (d, J = 3.1 Hz), 6.74 (d, J = 9.1 Hz, 2H), 7.20−7.43 (m, 14H), 7.55 (t, J = 7.3 Hz, 1H), 7.78 (d, J = 7.3 Hz, 2H); ^{13}C NMR δ 40.9, 45.9, 55.3, 58.2, 64.3, 113.9, 121.2, 126.3, 127.4, 127.9, 128.0, 128.5, 128.6, 128.8, 132.5, 133.6, 136.2, 137.1, 139.7, 155.4, 157.4, 197.4. Anal. Calcd for $C_{31}H_{28}N_2O_3$: C, 78.13; H, 5.92; N, 5.88. Found: C, 77.73; H, 6.04; N, 5.81.

Synthesis of *trans*-1-Benzyl-3-(4-methoxyphenyl)-4-phenyl-5-propylimidazolidin-2-one (7). Compound **5h** (0.13 mmol) and *p*-toluenesulfonohydrazide (1.3 mmol) were dissolved in DME (5 mL) and heated until refluxed, NaOAc aqueous solution (2.2 mmol, 5 mL) was added within 30 min, and the reaction mixture was further heated under reflux for 1 h. The resulting reaction mixture was cooled to room temperature, and water (20 mL) was added. The solution was extracted with ethyl acetate (3 × 20 mL), dried over Na_2SO_4, and evaporated under reduced pressure. The residue was then subjected to silica gel chromatography, and **7** was obtained as white prisms (from methylene chloride and hexanes): mp 129−130 °C; yield 100%; ^{1}H NMR δ 0.87 (t, J = 7.3 Hz, 3H), 1.21−1.41 (m, 2H), 1.58−1.64 (m, 2H), 3.25−3.28 (m, 1H), 3.71 (s, 3H), 4.10 (d, J = 15.4 Hz, 1H), 4.69 (d, J = 4.9 Hz, 1H), 4.95 (d, J = 15.4 Hz, 1H), 6.74 (m, J = 2.2 Hz, 2H), 7.17−7.32 (m, 12H); ^{13}C NMR δ 14.1, 17.1, 33.7, 45.6, 55.3, 61.0, 63.7, 113.9, 122.0, 126.3, 127.4, 127.9, 128.0, 128.5, 128.9, 132.6, 137.1, 140.9, 155.6, 158.0. Anal. Calcd for $C_{26}H_{28}N_2O_2$: N, 6.99. Found: N, 6.65.

Supporting Information Available: Characterization data for compounds **2b**−**e**, **3b**−**e**, and **5b**−**e**,**i**,**k** and X-ray crystal structures for **2a** and **5a**. This material is available free of charge via the Internet at http://pubs.acs.org.

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