(Supporting Information)

Palladium-Catalyzed Addition of Nitrogen Pronucleophiles to Alkylidenecyclopropanes

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General experimental methods

All reactions were performed under argon atmosphere. All alkylidenecyclopropanes were synthesized according to the procedure reported by Utimoto et al.¹ Nitrogen pronucleophiles, palladium catalysts, phosphine ligands, and organic solvents were purchased. All reagents were used as obtained commercially. Column chromatography was performed with silica-gel. All yields given refer to as isolated yields. NMR spectra were recorded on a 400 MHz spectrometer. IR spectra were recorded on a FT-IR spectrometer. HRMS experiments were performed on a high resolution magnetic sector mass spectrometer.

1. Utimoto, K.; Tamaru, M.; Sisido, K. *Tetrahedron* **1973**, 29, 1169

Representative Procedure for the Addition of Cyclic Amides 16a to the Alkylidenecyclopropane 7c. To a screw-capped Wheaton microreactor, containing Pd(PPh₃)₄ (17.3 mg, 0.015 mmol) and methylenecyclopropane (61.8 mg, 0.3 mmol), was added the cyclic amide (0.045 mL, 0.6 mmol) under Ar atmosphere and the mixture was stirred at 120 °C for the specified time. The reaction progress was monitored with GC-Mass. After completion, the turbid reaction mixture was filtered through a short silica column with ethyl acetate as eluent. Separation by passing through a florisil column (hexane as the eluent) and purification by medium-pressure liquid column chromatography (silica gel) using hexane as the eluent afforded the allylation product **17a** in 91% yield.

Representative Procedure for the Intramolecular Hydroamination of Alkylidenecyclopropane 19a. To a mixture of Pd(PPh₃)₄ (28.9 mg, 0.025 mmol) and P(O)*n*-Bu₃ (8.4 mg, 0.075 mmol) was added methylenecyclopropane **19a** (110.5 mg, 0.5 mmol), under Ar atmosphere in a screw-capped Wheaton microreactor. After heating at 120 °C for 2 days, the mixture was filtered through a short florisil column using ethyl acetate as an eluent. Separation by passing through a florisil column (hexane/ethyl acetate as the eluent) and purification by

medium-pressure liquid column chromatography (RP-18) using EtOAc as the eluent afforded adducts **20a** in 86% yield.

Representative Procedure for the Intramolecular Hydroamination of Alkylidenecyclopropane

24. To a mixture of Pd(PPh₃)₄ (28.9 mg, 0.025 mmol) in CH₃CN was added methylenecyclopropane **24** (2.3 mg, 1.0 mmol), under Ar atmosphere in a screw-capped Wheaton microreactor. After heating at 120 °C for 2 days, the mixture was filtered through a short florisil column using ethyl acetate as an eluent. Separation by passing through a florisil column (hexane/ethyl acetate as the eluent) and purification by medium-pressure liquid column chromatography (RP-18) using EtOAc as the eluent afforded adducts **25** in 57% yield.

1-(2-Methylene-3, 3-diphenylpropyl)-pyrrolidin-2-one (17a). IR (neat) 3352, 2939, 1958, 1888, 1680, 1597, 1490, 1417 cm^{-1} . ^1H NMR (CDCl_3 , 400 MHz) δ 1.79 (t, $J = 5.0$ Hz, 2H), 2.26 (t, $J = 8.2$ Hz, 2H), 3.19 (t, $J = 5.4$ Hz, 2H), 3.90 (s, 2H), 4.73 (d, $J = 16.4$ Hz, 2H), 5.15 (s, 1H), 7.12-7.30 (m, 10H). ^{13}C NMR (CDCl_3 , 100 MHz) δ 44.3, 49.9, 55.6, 61.6, 116.3, 126.5, 128.3, 128.88, 128.89, 141.1, 146.2, 158.2. Anal. Calcd for $\text{C}_{20}\text{H}_{21}\text{NO}$ (291.39): C, 82.44; H, 7.26; N, 4.81; O, 5.49. Found: C, 82.38; H, 7.23; N, 4.79. HRMS (EI) Calcd for $\text{C}_{20}\text{H}_{21}\text{NO}$: m/z 291.1623. Found: m/z 291.1620.

1-(2-Nonyl-allyl)-pyrrolidin-2-one (17b). IR (neat) 3367, 3075, 2925, 2854, 1687, 1654, 1594, 1492, 1460, 1422 cm^{-1} . ^1H NMR (CDCl_3 , 400 MHz) δ 0.87 (t, $J = 6.8$ Hz, 3H), 1.26-1.31 (m, 12H), 1.44 (t, $J = 7.2$ Hz, 2H), 1.92-2.04 (m, 4H), 2.42 (t, $J = 8.4$ Hz, 2H), 3.29 (t, $J = 5.4$ Hz, 2H), 3.83 (s, 2H), 4.82 (d, $J = 16.7$ Hz, 2H). ^{13}C NMR (CDCl_3 , 100 MHz) δ 14.2, 17.9, 22.7, 27.5, 29.37, 29.40, 29.5, 29.6, 31.1, 31.9, 33.6, 46.8, 47.6, 111.5, 144.2. Anal. Calcd for $\text{C}_{16}\text{H}_{29}\text{NO}$ (251.41): C, 76.44; H, 11.63; N, 5.57. Found: C, 76.24; H, 11.57; N, 5.67. HRMS (EI) Calcd for $\text{C}_{16}\text{H}_{29}\text{NO}$: m/z 251.2249. Found: m/z 251.2248.

1-(2-Octyl-allyl)-pyrrolidin-2-one (17c). IR (neat) 3370, 2925, 2855, 1765, 1692, 1494, 1462, 1423 cm^{-1} . ^1H NMR (CDCl_3 , 400 MHz) δ 0.87 (t, $J = 6.8$ Hz, 3H), 1.19-1.31 (m, 10H), 1.44 (t, $J = 6.8$ Hz, 2H), 1.92-2.07 (m, 4H), 2.42 (t, $J = 8.4$ Hz, 2H), 3.29 (t, $J = 5.4$ Hz, 2H), 3.83 (s, 2H), 4.82 (d, $J = 16.8$ Hz, 2H). ^{13}C NMR (CDCl_3 , 100 MHz) δ 14.2, 17.9, 22.7, 27.5, 29.3, 29.4, 29.5, 31.1, 31.9, 33.6, 46.8, 47.6, 111.5, 144.2, 174.8. Anal. Calcd for $\text{C}_{15}\text{H}_{27}\text{NO}$ (237.38): C, 75.90; H, 11.46; N, 5.90. Found: C, 75.86; H, 11.43; N, 5.87. HRMS (EI) Calcd for $\text{C}_{15}\text{H}_{27}\text{NO}$: m/z 237.2093. Found: m/z 237.2089.

1-(3-Hexyl-2-methylenenonyl)-pyrrolidin-2-one (17d). IR (neat) 3369, 3074, 2924, 2855, 1685,

1649, 1594, 1492, 1450 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ 0.87 (t, $J = 7.4$ Hz, 3H), 1.35 (m, 19H), 1.42-1.46 (m, 2H), 1.53 (s, 4H), 2.02 (m, 3H), 2.42 (t, $J = 3.2$ Hz, 2H), 3.32 (t, $J = 12$ Hz, 2H), 3.78 (s, 2H), 4.83 (d, $J = 12$ Hz, 2H). ^{13}C NMR (CDCl_3 , 100 MHz) δ 14.2, 17.9, 22.7, 27.4, 29.5, 31.1, 31.9, 33.9, 44.1, 45.7, 46.9, 111.1, 146.4, 174.7. Anal. Calcd for $\text{C}_{20}\text{H}_{37}\text{NO}$ (307.51): C, 78.11; H, 12.13; N, 4.55. Found: C, 78.09; H, 12.12; N, 4.64. HRMS (EI) Calcd for $\text{C}_{20}\text{H}_{37}\text{NO}$: m/z 307.2875. Found: m/z 307.2874.

1-[2-(3-Benzylamino-propyl)-allyl]-pyrrolidin-2-one (17e). IR (neat) 3065, 3026, 2935, 2855, 2789, 2718, 1646, 1602, 1492, 1450 cm^{-1} . ^1H NMR (CDCl_3 , 400 MHz) δ 1.20-1.80 (m, 14H), 2.62 (d, $J = 10.6$ Hz, 2H), 2.78-2.84 (m, 4H), 3.95 (d, $J = 13.6$ Hz, 2H), 4.82 (s, 2H), 4.82 (s, 2H). ^{13}C NMR (CDCl_3 , 100 MHz) δ 17.7, 24.8, 25.9, 32.1, 53.0, 59.5, 70.7, 112.3, 126.4, 128.0, 128.6, 139.9, 148.5. Anal. Calcd for $\text{C}_{17}\text{H}_{24}\text{N}_2\text{O}$ (272.38): C, 74.96; H, 8.88; N, 10.28. Found: C, 74.93; H, 8.84; N, 10.29. HRMS (EI) Calcd for $\text{C}_{17}\text{H}_{24}\text{N}_2\text{O}$: m/z 272.1889. Found: m/z 272.1887.

N-{2-(Diphenylmethyl)allyl}phthalimide (17f). IR (neat) 3466, 3060, 3026, 2921, 2851, 2499, 1958, 1772, 1707, 1613, 1493 cm^{-1} . ^1H NMR (CDCl_3 , 400 MHz) δ 4.31 (s, 2H), 4.66 (s, 1H), 4.88 (s, 1H), 5.22 (s, 1H), 7.117.25 (m, 10H), 7.65-7.79 (m, 4H). ^{13}C NMR (CDCl_3 , 100 MHz) δ 43.0, 55.7, 116.0, 123.1, 126.4, 128.2, 129.1, 129.1, 131.9, 133.7, 141.1, 145.7, 167.8. Anal. Calcd for $\text{C}_{24}\text{H}_{19}\text{NO}_2$ (353.41): C, 81.56; H, 5.42; N, 3.96. Found: C, 81.51; H, 5.39; N, 3.92. HRMS (EI) Calcd for $\text{C}_{24}\text{H}_{19}\text{NO}_2$: m/z 353.1416. Found: m/z 353.1414.

N-{2-(Diphenylmethyl)allyl}oxazolidin-2-one (17g). IR (neat) 3472, 3055, 2921, 1956, 1753,

1649, 1596, 1494, 1433 cm^{-1} . ^1H NMR (CDCl_3 , 400 MHz) δ 3.33 (t, $J = 7.4$ Hz, 2H), 3.88 (s, 2H), 4.10 (t, $J = 3.2$ Hz, 2H), 4.77 (s, 2H), 5.24 (s, 1H), 7.13-7.34 (m, 10H). ^{13}C NMR (CDCl_3 , 100 MHz) δ 44.3, 49.9, 55.2, 61.6, 116.3, 126.5, 128.3, 128.88, 128.89, 141.1, 146.2, 158.2. Anal. Calcd for $\text{C}_{19}\text{H}_{19}\text{NO}_2$ (293.36): C, 77.79; H, 6.53; N, 4.77. Found: C, 77.75; H, 6.49; N, 4.73. HRMS (EI) Calcd for $\text{C}_{19}\text{H}_{19}\text{NO}_2$: m/z 293.1416. Found: m/z 293.1412.

***N*-{2-(Diphenylmethyl)allyl}-5-chloromethyl-oxazolidin-2-one (17h).** IR (neat) 3083, 3059, 3025, 2905, 1954, 1768, 1650, 1598, 1492, 1444 cm^{-1} . ^1H NMR (CDCl_3 , 400 MHz) δ 3.27-3.30 (m, 1H), 3.88-3.47 (m, 2H), 3.54-3.58 (m, 1H), 3.88 (d, $J = 5.6$ Hz, 2H), 4.47-4.53 (m, 1H), 4.78 (s, 2H), 5.26 (s, 1H). ^{13}C NMR (CDCl_3 , 100 MHz) δ 44.5, 47.3, 49.9, 55.1, 71.1, 116.9, 126.58, 126.61, 128.4, 128.9, 141.0, 145.9, 156.8. Anal. Calcd for $\text{C}_{20}\text{H}_{20}\text{NO}_2\text{Cl}$ (341.83): C, 70.27; H, 5.90; N, 4.10; Cl, 10.37. Found: C, 70.24; H, 5.89; N, 4.12; Cl, 10.33. HRMS (EI) Calcd for $\text{C}_{20}\text{H}_{20}\text{NO}_2\text{Cl}$: m/z 341.1183. Found: m/z 341.1178.

1,3-Bis{2-(diphenylmethyl)allyl-imidazolidin-2-one (17i). IR (neat) 3381, 2852, 2379, 1952, 1820, 1694, 1599, 1490 cm^{-1} . ^1H NMR (CDCl_3 , 400 MHz) δ 3.03 (s, 4H), 3.76 (s, 4H), 5.18 (s, 2H), 7.14-7.28 (m, 20H), 4.67 (s, 1H), 4.76 (s, 1H). ^{13}C NMR (CDCl_3 , 100 MHz) δ 42.5, 50.0, 55.1, 115.3, 126.3, 128.2, 129.1, 141.6, 147.5, 160.6. Anal. Calcd for $\text{C}_{19}\text{H}_{20}\text{N}_2\text{O}$ (292.37): C, 84.30; H, 6.87; N, 5.62. Found: C, 84.33; H, 6.85; N, 5.26. HRMS (EI) Calcd for $\text{C}_{19}\text{H}_{20}\text{N}_2\text{O}$: m/z 498.2671. Found: m/z 498.2675.

3-Methylene-4-phenyl-1,2,3,4-tetrahydroquinoline (20a). IR (neat) 3397, 3077-2812, 1653,

1598, 1497, 1450 cm^{-1} . ^1H NMR (CDCl_3 , 400 MHz) δ 3.68 (d, J = 12 Hz, 1H), 3.78 (d, J = 12 Hz, 1H), 3.92 (bs, 1H), 4.69 (s, 1H), 5.00 (s, 1H), 5.06 (s, 1H), 6.61 (d, J = 8.0 Hz, 1H), 6.66 (t, J = 7.3 Hz, 1H), 6.87 (d, J = 6.6 Hz, 1H), 7.05 (t, J = 7.4 Hz, 1H), 7.15-7.29 (m, 5H). ^{13}C NMR (CDCl_3 , 400 MHz) δ 46.1, 50.7, 110.9, 114.5, 117.9, 126.2, 127.4, 128.2, 128.3, 130.3, 144.0, 144.60, 144.64. Anal. Calcd for $\text{C}_{16}\text{H}_{15}\text{N}$ (221.29): C, 86.84; H, 6.83; N, 6.33. Found: C, 86.81; H, 6.85; N, 6.31. HRMS (EI) Calcd for $\text{C}_{16}\text{H}_{15}\text{N}$: m/z 221.1204. Found: m/z 221.1202.

6-Chloro-3-methylene-4-phenyl-1,2,3,4-tetrahydroquinoline (20b). IR (neat) 3399, 3077-2818, 1598, 1489, 1450 cm^{-1} . ^1H NMR (CDCl_3 , 400 MHz) δ 3.68 (d, J = 12 Hz, 1H), 3.77 (d, J = 12 Hz, 1H), 3.93 (bs, 1H), 4.63 (s, 1H), 5.01 (s, 1H), 5.07 (s, 1H), 6.52 (d, J = 8.5 Hz, 1H), 6.85 (d, J = 2.4 Hz, 1H), 6.99 (dd, J = 2.4, 6.1 Hz, 1H), 7.12-7.31 (m, 5H). ^{13}C NMR (CDCl_3 , 400 MHz) δ 45.9, 50.5, 111.4, 115.5, 122.2, 124.3, 126.5, 127.4, 128.2, 128.3, 129.7, 143.1, 143.2, 143.8. Anal. Calcd for $\text{C}_{16}\text{H}_{14}\text{ClN}$ (255.74): C, 75.14; H, 5.52; Cl, 13.86; N, 6.33. Found: C, 75.12; H, 5.53; Cl, 13.82; N, 6.31. HRMS (EI) Calcd for $\text{C}_{16}\text{H}_{14}\text{ClN}$: m/z 255.0815. Found: m/z 255.0812.

6-Chloro-4-(2-chlorophenyl)-3-methylene-1,2,3,4-tetrahydroquinoline (20c). IR (neat) 3418, 3063-2849, 1602, 1495 cm^{-1} . ^1H NMR (CDCl_3 , 400 MHz) δ 3.75 (d, J = 12 Hz, 1H), 3.85 (d, J = 12 Hz, 1H), 3.99 (bs, 1H), 5.05 (d, J = 4.1 Hz, 2H), 5.22 (s, 1H), 6.54 (d, J = 8.5 Hz, 1H), 6.75 (d, J = 2.2 Hz, 1H), 6.96-7.00 (m, 2H), 7.14-7.18 (m, 2H), 7.37-7.42 (m, 1H). ^{13}C NMR (CDCl_3 , 400 MHz) δ 46.3, 46.5, 112.8, 115.6, 122.8, 126.9, 127.4, 127.9, 129.7, 130.9, 133.8, 140.8, 141.3, 143.6. HRMS (EI) Calcd for $\text{C}_{16}\text{H}_{13}\text{Cl}_2\text{N}$: m/z 289.0425. Found: m/z 289.0424.

7-Methyl-3-methylene-4-phenyl-1,2,3,4-tetrahydroquinoline (20d). IR (neat) 3399, 3055-2822, 1655, 1616, 1578 cm^{-1} . ^1H NMR (CDCl_3 , 400 MHz) δ 2.24 (s, 3H), 3.66 (d, $J = 12$ Hz, 1H), 3.76 (d, $J = 12$ Hz, 1H), 3.84 (bs, 1H), 4.65 (s, 1H), 4.98 (s, 1H), 5.03 (s, 1H), 6.42 (s, 1H), 6.48 (d, $J = 7.6$ Hz, 1H), 6.75 (d, $J = 7.8$ Hz, 1H), 7.15-7.28 (m, 5H). ^{13}C NMR (CDCl_3 , 400 MHz) δ 21.3, 46.1, 50.3, 110.8, 115.0, 119.0, 120.2, 126.2, 128.1, 128.3, 130.1, 137.1, 144.4, 144.5, 144.8. HRMS (EI) Calcd for $\text{C}_{17}\text{H}_{17}\text{N}$: m/z 235.1361. Found: m/z 235.1360.

4-Methyl-3-methylene-1, 2, 3, 4-tetrahydroquinoline (20e). IR (neat) 3397, 3072-2816, 1654, 1605, 1497 cm^{-1} . ^1H NMR (CDCl_3 , 400 MHz) δ 1.41 (d, $J = 7.1$ Hz, 3H), 3.49 (q, $J = 7.1$ Hz, 1H), 3.72 (d, $J = 12$ Hz, 1H), 3.87 (bs, 1H), 3.95 (d, $J = 12$ Hz, 1H), 4.94 (s, 1H), 4.99 (s, 1H), 6.53 (d, $J = 7.8$ Hz, 1H), 6.69 (t, $J = 5.0$ Hz, 1H), 6.96-7.04 (m, 2H). ^{13}C NMR (CDCl_3 , 400 MHz) δ 23.8, 38.8, 46.1, 108.8, 114.4, 117.8, 126.6, 126.7, 128.1, 143.9, 145.5. HRMS (EI) Calcd for $\text{C}_{11}\text{H}_{13}\text{N}$: m/z 159.1048. Found: m/z 159.1047.

1,2-Dimethylindolizine (25). IR (neat) 2919, 1716, 1628, 1434 cm^{-1} . ^1H NMR (C_6D_6 , 400 MHz) δ 1.77 (s, 3H), 1.79 (s, 3H), 5.59-5.63 (m, 1H), 5.93-5.97 (m, 1H), 6.31 (s, 1H), 6.70-6.80 (m, 2H). ^{13}C NMR (CDCl_3 , 400 MHz) δ 8.4, 10.6, 108.8, 110.1, 114.9, 116.6, 124.4, 128.4, 133.6, 133.8. HRMS (EI) Calcd for $\text{C}_{10}\text{H}_{11}\text{N}$: m/z 145.0891. Found: m/z 145.0886.

¹H-NMR of 1-(2-Methylene-3, 3-diphenylpropyl)-pyrrolidin-2-one (17a).

¹³C-NMR of 1-(2-Methylene-3,3-diphenylpropyl)-pyrrolidin-2-one (17a).

¹H-NMR of 1-(2-Nonyl-allyl)-pyrrolidin-2-one (17b).

¹³C-NMR of 1-(2-Nonyl-allyl)-pyrrolidin-2-one (17b).

D6ZV71X\k-m-Lftf
 COMNT
 DATIM Tue Dec 02 10:15:2
 OBNUC 13C
 EXMOD BCM
 OBFRQ 100.40 MHz
 OBSET 125.00 KHz
 OBFIN 10500.0 Hz
 POINT 32768
 FREQU 27173.9 Hz
 SCANS 1391
 ACQTM 1.206 sec
 PD 1.794 sec
 PW1 5.0 us
 1H
 IRNUC 23.6 c
 CTEMP
 SLVNT CDCL3
 EXREF 77.00 ppm
 BF 0.12 Hz
 RGAIN 22

¹H-NMR of 1-(2-Octyl-allyl)-pyrrolidin-2-one (17c).

¹³C-NMR of 1-(2-Octyl-allyl)-pyrrolidin-2-one (17c).

¹H-NMR of 1-(3-Hexyl-2-methylenenonyl)-pyrrolidin-2-one (17d).

¹³C-NMR of 1-(3-Hexyl-2-methylenenonyl)-pyrrolidin-2-one (17d).

¹H-NMR of 1-[2-(3-Benzylamino-propyl)-allyl]-pyrrolidin-2-one (17e).

¹³C-NMR of 1-[2-(3-Benzylamino-propyl)-allyl]-pyrrolidin-2-one (17e).

¹H-NMR of N-{2-(diphenylmethyl)allyl}phthalimide (17f).

^{13}C -NMR of N-[2-(diphenylmethyl)allyl]phthalimide (17f).

¹H-NMR of N-{2-(diphenylmethyl)allyl}oxazolidin-2-one (17g).

¹³C-NMR of N-{2-(diphenylmethyl)allyl}oxazolidin-2-one (17g).

¹H-NMR of N-{2-(diphenylmethyl)allyl}-5-chloromethyl-oxazolidin-2-one (17h).

¹³C-NMR of N-{2-(diphenylmethyl)allyl}-5-chloromethyl-oxazolidin-2-one (17h).

¹H-NMR of 1,3-Bis-{2-(diphenylmethyl)allyl}imidazolidin-2-one (17i).

¹³C-NMR of 1,3-Bis-{2-(diphenylmethyl)allyl}imidazolidin-2-one (17i).

¹H-NMR of 3-Methylene-4-phenyl-1,2,3,4-tetrahydroquinoline (20a).

¹³C-NMR of 3-Methylene-4-phenyl-1,2,3,4-tetrahydroquinoline (20a).

¹H-NMR of 6-Chloro-3-methylene-4-phenyl-1,2,3,4-tetrahydroquinoline (20b).

^{13}C -NMR of 6-Chloro-3-methylene-4-phenyl-1,2,3,4-tetrahydroquinoline (20b).

¹H-NMR of 6-Chloro-4-(2-chlorophenyl)-3-methylene-1,2,3,4-tetrahydroquinoline (20c).

¹³C-NMR of 6-Chloro-4-(2-chlorophenyl)-3-methylene-1,2,3,4-tetrahydroquinoline (20c).

^1H -NMR of 7-Methyl-3-methylene-4-phenyl-1,2,3,4-tetrahydroquinoline (20d).

¹³C-NMR of 7-Methyl-3-methylene-4-phenyl-1,2,3,4-tetrahydroquinoline (20d).

¹H-NMR of 4-Methyl-3-methylene-1,2,3,4-tetrahydroquinoline (20e).

¹³C-NMR of 4-Methyl-3-methylene-1,2,3,4-tetrahydroquinoline (20e).

¹H-NMR of 1, 2-Dimethylindolizine (25).

¹³C-NMR of 1, 2-Dimethylindolizine (25).

The reaction of **24** in the presence of the proton source

Table S1. Various H^+ Sources.^a

H^+ Sources	NMR Yield of 25 , %
H_2O	55
H_2O^b	Decomposition of 24
H_2O^c	Decomposition of 24
dil. HCl	Decomposition of 24
isopropanol	48 ^c
No H^+ Source	57 ^d

^a 0.2 equiv of H^+ source was used. ^b CH_3CN was not used. ^c 0.5 mL of H^+ source was used.

^d Isolated yield.