Supporting Information for

Efficient Aziridination of Olefins Catalyzed by A Unique Disilver(I) Compound

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General. All reagents were obtained commercially unless otherwise noted. All reactions were performed under a purified N₂ atmosphere. Organic solutions were concentrated under reduced pressure (ca. 15 mm Hg) by rotary evaporation. Dichloromethane and acetonitrile were freshly distilled from CaH₂. Chromatographic purification of products was accomplished using forced-flow chromatography on EM Science Geduran silica gel 60 (35-75 μ m). Thin layer chromatography was performed on EM Science silica gel 60 F254 plates (250 μ m). Visualization of the developed chromatogram was accomplished by fluorescence quenching and by staining with ethanolic anisaldehyde, aqueous potassium permanganate, or aqueous ceric ammonium molybdate (CAM) solution. Nuclear magnetic resonance (NMR) spectra were acquired on a Varian XL-400 operating at 400 and 100 MHz for ¹H and ¹³C, respectively, and are referenced internally according to residual protio solvent signals. Data for ¹H NMR are recorded as follows: chemical shift (ppm), multiplicity (s, singlet; d, doublet; m, multiplet), integration, coupling constant (Hz). Data for ¹³C NMR are reported in terms of chemical shift (δ , ppm). Low-resolution mass spectra were obtained from the Mass Spectrometry Facility, University of Chicago. X-ray diffraction data were collected on a Bruker Platform goniometer with a Charged Coupled Device (CCD) detector (Smart Apex). Structures were solved by direct methods using the SHELXTL (version 5.1) program library (G. Sheldrick, Bruker Analytical X-ray Systems, Madison, WI).

Catalyst preparation and recovery

[Ag₂(tBu₃tpy)₂(NO₃)](NO₃) (1)

A mixture of Ag(NO₃) (340 mg, 2 mmol) and **tBu₃tpy** (802 mg, 2 mmol) was dissolved in CH₃CN (10 mL), stirred at r.t. for 5 minutes and then filtered. The filtrate was allowed to stand in air. Pale yellow plate-like crystals were deposited after 3 days, washed with diethyl ether and dried in air. Yield: 95% (on a silver basis). Anal. Calcd (found): C, 56.75 (55.39); H, 6.17 (6.07); N, 9.80 (9.48). IR (KBr pellet, cm⁻¹): 2967 w, 2913 w, 2873 w, 1603 s, 1549 m, 1481 w, 1395 s, 1367 s, 1334 vs, 1250 m, 1203 w, 1124 w, 1036 w, 1013 w, 916 w, 896 w, 845 m, 751 w, 727 w, 691 w, 667 w, 629 w. ¹H NMR (CD₃CN, 400 MHz) δ 8.18 (s, 2H, Ar-H), 8.13 (s, 2H, Ar-H), 8.01(d, 2H, J = 7.6, Ar-H), 7.32 (d, 2H, J = 5.6, Ar-H), 1.92 (s, 6H, CH₃CN), 1.65 (s, 9H, tBu), 1.34 (s, 18H, tBu).

[Ag₂(tBu₃tpy)₂(CF₃SO₃)](CF₃SO₃)·2EtOAc·0.5Hexane (2)

In a 10 mL dry Schlenk tube, a mixture of Ag(CF₃SO₃) (5.1 mg, 0.02mmol) and **tBu₃tpy** (8.0 mg, 0.02 mmol) was stirred in 3 mL of CH₃CN for 5~10 minutes, and then 4 Å activated molecular sieves (0.5 g) was added. The resulting slightly yellow mixture was cooled to 0 °C and then PhI=NTS (186.5 mg, 0.5 mmol) was added. The suspension turned deep brown. Styrene (260 mg, 2.5 mmol, 5 equiv.) was added and the solution was

stirred at 0°C for 0.5 h and warmed to r.t. and further stirred for 6 hours, during which time the solution color faded to pale yellow. The reaction mixture was diluted with 10 mL of CH₂Cl₂ and filtered through Celite. The filter cake was washed thoroughly with (2 × 5 mL) CH₂Cl₂ and the combined filtrates were concentrated under reduced pressure. The isolated material was purified by chromatography on silica gel using hexane/EtOAc = 4:1 as the eluent solvent to afford the aziridine (yield: 90%). Pure EtOAc was used as the eluent solvent to give the dimeric silver cluster. X-ray quality crystals were obtained by slowly evaporating the solution at r.t. for 4 days, washed with diethyl ether and dried in air. Yield: 87% (on a silver basis). The sample was dried and submitted for elemental analysis. Anal. Calcd for C₅₆H₇₀Ag₂F₆N₆O₆S₂ {[Ag₂(tBu₃tpy)₂(CF₃SO₃)](CF₃SO₃)}: C, 51.07; H, 5.36; N, 6.38%. Found: C, 50.50; H, 5.27; N, 6.28%. The microanalysis results are not consistent with X-ray crystallography, with the loss of EtOAc and hexane solvent molecules upon drying. IR (KBr pellet, cm⁻¹): 2962 w, 2914 w, 2874 w, 1605 s, 1550 m, 1482 w, 1396 s, 1367 m, 1295 vs, 1223 m, 1204 w, 1148 s, 1029 vs, 1002 w, 914 w, 881 w, 849 m, 752 w, 729 w, 667 w, 635 s.

Aziridination reaction of styrene catalyzed by [Ag₂(tBu₃tpy)₂(NO₃)](NO₃) (1)

The isolated crystals of **1** (11.4 mg, 0.01 mmol) were dissolved in 2 mL CH₃CN and then 4 Å activated molecular sieves (0.5 g) was added. The resulting solution was cooled to 0 °C and PhI=NTS (186.5 mg, 0.5 mmol) was added. The suspension turned deep brown quickly. Styrene (260 mg, 2.5 mmol, 5 equiv.) was added and the solution was stirred at 0°C for 0.5 h, warmed to r.t., and stirred for 6 hours. The color of this solution faded to pale yellow at this point. The reaction mixture was diluted with 10 mL of CH₂Cl₂ and

filtered through Celite. The filter cake was washed thoroughly with (2×5 mL) CH_2Cl_2 and the combined filtrates were concentrated under reduced pressure. The isolated material was purified by chromatography on silica gel (hexane/EtOAc = 4:1) affording the white crystalline aziridine (Yield: 89%).

Experimental procedure and product characterization for the aziridination reactions (see Table 1 in main text for more details):

In a 10 mL dry Schlenk tube, a mixture of $\text{Ag}(\text{NO}_3)$ (1.7 mg, 0.01 mmol) and **tBu₃tpy** (4.0 mg, 0.01 mmol) was stirred in 2 mL of CH_3CN for 5~10 minutes. The resulting slightly yellow mixture was cooled to 0 °C and then PhI=NTS (186.5 mg, 0.5 mmol) was added together with 4 Å activated molecular sieves (0.5 g). The suspension turned deep brown quickly. Styrene (260 mg, 2.5 mmol, 5 equiv.) was added and the solution was stirred at 0°C for 0.5 h, warmed to r.t., and further stirred for 8-20 hours (8 h for styrene, cyclooctene, tran-methy-styrene and cyclohexene, 12 h for p-chlor-styrene, o-bromo-styrene and cis- and trans-stillbene, and 20 h for 1-petene and 1-hexene), during which time the solution color faded to pale yellow. The reaction mixture was diluted with 10 mL of CH_2Cl_2 and filtered through Celite. The filter cake was washed thoroughly with (2×5 mL) CH_2Cl_2 and the combined filtrates were concentrated under reduced pressure. The isolated material was purified by chromatography on silica gel to afford the desired aziridine product. All products were identified by comparing the ^1H and ^{13}C NMR spectra to those of the known compounds.¹⁻³

Purified by chromatography on silica gel (4:1 hexanes/EtOAc); colorless oil (66%): ^1H NMR (CDCl_3 , 400 MHz) δ 7.82 (d, 2H, J = 8.2 Hz, Ar-H), 7.30 (d, 2H, J = 8.2 Hz, Ar-H), 2.73 (m, 1H, CH-aziridine), 2.60 (d, 1H, J = 7.2, CH-aziridine), 2.42 (s, 3H, Ar-CH₃), 2.04 (d, 1H, J = 4.8, CH-aziridine), 1.58-1.19 (m, 4H, aliphatic-CH), 0.84 (t, 3H, J = 6.6 Hz, aliphatic-CH₃) ppm; ^{13}C NMR (CDCl_3 , 100 MHz) 144.8, 135.6, 130.0, 128.4, 40.6, 34.2, 33.7, 31.1, 22.1, 14.0 ppm; MS (m/z) 239 (M^+)

Purified by chromatography on silica gel (4:1 hexanes/EtOAc); viscous liquid

(71%): ^1H NMR (CDCl_3 , 400 MHz) δ 7.83 (d, 2H, J = 8.1 Hz, Ar-H), 7.25 (d, 2H, J = 8.0 Hz, Ar-H), 2.70 (m, 1H, CH-aziridine), 2.61 (d, 1H, J = 7.2, CH-aziridine), 2.42 (s, 3H, Ar-CH₃), 2.03 (d, 1H, J = 5.6, CH-aziridine), 1.46-1.12 (m, 6H, aliphatic-CH), 0.86 (t, 3H, J = 6.6 Hz, aliphatic-CH₃) ppm; ^{13}C NMR (CDCl_3 , 100 MHz) 144.8, 130.2, 128.9, 127.8, 40.6, 33.7, 33.2, 30.1, 22.0, 20.5, 13.3 ppm; MS (m/z) 253 (M^+)

Purified by chromatography on silica gel (4:1 hexanes/EtOAc); white solid (81%): ^1H NMR (CDCl_3 , 400 MHz) δ 7.85 (d, 2H, J = 8.2 Hz, Ar-H), 7.31 (d, 2H, J = 8.0 Hz, Ar-H), 2.95 (t, 2H, J = 1.2 Hz, CH-aziridine), 2.41 (s, 3H, Ar-CH₃), 1.76 (m, 4H, ring-CH), 1.44-1.35 (m, 2H, ring-CH), 1.23-1.18 (m, 2H, ring-CH) ppm; ^{13}C NMR (CDCl_3 , 100 MHz) 144.0, 135.8, 129.4, 127.6, 39.8, 22.8, 21.5, 19.4 ppm; MS (m/z) 251 (M^+)

Purified by chromatography on silica gel (6:1 hexanes/EtOAc), white solid (88%): ^1H NMR (CDCl_3 , 400 MHz) δ 7.86 (d, 2H, J = 8.2 Hz, Ar-H), 7.34 (d, 2H, J = 8.2 Hz, Ar-H), 2.78 (m, 1H, CH-aziridine), 2.45 (s, 3H, Ar-CH₃), 2.00 (m, 1H, CH-aziridine), 1.3-1.65 (m, 12H) ppm; ^{13}C NMR (CDCl_3 , 100 MHz) 144.1, 135.8, 130.5, 127.6, 44.0, 26.4, 26.2, 25.3, 21.6 ppm; MS (m/z) 279 (M^+)

Purified by chromatography on silica gel (4:1 hexanes/EtOAc); white solid (91%): ^1H NMR (CDCl_3 , 400 MHz) δ 7.86 (d, 2H, J = 8.3 Hz, Ar-H), 7.28 (m, 7H, Ar-H), 3.78 (m, 1H, CHPh), 2.98 (d, 1H, J = 7.8, CH-aziridine), 2.43 (s, 3H, Ar-CH₃), 2.37 (d, 1H, J = 4.4 Hz, CH-aziridine); ^{13}C NMR (CDCl_3 , 100 MHz) δ 145.2, 135.6, 130.3, 129.2, 128.8, 128.5, 127.0, 41.6, 36.5, 22.2. MS (m/z) 273 (M^+)

Purified by chromatography on silica gel (3:1 hexanes/EtOAc); white solid (74%): ^1H NMR (CDCl_3 , 400 MHz) δ 7.86 (d, 2H, $J = 8.3$ Hz, Ar-H), 7.34 (2H, $J = 8.0$ Hz, Ar-H), 7.26 (d, 2H, $J = 8.2$, Ar-H), 7.14 (d, 2H, $J = 7.8$, Ar-H), 3.73 (m, 1H, CH-aziridine), 2.98 (m, 1H, $J = 7.2$, CH-aziridine), 2.44 (s, 3H, Ar- CH_3), 2.35 (s, $J = 4.3$ Hz, CH-aziridine); ^{13}C NMR (CDCl_3 , 100 MHz) δ 144.9, 134.7, 134.2, 133.6, 129.8, 128.8, 128.1, 127.9, 40.3, 36.1, 21.7. MS (m/z) 307 (M^+)

Purified by chromatography on silica gel (3:1 hexanes/EtOAc); white solid (80%): ^1H NMR (CDCl_3 , 400 MHz) δ 7.89 (d, 2H, $J = 7.4$ Hz, Ar-H), 7.48 (m, 2H, Ar-H), 7.34 (d, 2H, $J = 8.0$ Hz, Ar-H), 7.22-7.09 (m, 3H, Ar-H), 3.96 (m, 1H, CH-aziridine), 3.01 (d, 1H, $J = 7.8$, CH-aziridine), 2.41 (s, 3H, Ar- CH_3), 2.25 (d, $J = 4.3$ Hz, $J = 6.8$, CH-aziridine); ^{13}C NMR (CDCl_3 , 100 MHz) δ 145.3, 135.1, 132.8, 132.5, 130.5, 130.2, 128.5, 128.2, 128.0, 123.7, 41.7, 36.3, 22.1. MS (m/z) 353 (M^+)

Purified by chromatography on silica gel (6:1 hexanes/EtOAc); white solid (90%): ^1H NMR (CDCl_3 , 400 MHz) δ 7.82 (d, 2H, $J = 8.2$ Hz, Ar-H), 7.24-7.15 (d, 5H, Ar-H), 7.11 (d, 2H, $J = 7.4$, Ar-H), 3.79 (d, 1H, $J = 4.4$, CH-aziridine), 2.90 (m, 1H, CH-aziridine), 2.35 (s, 3H, Ar- CH_3), 1.84 (d, 3H, $J = 6.0$ Hz, CH_3); ^{13}C NMR (CDCl_3 , 100 MHz) δ 144.1, 138.1, 135.8, 129.5, 128.4, 128.1, 127.1, 126.2, 49.3, 21.5, 14.2 ppm; MS (m/z) 287 (M^+)

Purified by chromatography on silica gel (6:1 hexanes/EtOAc); white solid (86%, 20:1/*cis:trans*): ^1H NMR (CDCl_3 , 400 MHz) δ 7.95 (d, 2H, $J = 8.2$ Hz, Ar-H), 7.36 (d, 2H, $J = 8.2$ Hz, Ar-H), 7.10-7.01 (m, 10H, Ar-H), 4.22 (s, 2H, CH-aziridine), 2.43 (s, 3H, Ar-

CH₃); ¹³C NMR (CDCl₃, 100 MHz) δ 145.2, 135.2, 132.4, 130.0, 128.7, 128.5, 128.3, 127.9, 47.9, 22.1 ppm; MS (m/z) 349 (M⁺).

Purified by chromatography on silica gel (6:1 hexanes/EtOAc); white solid (88%): ¹H NMR (CDCl₃, 400 MHz) δ 7.70 (d, 2H, J = 8.2 Hz, Ar-H), 7.49-7.35 (m, 10H, Ar-H), 7.21 (d, 2H, J = 8.2, Ar-H), 4.27 (s, 2H, CH-aziridine), 2.38 (s, 3H, Ar-CH₃); ¹³C NMR (CDCl₃, 100 MHz) δ 143.8, 134.8, 132.0, 129.9, 128.5, 128.0, 127.8, 127.5, 49.9, 21.3 ppm; LRMS (EI). MS (m/z) 349 (M⁺)

X-ray crystallography

Diffraction data were collected on a Bruker SMART APEX diffractometer at 100 K. Integration of intensities and refinement of cell parameters were done using SAINT.⁴ Absorption corrections were applied using SADABS¹ or psi-scans. The space groups P3(1) for **1** and P2(1)/n for **2** were determined based on systematic absences and intensity statistics. The structures were solved by direct methods and refined by full-matrix least-squares calculations using the SHELXTLPC program and anisotropic thermal parameters for all non-hydrogen atoms except the disordered atoms of the free anions and solvent molecules. Hydrogen atoms were fixed at calculated positions for all non-disordered carbon atoms and refined isotropically. Two *tert*-butyl groups of tBu₃tpy in complex **1** were split into two parts, each having a site occupancy factor of 0.5. The nitrate anions in complex **1** are disordered, the geometry of which is partly constrained and refined accordingly. The final highest peaks are close to the silver ions. The space group was checked for any missing symmetry using the Platon program.

$$R_{\text{int}} = \frac{\sum |F_o^2 - \langle F_o^2 \rangle|}{\sum F_o^2}$$

$$R_1 = \frac{\sum ||F_o| - |F_c||}{\sum |F_o|}$$

$$wR_2 = \left[\frac{\sum [w(F_o^2 - F_c^2)^2]}{\sum [w(F_o^2)^2]} \right]^{1/2} \text{ where: } w = 1/[\sigma^2(F_o^2) + (aP)^2 + bP];$$

$$GoF = S = \left[\frac{\sum [w(F_o^2 - F_c^2)^2]}{(n-p)} \right]^{1/2}$$

n = number of independent reflections;

p = number of parameters refined.

References

1. Evans, D. A.; Faul, M. M.; Bilodeau, M. T. *J. Am. Chem. Soc.*, **1994**, 116, 2742.
2. Farmer, J. J.; Schroeder, F. C.; Meinwald, J. *Helv. Chim. Acta.*, **2000**, 83, 2607.
3. Jiang, J. L; Huang, J. S.; Yu, X. Q.; Zhu, N.; Che, C. M. *Chem. Eur. J.*, **2002**, 83, 563.
4. All software and sources of scattering factors are contained in the SHELXTL (version 5.1) program library (G.Sheldrick, Bruker Analytical X-ray Systems, Madison, WI).

¹H NMR spectra (CD₃CN) of monitoring the formation of **1**

Figure S1. ¹H NMR spectra (400 Hz, CD₃CN, 23 °C) of (top) immediate mixing solutions of Ag(NO₃) and tBu₃tpy (molar ration: 1:1); (middle) 5 min after mixing; (bottom) the solution of crystallized **1**.

UV-Vis spectra

Figure S2. UV-vis spectra of the CH₃CN solutions of in situ-generated **1** (**a**, final concentration = 2.5×10^{-6} M, $\lambda_{\text{max}} = 224$ nm, $\epsilon = 3.086 \times 10^5$ M⁻¹) and after addition of 1 equiv of PhI=NTs (**b**, final concentration = 1.25×10^{-6} M, $\lambda_{\text{max}} = 237$ nm, $\epsilon = 1.087 \times 10^6$ M⁻¹). All UV-Vis spectra were taken in a 1 cm quartz cuvette with an acetonitrile blank at r.t.

Figure S3. ORTEP drawing of the cation $[\text{Ag}_2(\text{tBu}_3\text{tpy})_2(\text{NO}_3)]^+$ of **1** (35% thermal ellipsoids).

Figure S4. ORTEP drawing of the cation $[\text{Ag}_2(\text{tBu}_3\text{tpy})_2(\text{CF}_3\text{SO}_3)]^+$ of **2** (40% thermal ellipsoids).

Fig.S5. A space filling model of the cation $[\text{Ag}_2(\text{tBu}_3\text{tpy})_2(\text{CF}_3\text{SO}_3)]^+$ of **2**.

Table 1. Crystal data and structure refinement for complex **1**.

Identification code	1
Empirical formula	C ₅₄ H ₇₀ Ag ₂ N ₈ O ₆
Formula weight	1142.92
Temperature	100(2) K
Wavelength	0.71073 Å
Crystal system	Trigonal
Space group	P3(1)
Unit cell dimensions	
	a = 16.688(3) Å alpha = 90°
	b = 16.688(3) Å beta = 90°
	c = 16.208(4) Å gamma = 120°
Volume, Z	3909.0(12) Å ³ , 3
Density (calculated)	1.457 Mg/m ³
Absorption coefficient	0.809 mm ⁻¹
F(000)	1776
Crystal size	0.15 × 0.10 × 0.5 mm
Theta range for data collection	1.89 to 22.51°
Limiting indices	-19 ≤ h ≤ 20, -20 ≤ k ≤ 20, -13 ≤ l ≤ 20
Reflections collected	12920
Independent reflections	5257 (R _{int} = 0.0281)
Completeness to theta = 22.51°	99.9 %
Refinement method	Full-matrix least-squares on F ²
Data / restraints / parameters	5257 / 19 / 554
Goodness-of-fit on F ²	1.050
Final R indices [I > sigma (I)]	R ₁ = 0.0854, wR ₂ = 0.2274
R indices (all data)	R ₁ = 0.0908, wR ₂ = 0.2331
Absolute structure parameter	0.00
Largest diff. peak and hole	3.710 and -0.756 e.Å ⁻³

Table 2. Bond lengths (Å) and angles (°) for complex **1**.

Ag(1)-Ag(2)	2.8415(17)
Ag(1)-N(1)	2.294(12)
Ag(1)-N(2)	2.448(12)
Ag(1)-N(4)	2.242(11)
Ag(1)-N(5)	2.447(11)
Ag(2)-N(3)	2.270(12)
Ag(2)-N(6)	2.268(13)
Ag(2)-O(1)	2.331(18)
Ag(2)-O(2)	2.70(2)
N(1)-Ag(1)-N(2)	70.3(5)
N(1)-Ag(1)-N(4)	155.0(5)
N(1)-Ag(1)-N(5)	119.8(5)
N(2)-Ag(1)-N(4)	118.9(4)
N(2)-Ag(1)-N(5)	136.4(4)
N(3)-Ag(2)-N(6)	149.8(5)
N(3)-Ag(2)-O(1)	121.9(5)
N(3)-Ag(2)-O(2)	91.1(5)
N(4)-Ag(1)-N(5)	71.2(4)
N(6)-Ag(2)-O(1)	85.3(5)
N(6)-Ag(2)-O(2)	118.4(5)
O(1)-Ag(2)-O(2)	48.7(6)

Table 3. Crystal data and structure refinement for complex **2**.

Identification code	2
Empirical formula	C ₆₇ H ₉₃ Ag ₂ F ₆ N ₆ O ₁₀ S ₂
Formula weight	1536.33
Temperature	100(2) K
Wavelength	0.71073 Å
Crystal system	Monoclinic
Space group	P2(1)/n
Unit cell dimensions	
	a = 16.194(3) Å alpha = 90°
	b = 14.666(3) Å beta = 04.752(3)°
	c = 30.836(5) Å gamma = 90°
Volume, Z	7082(2), 4
Density (calculated)	1.441 Mg/m ³
Absorption coefficient	0.687 mm ⁻¹
F(000)	3188
Crystal size	0.1 × 0.1 × 0.05
Theta range for data collection	1.37 to 22.51°
Limiting indices	-17 ≤ h ≤ 20, -18 ≤ k ≤ 18, -38 ≤ l ≤ 33
Reflections collected	27926
Independent reflections	9263 (Rint = 0.0306)
Completeness to theta = 22.51°	99.8 %
Refinement method	Full-matrix least-squares on F ²
Data / restraints / parameters	9263 / 3 / 755
Goodness-of-fit on F ²	1.271
Final R indices [I > 2σ(I)]	R1 = 0.0744, wR2 = 0.1570
R indices (all data)	R1 = 0.0771, wR2 = 0.1583
Largest diff. peak and hole	1.494 and -1.125 e.Å ⁻³

Table 4. Bond lengths (Å) and angles (°) for complex **2**.

Ag(1)-Ag(2)	2.8966(9)
Ag(1)-N(1)	2.244(6)
Ag(1)-N(2)	2.438(6)
Ag(1)-N(5)	2.522(6)
Ag(1)-N(6)	2.224(6)
Ag(2)-N(3)	2.181(6)
Ag(2)-N(4)	2.214(6)
Ag(2)-N(5)	2.612(6)
Ag(2)-O(1)	2.517(5)

N(1)-Ag(1)-N(2)	71.7(2)
N(1)-Ag(1)-N(5)	114.9(2)
N(1)-Ag(1)-N(6)	159.7(2)
N(2)-Ag(1)-N(5)	126.11(19)
N(2)-Ag(1)-N(6)	121.8(2)
N(3)-Ag(2)-N(4)	156.4(2)
N(3)-Ag(2)-N(5)	108.7(2)
N(3)-Ag(2)-O(1)	117.2(2)
N(4)-Ag(2)-N(5)	69.1(2)
N(4)-Ag(2)-O(1)	86.40(19)
N(5)-Ag(1)-N(6)	71.1(2)
N(5)-Ag(2)- O(1)	85.53(18)