**SUPPORTING INFORMATION, PART A**

**General.** All chemicals if not noted were purchased from commercial sources. The control pore glass supports were obtained from Prime Synthesis Ltd. $^1$H NMR spectra were recorded on a Bruker Avance 500 (500 MHz) spectrometer or Bruker Avance II spectrometer (400 MHz). Chemical shifts are reported in ppm from tetramethylsilane with the solvent resonance resulting from incomplete deuterium incorporation as the internal standard (CDCl$_3$: $\delta$ 7.26 ppm). Data are reported as follows: chemical shift, multiplicity ($s$ = singlet, $d$ = doublet, $q$ = quartet, $br$ = broad, $m$ = multiplet), and coupling constants (Hz), integration. $^{13}$C MR spectra were recorded on a Bruker Avance 500 (125.8 MHz) spectrometer or Bruker Avance II spectrometer (400 MHz) spectrometer with complete proton decoupling. Chemical shifts are reported in ppm from tetramethylsilane with the solvent resonance as the internal standard (CDCl$_3$: $\delta$ 77.26 ppm). High-resolution mass spectrometry was performed on a Agilent Technologies 6520- Q-TOF ESI-MS (positive mode) at the Stockholm University or Mid-Sweden University Mass Spectrometry Facility. Enantiomer ratios were determined by HPLC (Chiral Agilent Technologies Chiralpak OD, OJ-H, AS column or Chiralcel OD-R column (4.6 mm x 250 mm)) in comparison with authentic racemic materials. Optical rotations were measured on a Perkin-Elmer 341 Polarimeter. Unless otherwise noted, all reactions were performed with distilled solvents under an atmosphere of N$_2$ in oven- (135 °C) or flame-dried glassware with standard vacuum line techniques. The Pd catalyst on the support was characterized for palladium loading by Inductively Coupled Plasma (ICP; Mikroanalytisches Laboratorium Kolbe, Germany). Elemental analyses on the Pd contents of the Pd-Amp-CPG were carried out by Medac LTD Analytical and chemical consultancy services (United Kingdom) by ICP-OES. The samples for TEM imaging were done by sonicating the Pd-Amp-CPG particles in ethanol, and then drop the suspension on a standard TEM grip and let it dry in air. The TEM was done on a JEOL 2000FX (JEOL) microscope at an accelerate voltage of 160 KV.
**Preparation of Pd⁰-CPG nanoparticles:** Amine-functionalized Amp-CPG (pore size 533Å, amine 166 μmol/g), CPG-Hybrid VBC (pore size 526Å, amine 398 μmol/g), CPG-Hybrid COPO (pore size 590Å, amine 360 μmol/g) and CPG-Hybrid (pore size 1400Å, amine 353 μmol/g) obtained from Prime Synthesis were used as supports. Thus, each amine-functionalized CPG (1g, 1 equiv, amine content) was added to a deionized water solution (45 mL, pH = 9). In parallel, Li₂PdCl₄ (2 equiv) was solubilized in a pH adjusted deionized water solution (pH = 9) and next added to the suspension. After 24 h of stirring, the Pd(II)-CPG catalyst was transferred to a centrifuge vial (45 mL) and next washed (3 x H₂O (40 mL), 3 x acetone (40 mL)) followed by drying overnight under vacuum. Next, the dry Pd(II)-CPG catalyst was suspended in deionized water (30 mL) followed by the slow addition of NaBH₄ (15 equiv), which had been dissolved in deionized water (15 mL), at room temperature. After stirring for 30 min, the obtained Pd(0)-CPG catalyst was transferred to a 45 mL centrifuge vial and washed (3 x H₂O (40 mL), 3 x acetone (40 mL)) and dried overnight under vacuum. Some of the catalysts were also further washed (3 x MeOH (40 mL), 3 x CH₂Cl₂ (40 mL)) and dried overnight under vacuum. Elemental analyses on the Pd contents of the Pd-Amp-CPG were carried out by Medac LTD Analytical and chemical consultancy services (United Kingdom) by ICP-OES.

**Typical experimental procedure for the Pd-CPG catalyst screening:**

To a suspension of Pd-CPG-catalyst (5 mol% to the alcohol) in toluene (0.5 mL) was added 1a (0.24 mmol, 1.2 equiv). The vial was capped, evacuated and an oxygen balloon was connected to the reaction vessel. The reaction was stirred at 70 °C for the time shown in Table 1. Next, the reaction was cooled to room temperature followed by addition of 3 (0.2 mmol, 1 eq) and 5 (20 mol%) and the reaction mixture was vigorously stirred for the time shown in Table 1. After removal of the Pd-catalyst, the crude reaction mixture was directly loaded on a silica-gel column and next chromatograph (pentane/EtOAc) afforded the corresponding product 4a.

**Typical experimental procedure for the Pd⁰-Amp-CPG/chiral amine-catalyzed catalytic aerobic oxidation/Michael/carbocyclization relay sequence:**

In a microwave vial, to a suspension of Pd(0)-Amp-CPG (6 mol% Pd to 1, 36 mg) in toluene (0.5 mL) was added alcohol 1 (0.12 mmol, 1.2 eq). The vial was capped, evacuated and the oxygen balloon was connected to the reaction vessel. The reaction was stirred at 70 °C for the
time shown in Table 2. Next, the reaction was cooled to room temperature. Propargyl 3 (0.1 mmol, 1 eq) and 5 (20 mol%) were added and the reaction mixture was vigorously stirred for the time shown in Table 2. After removal of the Pd-catalyst, the crude reaction mixture was directly loaded on a silica-gel column and next chromatography (pentane/EtOAc) afforded the corresponding products 4.

(1R,2R)-methyl 1-cyano-3-formyl-4-methyl-2-phenylcyclopent-3-enecarboxylate: oil. $^1$H NMR (400MHz, CDCl$_3$): $\delta$ 9.92 (s, 1H), 7.38-7.32 (m, 3H), 7.17-7.15 (m, 2H), 4.72 (bs, 1H), 3.89 (s, 3H), 3.41 (d, $J$ = 14.8 Hz, 1H), 3.26 (dt, $J$ = 12.4 Hz, $J$’ = 1.2 Hz, 1H), 2.33 (d, $J$ = 0.8 Hz, 3H); $^{13}$C NMR (100MHz, CDCl$_3$): 186.2, 168.8, 157.8, 136.8, 136.6, 129.1, 128.7, 128.0, 117.4, 58.4, 54.4, 51.7, 47.9, 14.3; HRMS (ESI) : calcd for [M+Na] (C$_{16}$H$_{15}$NO$_3$) requires m/z 292.0944, found 292.0946; $\left[\alpha\right]_D^{25} = -70.9$ (c=1.0 CHCl$_3$). The enantiomeric excess was determined by HPLC analysis in comparison with authentic racemic material (ODH-column, n-hexane/i-PrOH = 85/15, $\lambda$ = 210 nm, 1.0 ml/min) $t_r$ (major enantiomer) = 19.3 min, $t_r$ (minor enantiomer) = 29.9 min.

(1R,2R)-methyl 1-cyano-3-formyl-4-methyl-2-(4-nitrophenyl)cyclopent-3-enecarboxylate: oil. $^1$H NMR (400MHz, CDCl$_3$): $\delta$ 9.95 (s, 1H), 8.22 (d, $J$ = 8.8 Hz, 2H), 7.34 (d, $J$ = 8.8 Hz, 2H), 4.82 (bs, 1H), 3.91 (s, 3H), 3.49 (d, $J$ = 18.8 Hz, 1H), 3.32 (d, $J$ = 18.8 Hz, 1H), 2.36 (d, $J$ = 1.2 Hz, 1H); $^{13}$C NMR (100MHz, CDCl$_3$): 185.8, 168.2, 159.0, 148.1, 144.1, 136.2, 129.2, 124.2, 117.0, 57.7, 54.7, 51.3, 48.3, 14.4; HRMS (ESI) : calcd for [M+Na] (C$_{16}$H$_{14}$N$_2$O$_5$) requires m/z 337.0792, found 337.0795; $\left[\alpha\right]_D^{25} = -88.2$ (c=1, CHCl$_3$). The enantiomeric excess was determined by HPLC analysis in comparison with authentic
racemic material (ODH-column, n-hexane/i-PrOH = 75/25, \( \lambda = 254 \) nm, 1.0 ml/min) \( t_r \) (major enantiomer) = 33.6 min, \( t_r \) (minor enantiomer) = 41.1 min.

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\text{(1R,2R)-methyl 1-cyano-3-formyl-4-methyl-2-(3-nitrophenyl)cyclopent-3-ene carboxylate: oil.} \quad ^1H \text{ NMR (400MHz, CDCl}_3\text{): } \delta \ 9.97 \text{ (s, 1H), 8.22-8.19 (m, 1H), 7.98 (t, } J= 1.2 \text{ Hz, 1H), 7.59-7.53 (m, 2H), 4.83 (bs, 1H), 3.92 (s, 3H), 3.49 (d, } J = 18.8 \text{ Hz, 1H), 3.33 (d, } J = 18.8 \text{ Hz, 1H), 2.38 (d, } J = 1.6 \text{ Hz, 3H);} \quad ^13C \text{ NMR (100MHz, CDCl}_3\text{): } 185.8, 168.2, 159.3, 148.6, 139.0, 136.1, 134.7, 130.1, 123.8, 122.8, 117.1, 57.8, 54.8, 51.3, 48.1, 14.5; \quad \text{HRMS (ESI): } \text{calcd for [M+Na] (C}_{16}\text{H}_{14}\text{N}_2\text{O}_5 \text{ requires m/z 337.0792, found 337.0794; } [\alpha]_D^{25} = -89.3 \ (c=1, \text{ CHCl}_3). \quad \text{The enantiomeric excess was determined by HPLC analysis in comparison with authentic racemic material (ODH-column, n-hexane/i-PrOH = 70/30, } \lambda = 254 \text{ nm, 1.0 ml/min) } t_r \text{ (major enantiomer) = 19.4 min, } t_r \text{ (minor enantiomer) = 34.6 min.} \]

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\text{(1R,2R)-methyl 2-(2-chlorophenyl)-1-cyano-3-formyl-4-methylcyclopent-3-ene carboxylate: oil.} \quad ^1H \text{ NMR (400MHz, CDCl}_3\text{): } \delta \ 9.93 \text{ (s, 1H), 7.47 (dd, } J= 6.4 \text{ Hz, } J' = 1.6 \text{ Hz, 1H), 7.29-7.20 (m, 2H), 6.97 (dd, } J= 6.0 \text{ Hz, } J' = 1.6 \text{ Hz, 1H), 5.30 (bs, 1H), 3.89 (s, 3H), 3.41 (d, } J = 18.8 \text{ Hz, 1H), 3.27 (d, } J = 18.8 \text{ Hz, 1H), 2.34 (d, } J = 0.8 \text{ Hz, 3H);} \quad ^13C \text{ NMR (100MHz, CDCl}_3\text{): } 185.9, 169.0, 158.4, 136.3, 134.8, 134.4, 130.3, 129.9, 128.4, 127.2, 117.3, 54.8, 54.5, 50.9, 48.9, 14.4; \quad \text{HRMS (ESI): } \text{calcd for [M+Na] (C}_{16}\text{H}_{14}\text{ClNO}_3 \text{ requires m/z 326.0554, found 326.0562; } [\alpha]_D^{25} = -80.3 \ (c=1, \text{ CHCl}_3). \quad \text{The enantiomeric excess was determined by HPLC analysis in comparison with authentic racemic material (AD-column, i-hexane/i-PrOH = 90/10, } \lambda = 254 \text{ nm, 1.0 ml/min) } t_r \text{ (major enantiomer) = 33.2 min, } t_r \text{ (minor enantiomer) = 41.9 min.} \]
(1R,2R)-methyl 2-(4-bromophenyl)-1-cyano-3-formyl-4-methylcyclopent-3-enecarboxylate: oil. \[\text{\textsuperscript{1}H NMR (400MHz, CDCl}_3\text{): } \delta 9.91\ (s, 1H), 7.48\ (d, J = 6.8 Hz, 2H), 7.09\ (d, J = 6.8 Hz, 2H), 4.67\ (bs, 1H), 3.88\ (s, 3H), 3.42\ (d, J = 14.8 Hz, 1H), 3.25\ (d, J = 12.8 Hz, 1H); \text{\textsuperscript{13}C NMR (100MHz, CDCl}_3\text{): } 186.0, 168.6, 158.3, 136.5, 135.7, 132.2, 129.8, 122.8, 117.3, 57.8, 54.3, 51.3, 47.9, 14.3; HRMS (ESI): calcd for [M+Na] (C\textsubscript{16}H\textsubscript{14}BrNO\textsubscript{3}) requires m/z 370.0049, found 370.0050; [\alpha\textsubscript{D}]\textsuperscript{25} = -55.8\ (c=1, CHCl\textsubscript{3})

The enantiomeric excess was determined by HPLC analysis in comparison with authentic racemic material (AD-column, \textit{n}-hexane/i-PrOH = 90/10, \(\lambda = 250\) nm, 1.0 ml/min) \(t_r\) (major enantiomer) = 22.3 min, \(t_r\) (minor enantiomer) =34.9 min.

(1R,2R)-methyl 1-cyano-3-formyl-4-methyl-2-(p-tolyl)cyclopent-3-enecarboxylate: oil. \[\text{\textsuperscript{1}H NMR (400MHz, CDCl}_3\text{): } \delta 9.91\ (s, 1H), 7.16\ (d, J = 6.4 Hz, 2H), 4.68\ (bs, 1H), 3.88\ (s, 3H), 3.39\ (d, J = 14.8 Hz, 1H), 2.33\ (s, 3H); \text{\textsuperscript{13}C NMR (100MHz, CDCl}_3\text{): } 186.3, 168.9, 157.7, 138.4, 136.8, 133.6, 129.8, 127.9, 117.6, 58.2, 54.4, 51.8, 47.8, 21.4, 14.3; HRMS (ESI): calcd for [M+Na] (C\textsubscript{17}H\textsubscript{17}NO\textsubscript{3}) requires m/z 306.1101, found 306.1104; [\alpha\textsubscript{D}]\textsuperscript{25} = -85.2\ (c=1.30CHCl\textsubscript{3})

The enantiomeric excess was determined by HPLC analysis in comparison with authentic racemic material (ODH-column, \textit{n}-hexane/i-PrOH = 90/10, \(\lambda = 250\) nm, 1.0 ml/min) \(t_r\) (major enantiomer) = 23.6 min, \(t_r\) (minor enantiomer) =32.9 min.

(1R,2R)-methyl 1-cyano-3-formyl-4-methyl-2-propylcyclopent-3-enecarboxylate: oil. \[\text{\textsuperscript{1}H NMR (400MHz, CDCl}_3\text{): } \delta 9.93\ (s, 1H), 3.83\ (s, 3H), 3.52\ (m, 1H), 3.19\ (dd, J\textsubscript{1} = 18.4 Hz,
$J_2 = 36.3 \text{ Hz, 2H), 2.42 (s, 3H), 1.92 (m, 1H), 1.75 (m, 1H), 1.47-1.27 (m, 2H), 0.94 (t, 3H);}^{13}\text{C NMR (100MHz, CDCl}_3\text{): 186.9, 169.5, 156.9, 137.5, 118.2, 54.2, 52.5, 49.5, 48.5, 33.4, 20.2, 14.1, 14.0; HRMS (ESI) : calcd for [M+Na] (C}_{13}\text{H}_{17}\text{NO}_3 \text{ requires m/z 258.1101, found 258.1109, } [\alpha]_{D}^{25} = +15.5 (c=1.0 \text{ CHCl}_3). \text{ The enantiomeric excess was determined by HPLC analysis in comparison with authentic racemic material (ODH-column, } n\text{-hexane/i-PrOH = 98/2, } \lambda = 230 \text{ nm, 1.0 ml/min) } t_r \text{ (major enantiomer) } = 18.1 \text{ min, } t_r \text{ (minor enantiomer) } = 19.8 \text{ min.}

\text{(1R,2R)-methyl 2-butyl-1-cyano-3-formyl-4-methylcyclopent-3-enecarboxylate: oil. }^{1}\text{H NMR (400MHz, CDCl}_3\text{): } \delta 9.93 \text{ (s, 1H), 3.83 (s, 3H), 3.53 (bs, 1H), 3.26 (d, } J_1 = 18.8 \text{ Hz, 1H), 3.13 (d, } J_1 = 20.0 \text{ Hz, 1H), 2.18 (s, 3H), 1.99-1.90 \text{ (m, 1H), 1.82-1.73 (m, 1H), 1.40-1.28 (m, 4H), 0.90 (t, } J_1 = 6.8 \text{ Hz, 3H); }^{13}\text{C NMR (100MHz, CDCl}_3\text{): 186.9, 169.5, 156.9, 137.5, 118.2, 54.2, 52.5, 49.5, 48.5, 33.4, 20.2, 14.1, 14.0; HRMS (ESI) : calcd for [M+Na] (C}_{14}\text{H}_{19}\text{NO}_3 \text{ requires m/z 272.1257, found 272.1270, } [\alpha]_{D}^{25} = +30.2 (c=1.0 \text{ CHCl}_3). \text{ The enantiomeric excess was determined by HPLC analysis in comparison with authentic racemic material (ODH-column, } n\text{-hexane/i-PrOH = 98/2, } \lambda = 230 \text{ nm, 1.0 ml/min) } t_r \text{ (major enantiomer) } = 17.7 \text{ min, } t_r \text{ (minor enantiomer) } = 19.7 \text{ min.}

\text{(1R,2R)-methyl 2-(but-3-en-1-yl)-1-cyano-3-formyl-4-methylcyclopent-3-enecarboxylate: oil. }^{1}\text{H NMR (400MHz, CDCl}_3\text{): } \delta 9.93 \text{ (s, 1H), 5.78 (m, 1H), 5.08-4.97 (m, 2H), 3.83 (s, 3H), 3.54 (m, 1H), 3.22 (dd, } J_1 = 18.4 \text{ Hz, } J_2= 36.3 \text{ Hz, 2H), 2.18 (s, 3H), 2.18-2.03 \text{ (m, 3H), 1.88 (m, 1H); }^{13}\text{C NMR (100MHz, CDCl}_3\text{): 186.9, 169.3, 157.1, 137.4, 137.3, 118.1, 115.8, 54.2, 52.0, 49.3, 48.5, 31.1, 30.5, 14.1; HRMS (ESI) : calcd for [M+Na] (C}_{14}\text{H}_{19}\text{NO}_3 \text{ requires m/z 270.1102, found 270.1106, } [\alpha]_{D}^{25} = +12.3 (c=1.0 \text{ CHCl}_3). \text{ The enantiomeric excess was determined by HPLC analysis in comparison with authentic racemic material (ODH-column, } n\text{-hexane/i-PrOH = 98/2, } \lambda = 230 \text{ nm, 1.0 ml/min) } t_r \text{ (minor enantiomer) } = 22.5 \text{ min, } t_r \text{ (majoror enantiomer) } = 24.2 \text{ min.}
(1R,2R)-methyl-1-cyano-3-formyl-4-methyl-2-(p-tolyl)cyclopent-3-enecarboxylate:

Yellow oil. $^1$H NMR (400MHz, CDCl$_3$): $\delta$ 9.91 (s, 1H), 7.08 (d, $J$ = 8.7 Hz, 2H), 6.88 (d, $J$ = 8.7 Hz, 2H), 4.68 (br s, 1H), 3.88 (s, 3H), 3.79 (s, 3H), 3.38 (d, $J$ = 18.6 Hz, 1H), 3.24 (dt, $J$ = 18.6, 3.0 Hz, 1H), 2.32 (s, 3H); $^{13}$C NMR (100MHz, CDCl$_3$): 186.3, 168.9, 159.8, 157.6, 136.9, 129.2, 128.6, 117.6, 114.4, 57.9, 55.3, 54.4, 51.9, 47.7, 14.3; HRMS (ESI): calcd for [M+Na] (C$_{17}$H$_{17}$NO$_4$) requires m/z 322.1050, found 322.1051; $\left[\alpha\right]_{D}^{25}$ = -108.7 (c=1.6, CHCl$_3$). The enantiomeric excess was determined by HPLC analysis in comparison with authentic racemic material (AS-H-column, $n$-hexane/i-PrOH = 85/15, $\lambda$ = 210 nm, 1.0 ml/min) $t_r$ (major enantiomer) = 43.1 min, $t_r$ (minor enantiomer) =73.6 min.

TEM analysis:
The samples for TEM imaging were done by sonicating the Pd-Amp-CPG particles in ethanol, and then drop the suspension on a standard TEM grip and let it dry in air. The TEM was done on a JEOL 2000FX (JEOL) microscope at an accelerate voltage of 160 KV (Figure 1S and 2S).
Figure 1S.

Figure 2S.
Display Report

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Sample Name: id1476
Comment

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Bruker Compass DataAnalysis 4.0
printed: 2013-08-16 10:27:24
Display Report

Analysis Info
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Instrument / Ser#: microTOF 125

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Bruker Compass DataAnalysis 4.0 printed: 2013-08-16 10:22:14 Page 1 of 1
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Additional Info : Peak(s) manually integrated

DAD1 C, Sig=210,4 Ref=off (SAMSON/CP-27-008_RAC/CP-27-008_RAC.D)

Area Percent Report

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Dilution: : 1.0000
Sample Amount: : 10.00000 [ng/µl] (not used in calc.)

Use Multiplier & Dilution Factor with ISTDS

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**Additional Info:** Peak(s) manually integrated

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**Area Percent Report**

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