Electronic Supplementary Material (ESI) for Chemical Communications

Oxidative Imination of Toluenes Catalyzed by Pd-Au/Silica Gel Under Mild Reaction Conditions

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1. Experimental

All solvents and all chemicals purities are >98% and supplied by Shanghai Lianguan Biochemical Co., Ltd. and were used as received.

NMR spectra were measured using a Bruker ARX 400 or ARX 100 spectrometer at 400 MHz (^{1}H) and 100 MHz (^{13}C). All spectra were recorded in $CDCl_3$ and chemical shifts (δ) are reported in ppm relative to tetramethylsilane referenced to the residual solvent peaks. GC nanalysis (HP 6890/5973 equipped with a column (30 m × 0.25 mm) and a flame ionization detector (FID)) was carried out using biphenol as an internal standard.

X-ray diffraction (XRD)

XRD patterns of samples were obtained on a STADI P automated transmission diffractometer instrument equipped with an incident beam curved germanium monochromator selecting Cu Kα1 radiation (40 KV and 40 mA) was used as the X-ray source. The precipitated particles were dried in air and pressed on a glass slide for analysis. The samples were oscillated in the xy-axis during data collecting.

Transmission Electron Microscopy (TEM)

For the prepared catalysts, the particle disperion was diluted by ethanol, and then 10 μL of disperion was cast on the TEM grids with a micorpippet. TEM images were obtained on a Tecnai G2 F30 S-Twin operating at 300 kV. Single-particle EDX analysis was performed on a Tecnai G2 F30 S-Twin Field Emission TEM with STEM mode.

X-ray Photoelectron Spectrocopy (XPS)

The XPS measurements were performed with a VG ESCALAB 210 instrument provided with a dual Mg/Mg anode X-ray source, a hemispherical capacitor analyser and a 5 keV Ar^{+} ion-gun. All spectra were recorded using non-monochromatic Mg Kα (1253.6 eV) radiation.

BET and ICP-AES analysis

Nitrogen adsorption-desorption isotherms were measured at 77 K using Micromeritics 2010 instrument. The pore-size distribution was calculated by Barrett, Joyner and Halenda (BJH) method from desorption isotherm. The Pd and Au contents of the catalysts were measured by inductively coupled plasma-atomic emission spectrometry (ICP-AES), using an Iris advantage Thermo Jarrel Ash device. The Pd and Au ions in the filtered solution were out of determined limits.

Typical procedure for catalyst preparation

300 mg polyvinylpyrrolidone (PVP, Mw = 10000) and 300 mg hexadecyltrimethylammonium bromide (CTAB) were added into a 60 mL water/alcohol (V : V = 1 : 5) mixture and agitated till complete dissolution. Then, suitable amounts of $HAuCl_4$ (0.25 M) and H_2PdCl_4 (0.14 M) were dropwise added and the mixture was further stirred for 30 min at 40 ^oC. Subsequently, 5 mL tetraethylorthosilicate (TEOS) and 1.0 g urea were added and the resulting solution was transferred into a Teflon-lined stainless steel autoclave. After being hydrothermal treated at 140 ^oC for 12 h, the reaction mixture was cooled to room temperature, centrifuged, washed by water, dried at 120 ^oC for 4 h and calcined in static air at 400 ^oC for 4 h in air. ~1.5 g black solid sample was obtained and denoted as Pd-Au/SiO_2-P-C. The other samples with different amounts of Pd, Au, PVP or CTAB were prepared with the same procedure and denoted as Pd/SiO_2-P-C, Au/SiO_2-P-C, Pd-Au/SiO_2-P, Pd-Au/SiO_2-C, Pd-Au/SiO_2 and Pd-Au/SiO_2-P-C-1. The Pd and Au loadings were determined by ICP-AES analysis.

Typical procedure for the coupling reaction of sulfonamides with toluenes:

1.0 mmol sulfonamides, 50 mg catalyst (0.21-0.63 mol% Pd and Au) and 3 mL toluene were added into a 100 mL autoclave equipped with magnetic stirrer. The autoclave was sealed and exchanged with oxygen for 3 times and reacted at 120 ^oC under 1 MPa O_2 for 24 h. Then it was cooled to room temperature. ~10 mL acetone was added to dissolve the reaction mixture and filtered by celite. The crude reaction mixture was concentrated *in vacuo* and purified by column chromatography [petroleum ether (b.p. 60-90ºC)/ethyl acetate] to give the corresponding imine in good yields.

Typical procedure for the coupling reaction of amines with toluenes:

1.0 mmol amines, 50 mg catalyst (Pd-Au/SiO_2-P-C) and 3 mL toluene were added into a 100 mL autoclave equipped with magnetic stirrer. The autoclave was sealed and exchanged with oxygen for 3 times and reacted at 120 ^oC under 1 MPa O_2 for 24 h. Then it was cooled to room temperature. 77 mg diphenyl was added as internal standard and ~10 mL ethanol was added to dissolve the reaction mixture. The GC-yield was obtained by GC-FID analysis (HP 6890).

2. Table S1 Results of the reusability study and activity measurement of the filtrate

Entry	Recycle Time	Catalyst	GC-yield[a]
1	1	Pd-Au/SiO_2-P-C	91/86[b]
2	2	Pd-Au/SiO_2-P-C	89
3	3	Pd-Au/SiO_2-P-C	87
4	4	Pd-Au/SiO_2-P-C	82
5	5	Pd-Au/SiO_2-P-C	85/83[b]
6[c]	Filtrate	----	No reaction

[a] Determined by GC-FID with biphenol as internal standard; [b] isolated yields; [c] 1 mmol benzenefulfonamide was added after removing the catalyst and reacted for 24 h in the presence of 1 MPa O_2.

3. Table S2 ICP-AES analysis of the catalysts

Entry	Catalyst	Pd	Au	Mole ratio
1	Pd/SiO_2-P-C	0.9	0	
2	Pd-Au/SiO_2-P	0.87	0.81	2.00
3	Pd-Au/SiO_2-P-C	0.88	0.83	1.97
4	Au/SiO_2-P-C		0.82	
5	Pd-Au/SiO_2-C	0.86	0.83	1.92
6	Pd-Au/SiO_2	0.88	0.84	1.95
7	Pd-Au/SiO_2-P-C-after reaction	0.86	0.84	1.92
8	Filtered solution	Not detectable	Not detectable	Not detectable

4. Figure S1

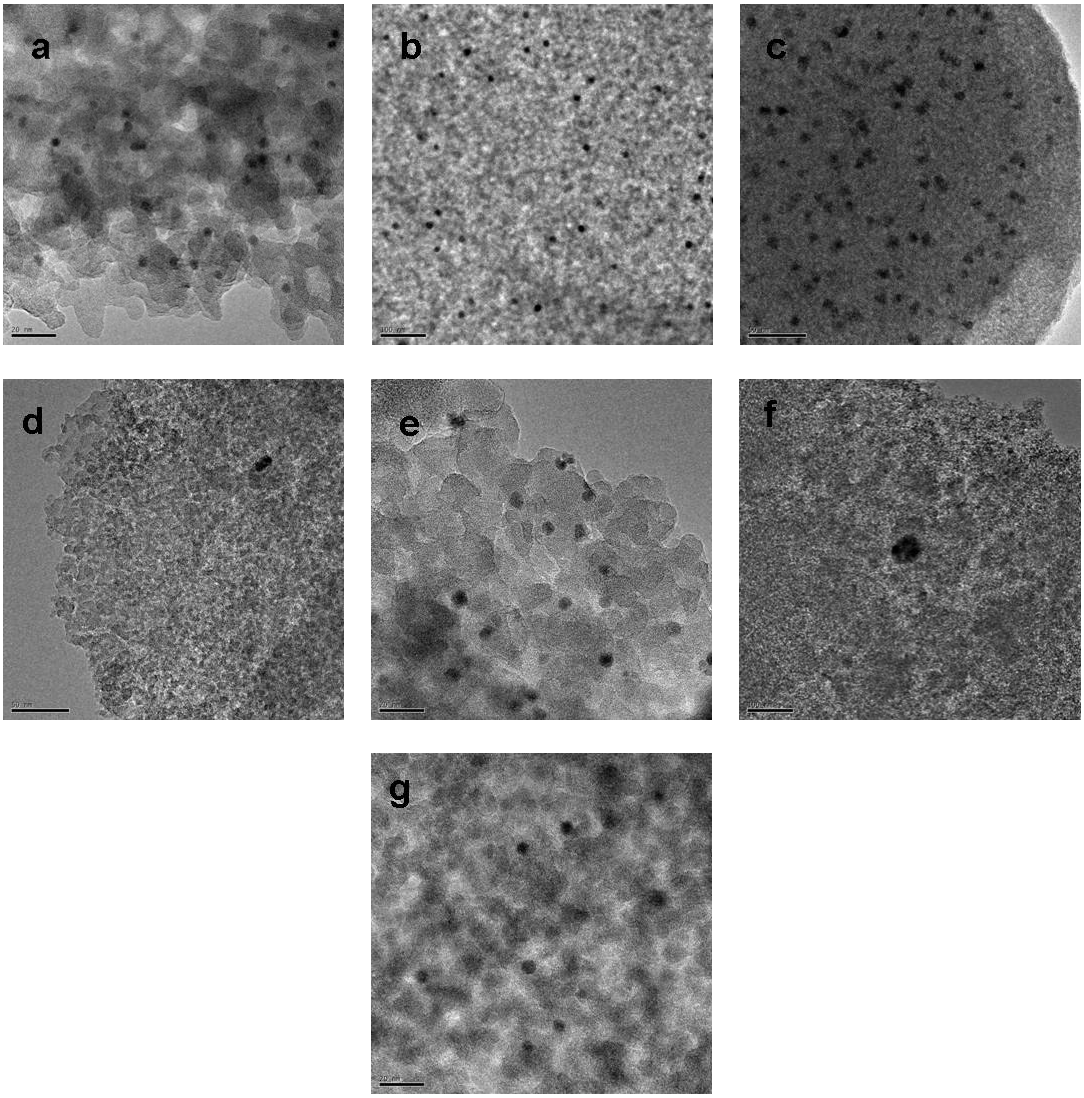


Figure S1. STEM images of the obtained samples: a) Pd/SiO_2-P-C; b) Au/SiO_2-P-C; c) Pd-Au/SiO_2-P; d) Pd-Au/SiO_2-C; e) Pd-Au/SiO_2-P-C; f) Pd-Au/SiO_2; g) Pd-Au/SiO_2-P-C-1

5. Figure S2

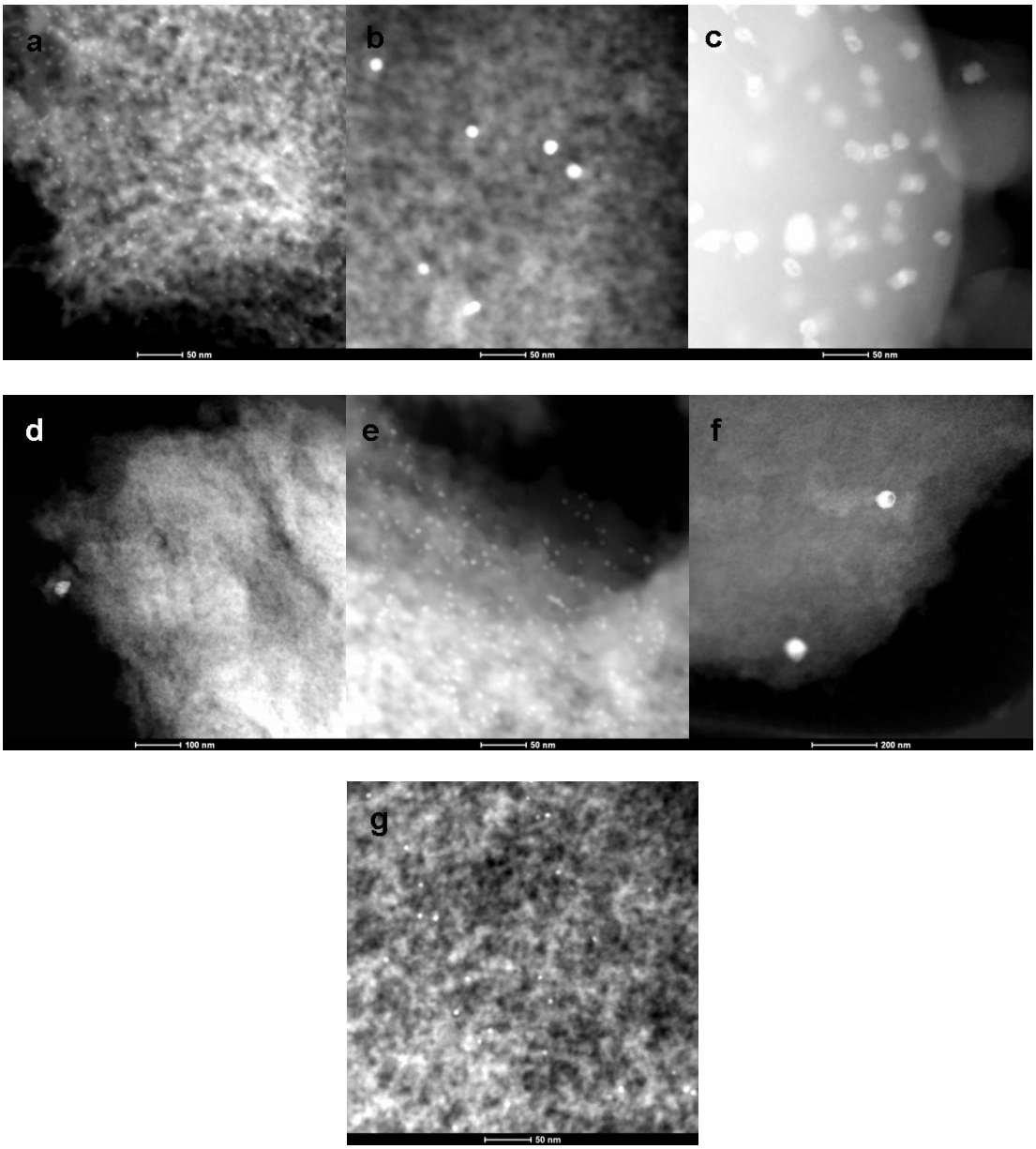
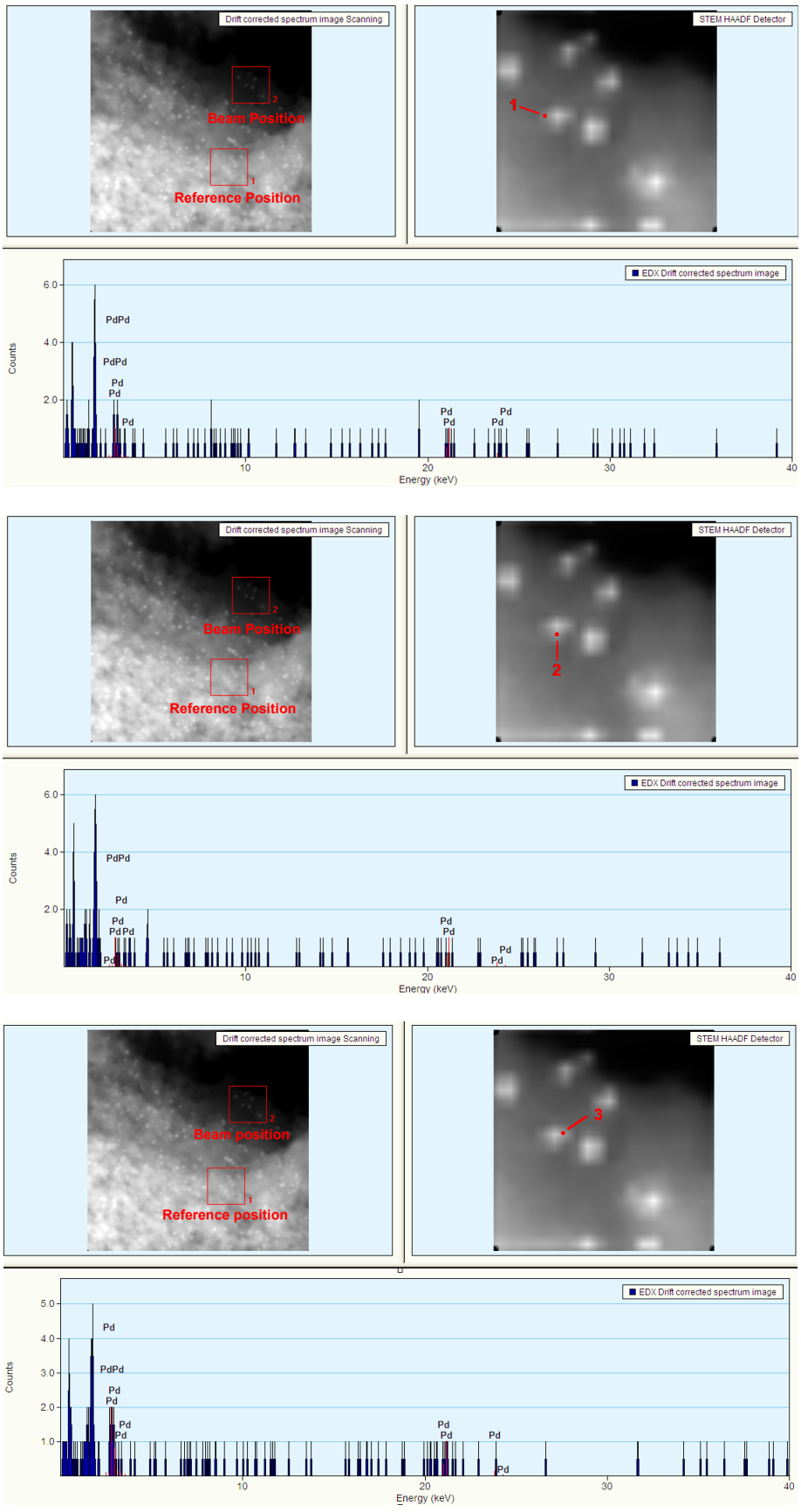


Figure S2. STEM-EDX images of the obtained samples: a) Pd/SiO_2-P-C ; b) Au/SiO_2-P-C; c) Pd-Au/SiO_2-P ; d) Pd-Au/SiO_2-C ; e) Pd-Au/SiO_2-P-C; f) Pd-Au/SiO_2; g) Pd-Au/SiO_2-P-C-1

6. Figure S3


Drift corrected spectrum image Scanning
Beam Position
Reference Position
STEM HAADF Detector
1
EDX Drift corrected spectrum image
Counts
Energy (keV)
Drift corrected spectrum image Scanning
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Reference Position
STEM HAADF Detector
2
EDX Drift corrected spectrum image
Counts
Energy (keV)
Drift corrected spectrum image Scanning
Beam position
Reference position
STEM HAADF Detector
3
EDX Drift corrected spectrum image
Counts
Energy (keV)

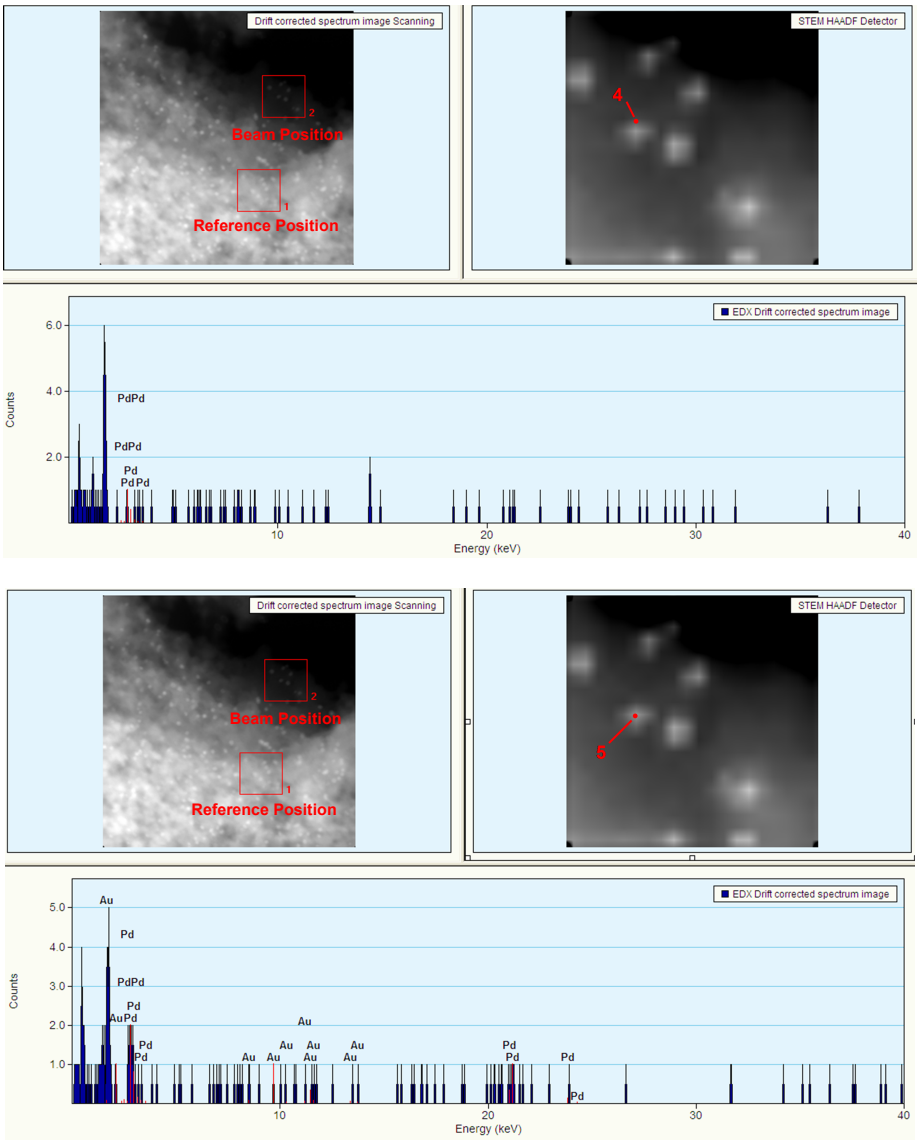

Figure S3. STEM-EDX element mapping of the Pd-Au particles in Pd-Au/SiO_2-P-C. The results showed that only palladium was observable around Pd-Au nano-particle, and a mixture of Pd and Au could be observed in the center of the nano-particle.

7. Figure. S4

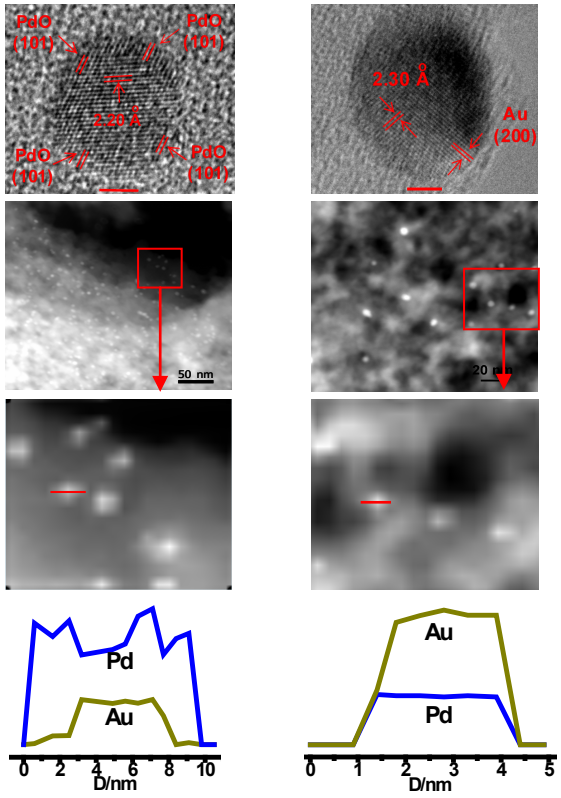

Figure S4. (a) HR-TEM image of Pd-Au/SiO_2-P-C (scale bar = 2 nm), (b) HAADF image of Pd-Au/SiO_2-P-C, (c) HAADF-STEM image of Pd-Au/SiO_2-P-C, (d) cross-sectional compositional line profiles of the line in (c), (e) HR-TEM image of Pd-Au/SiO_2-P-C-1 (scale bar = 2 nm), (f) HAADF image of Pd-Au/SiO_2-P-C-1, (g) HAADF-STEM image of Pd-Au/SiO_2-P-C-1 and (h) cross-sectional compositional line profiles of the line in (g).

8. Figure. S5

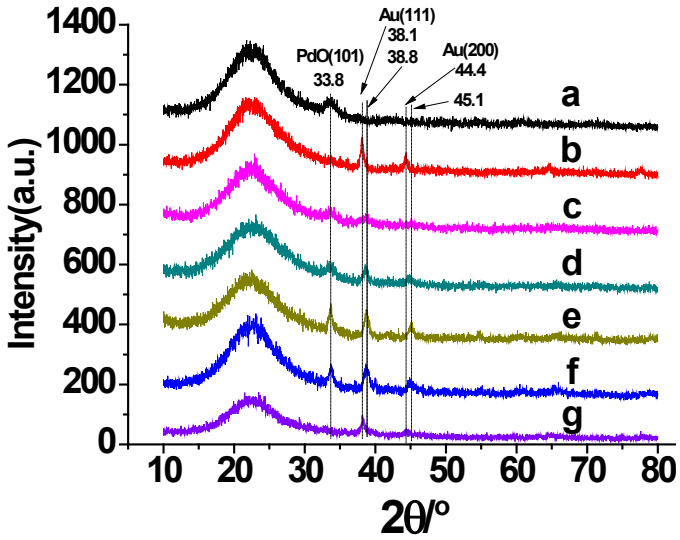

Figure S5. XRD spectra of the obtained samples: a) Pd/SiO_2-P-C ; b) Au/SiO_2-P-C; c) Pd-Au/SiO_2-P; d) Pd-Au/SiO_2-C; e) Pd-Au/SiO_2-P-C; f) Pd-Au/SiO_2; g) Pd-Au/SiO_2-P-C-1.

9. Figure. S6

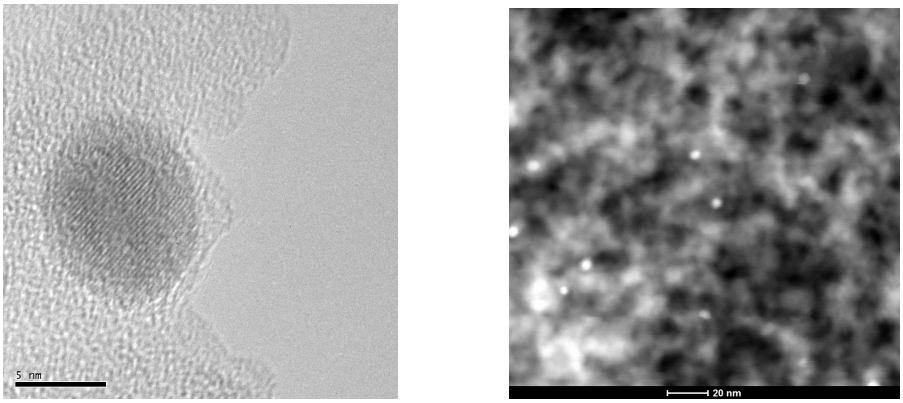

Figure S6. HRTEM and HADDF images of Pd-Au/SiO_2-P-C after reaction

10. Figure. S7

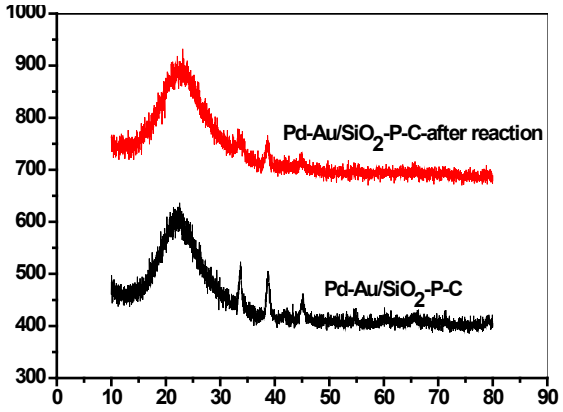

Figure S7. XRD spectra of Pd-Au/SiO_2-P-C and Pd-Au/SiO_2-P-C after reaction

11. Figure. S8

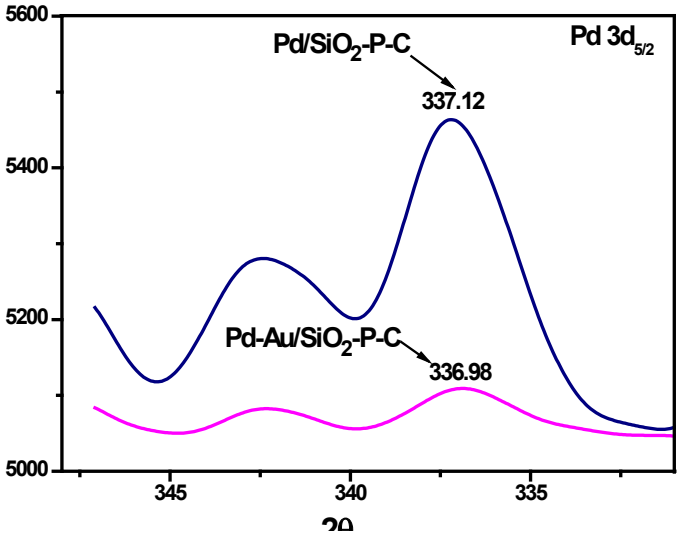

Figure S8. XPS spectra of Pd/SiO_2-P-C and $Pd\text{-}Au/SiO_2$-P-C

12. Characterization of compounds

N-benzylidenebenzenesulfonamide: (Table 2, Entry 1) (GC purity 98%); According to representative procedure 3.1, using benzenesulfonamide (157 mg, 1 mmol), toluene 3 mL, the title compound was obtained and purified by flash column chromatography eluting with petroleum ether (b.p. 60-90 ºC)/ethyl acetate (10 : 1), R_f = 0.23 to give a white solid (211 mg, 86%); ^{1}H NMR (400.1 MHz, $CDCl_3$): δ =7.48-7.58 (m, 4H), 7.61-7.65 (m, 2H), 7.93-7.95 (d, 2H), 8.01-8.03 (m, 2H), 9.07 (s, 1H); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ = 128.01, 129.15, 131.35, 132.32, 133.53, 135.04, 138.22, 170.59; MS (E.I., 70 eV) *m/z* (rel. int.) 245 (2), 195 (6), 194 (38), 155 (5), 118 (44), 117 (5), 104 (12), 92 (13), 91 (100), 90 (10), 89 (9), 77 (24), 65 (29), 63 (6), 51 (15), 39 (8), 28 (16), 18 (9).

N-benzylidene-4-methylbenzenesulfonamide: (Table 2, Entry 2) (GC purity 98%); According to representative procedure 3.1, using 4-methylbenzenesulfonamide (171 mg, 1 mmol), toluene 3 mL, the title compound was obtained and purified by flsah column chromatography eluting with petroleum ether (b.p. 60-90 ºC)/ethyl acetate (14 : 1), R_f = 0.21 to give a white solid (220 mg, 85%);^{1}H NMR (400.1 MHz, $CDCl_3$): δ =2.76, 7.33-7.38 (m, 2H), 7.48-7.52 (m, 3H), 7.61-7.65 (t, 1H), 7.93-7.95 (d, 2H), 8.09-8.11 (d, 1H), 9.10 (s, 1H); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ = 20.63, 126.31, 129.18, 131.31, 132.42, 133.62, 134.98, 136.50, 138.87, 170.65; MS (E.I., 70 eV) *m/z* (rel. int.) 259 (4), 195 (12), 194 (69), 178 (5), 155 (8), 137 (5), 119 (5), 118 (52), 117 (7), 104 (14), 92 (13), 92 (13), 91 (100), 90 (10), 89 (9), 77 (21), 65 (24), 63 (5), 51 (11), 39 (5).

N-benzylidene-2-methylbenzenesulfonamide: (Table 2, Entry 3) (GC purity 98%); According to representative procedure 3.1, using 2-methylbenzenesulfonamide (171 mg, 1 mmol), toluene 3 mL, the title compound was obtained and purified by flash column chromatography eluting with petroleum ether (b.p. 30-60 ºC)/ethyl acetate (16 : 1), R_f = 0.28 to give a white solid

(210 mg, 81%); ^{1}H NMR (400.1 MHz, $CDCl_3$): δ =2.44 (s, 3H), 7.33-7.36 (d, 2H), 7.46-7.51 (t, 2H), 7.62-7.64 (t,1H), 7.82-7.93 (m, 4H) 9.03 (s, 1H); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ = 21.61, 128.07, 129.10, 129.76, 131.28, 131.26, 132.39, 134.89, 135.16, 144.58, 170.08; MS (E.I., 70 eV) *m*/*z* (rel. int.) 259 (2), 195 (10), 194 (57), 178 (5), 155 (7), 137 (5), 118 (51), 104 (3), 91 (100), 77 (20), 65 (25), 51 (11), 39 (5).

N-benzylidene-4-methoxybenzenesulfonamide: (Table 2, Entry 4) (GC purity 98%); According to representative procedure 3.1, using -methoxybenzenesulfonamide (187 mg, 1 mmol), toluene 3 mL, the title compound was obtained and purified by flash column chromatography eluting with petroleum ether (b.p. 60-90 ºC)/ethyl acetate (11 : 1), R_f = 0.27 to give a white solid (223 mg, 81%); ^{1}H NMR (400.1 MHz, $CDCl_3$): δ = 3.15 (s, 3H), 7.52-7.56 (m, 2H), 7.65-7.69 (m, 1H), 7.96-7.99 (m, 2H), 9.05 (s, 1H); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ = 40.25, 128.94, 131.29, 132.09, 135.17, 171.65; MS (E.I., 70 eV) *m*/*z* (rel. int.) 183 (20), 118 (7), 105 (12), 104 (100), 103 (5), 80 (12), 79 (12), 78 (6), 77 (53), 76 (6), 65 (5), 51 (8).

N-benzylidene-4-methoxybenzenesulfonamide: (Table 2, Entry 5) (GC purity 98%); According to representative procedure 3.1, using -methoxybenzenesulfonamide (187 mg, 1 mmol), toluene 3 mL, the title compound was obtained and purified by flash column chromatography eluting with petroleum ether (b.p. 60-90 ºC)/ethyl acetate (10 : 1), R_f = 0.23 to give a white solid (223 mg, 81%); ^{1}H NMR (400.1 MHz, $CDCl_3$): δ = 3.87 (s, 3H), 7.00-7.03 (m, 2H), 7.46-7.51 (t, 2H), 7.59-7.64 (t, 1H),7.91-7.95 (m, 4H), 9.06 (s, 1H); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ =55.64, 114.38, 129.09, 129.49, 130.27, 131.20, 132.40, 134.79, 163.71, 169.57; MS (E.I., 70 eV) *m*/*z* (rel. int.) 275 (20), 173 (6), 172 (9), 171 (100), 123 (24), 108 (7), 92 (30), 78 (6), 77 (60), 64 (15), 63 (10), 51 (15), 50 (7).

N-benzylidene-4-chlorobenzenesulfonamide: (Table 2, Entry 6) (GC purity 98%); According to representative procedure 3.1, using 4-chlorobenzenesulfonamide (191 mg, 1

mmol), toluene 3 mL, the title compound was obtained and purified by flash column chromatography eluting with petroleum ether (b.p. 30-60 ºC)/ethyl acetate (15 : 1), R_f = 0.19 to give a white solid (251 mg, 90%); ^{1}H NMR (400.1 MHz, $CDCl_3$): δ =7.49-7.54 (m, 4H), 7.62-7.67 (t, 1H), 7.94-7.97 (t, 4H), 9.06 (s, 1H); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ = 129.22, 129.47, 131.43, 132.18, 135.26, 136.79, 140.26, 170.92; MS (E.I., 70 eV) *m*/*z* (rel. int.) 279 (28), 217 (6), 215 (19), 180 (5), 178 (5), 177 (37), 176 (12), 175 (97), 152 (5), 114 (34), 113 (34), 112 (17), 111 (100), 104 (33), 77 (31), 76 (10), 75 (28), 74 (5), 51 (16), 50 (9).

N-benzylidene-4-bromobenzenesulfonamide: (Table 2, Entry 7) (GC purity 98%); According to representative procedure 3.1, using 4-bromobenzenesulfonamide (243 mg, 1 mmol), toluene 3 mL the title compound was obtained and purified by flash column chromatography eluting with petroleum ether (b.p. 30-60 ºC)/ethyl acetate (10 : 1), R_f = 0.20 to give a white solid (246 mg, 93%); ^{1}H NMR (400.1 MHz, $CDCl_3$): δ =7.49-7.54 (m, 4H), 7.62-7.67 (t, 1H), 7.94-7.97 (T, 4H), 9.06 (S,1H); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ =7.49-7.52 (t, 2H), 7.62-7.71 (m, 3H), 7.86-7.95 (m, 4H), 9.06 (s, 1H); ^{3}C NMR (100.6 MHz, $CDCl_3$): δ = 128.78, 129.24, 129.54, 131.45, 132.17, 132.48, 135.27, 137.31, 170.96; MS (E.I., 70 eV) *m*/*z* (rel. int.) 324 (28), 267 (18), 260 (5), 259 (18), 222 (5), 221 (12), 220 (93), 219 (12), 218 (90), 180 (7), 158 (12), 157 (98), 156 (13), 155 (100), 152 (7), 141 (6), 104 (47), 77 (50), 76 (36), 75 (28), 74 (9), 51 (22), 50 (17).

4-methoxy-N-(4-methylbenzylidene)benzenesulfonamide: (Table 2, Entry 8) (GC purity 98%); According to representative procedure 3.1, using 4-methoxybenzenesulfonamide (187 mg, 1 mmol), toluene 3 mL, the title compound was obtained and purified by flash column chromatography eluting with petroleum ether (b.p. 30-60 ºC)/ethyl acetate (15 : 1), R_f = 0.25 to give a white solid (246 mg, 93%); ^{1}H NMR (400.1 MHz, $CDCl_3$): δ =2.43 (s, 3H), 3.87 (s, 3H), 6.99-7.02 (m, 2H), 7.27-7.30 (d, 2H), 7.80-7.82 (d, 2H), 7.92-7.94 (d, 2H); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ = 21.93, 55.63, 114.35, 129.89, 130.18, 131.33, 146.19, 163.57, 169.41; MS (E.I., 70 eV) *m*/*z* (rel. int.) 289 (20), 173 (5), 172 (11), 171 (100), 156 (5), 123

(26), 108 (10), 107 (56), 92 (29), 91 (15), 89 (7), 77 (41), 65 (16), 64 (14), 63 (11), 39 (6), 28 (10), 18 (6).

4-chloro-N-(4-methylbenzylidene)benzenesulfonamide: (Table 2, Entry 9) (GC purity 98%); According to representative procedure 3.1, using 4-chlorobenzenesulfonamide (191 mg, 1 mmol), p-xylene 3 mL, the title compound was obtained and purified by flash column chromatography eluting with petroleum ether (b.p. 60-90 ºC)/ethyl acetate (10 : 1), R_f = 0.23 to give a white solid (261 mg, 89%); ^{1}H NMR (400.1 MHz, $CDCl_3$): δ = 2.62 (s, 3H), 7.26-7.32 (t, 2H), 7.48-7.54 (m, 3H), 7.94-8.02 (m, 3H), 9.37 (s, 1H); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ = 19.69, 126.69, 129.40, 129.43, 130.82, 131.67, 134.92, 137.09, 142.48, 169.47; MS (E.I., 70 eV) *m*/*z* (rel. int.) 293 (21), 229 (12), 228 (6), 177 (21), 176 (7), 175 (51), 119 (10), 118 (100), 117 (6), 114 (9), 113 (32), 112 (25), 111 (100), 91 (38), 90 (7), 89 (12), 76 (7), 65 (26), 63 (7), 51 (7), 39 (6), 28 (10).

N-(4-methylbenzylidene)benzenesulfonamide: (Table 2, Entry 9) (GC purity 98%); According to representative procedure 3.1, using benzenesulfonamide (157 mg, 1 mmol), p-xylene 3 mL, the title compound was obtained and purified by flash column chromatography eluting with petroleum ether (b.p. 60-90 ºC)/ethyl acetate (13 : 1), R_f = 0.23 to give a white solid (218 mg, 84%); ^{1}H NMR (400.1 MHz, $CDCl_3$): δ = 2.62 (s, 3H), 7.26-7.31 (t, 2H), 7.47-7.51 (t, 1H), 7.54-7.58 (t, 2H), 7.61-7.66 (t, 1H), 8.01-8.04 (d, 3H), 9.37 (s, 1H); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ = 18.67, 126.61, 127.92, 129.09, 130.71, 131.57, 133.41, 134.67, 142.36, 169.11; MS (E.I., 70 eV) *m*/*z* (rel. int.) 259 (16), 195 (5), 194 (7), 142 (7), 141 (32), 119 (7), 118 (76), 116 (5), 89 (7), 78 (20), 77 (100), 65 (15), 51 (16).

(E)-N-(4-chlorobenzylidene)benzenesulfonamide: (Table 2, Entry 11) (GC purity 98%); According to representative procedure 3.1, using benzenesulfonamide (157 mg, 1

mmol), 1-chloro-4-methylbenzene 3 g, the title compound was obtained and purified by flash column chromatography eluting with petroleum ether (b.p. 60-90 ºC)/ethyl acetate (15 : 1), R_f = 0.22 to give a white solid (231 mg, 74%); ^{1}H NMR (400.1 MHz, $CDCl_3$): δ = 7.46-7.49 (d, 2H), 7.54-7.59 (t, 2H), 7.63-7.67 (t, 1H), 7.86-7.89 (d, 2H), 8.00-8.02 (d, 2H), 9.03 (s, 1H); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ = 128.05, 129.17, 129.63, 130.80, 132.39, 133.67, 138.03, 141.60, 169.05; MS (E.I., 70 eV) *m*/*z* (rel. int.) 279 (7), 207 (9), 142 (9), 141 (51), 140 (9), 138 (20), 111 (12), 78 (13), 77 (1000, 75 (14), 51 (20), 50 (8), 32 (120), 28 (38).

N-(4-chlorobenzylidene)-4-methoxybenzenesulfonamide: (Table 2, Entry 12) (GC purity 98%); According to representative procedure 3.1, using 4-methoxybenzenesulfonamide (187 mg, 1 mmol), 1-chloro-4-methylbenzene 3 g, the title compound was obtained and purified by flash column chromatography eluting with petroleum ether (b.p. 60-90 ºC)/ethyl acetate (11 : 1), R_f = 0.23 to give a white solid (250 mg, 81%); ^{1}H NMR (400.1 MHz, $CDCl_3$): δ = 3.88 (s, 3H), 7.01-7.03, (d, 2H), 7.45-7.48 (d, 2H), 7.85-7.95 (m, 4H), 8.97 (s, 1H); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ = 55.69, 109.74, 114.47, 129.30, 129.57, 130.35, 130.89, 132.29, 141.30, 168.06; MS (E.I., 70 eV) *m*/*z* (rel. int.) 309 (21), 173 (7), 172 (9), 171 (100), 108 (5), 107 (41), 92 (20), 77 (31), 75 (10), 75 (10), 64 (10), 63 (7), 50 (5).

4-chloro-N-(4-chlorobenzylidene)benzenesulfonamide: (Table 2, Entry 13) (GC purity 98%); According to representative procedure 3.1, using 4-chlorobenzenesulfonamide (191 mg, 1 mmol), 1-chloro-4-methylbenzene 3 g, the title compound was obtained and purified by flash column chromatography eluting with petroleum ether (b.p. 60-90 ºC)/ethyl acetate (14 : 1), R_f = 0.19 to give a white solid (231 mg, 74%); ^{1}H NMR (400.1 MHz, $CDCl_3$): δ = 7.47-7.54 (m, 4H), 7.86-7.96 (m, 4H), 9.02 (s, 1H); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ = 129.53, 129.72, 130.64, 132.47, 136.59, 140.44, 141.84, 169.41; MS (E.I., 70 eV) *m*/*z* (rel. int.) 314 (6), 312 (9), 177 (27), 176 (9), 175 (70), 140 (8), 138 (22), 113 (33), 112 (14), 111 (100), 76 (9), 75 (40), 51 (6), 28 (7).