Electronic Supplementary information (ESI) for

Selective imines formation from alcohols and amines catalyzed by polymer incarcerated gold/palladium alloy nanoparticles with molecular oxygen as oxidant

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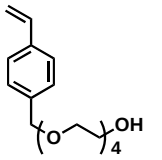
1. General experimental

Reactions were monitored with analytical thin-layer chromatography (TLC) on silica gel 60 F_{254} plates and visualized under UV (254 nm) and/or by staining with $KMNO_4$. NMR spectra were recorded with JEOL JMN-LA400, 500 or 600 spectrometers. Chemical shifts are given in parts per million, referenced to the solvent peak of $CDCl_3$, defined at 77.2 ppm (^{13}C NMR) and 7.24 ppm (^{1}H NMR) or peak of MeOH-d_4, defined at 49.15 ppm (^{13}C NMR) and 3.31 ppm (^{1}H NMR). The structures of the known compounds were confirmed by comparison with commercially available compounds or data shown in literature. Inductively coupled plasma-atomic emission spectrometry (ICP-AES) analysis was performed on Shimadzu ICPS-7510 equipment. STEM/EDS images were obtained using a JEOL JEM-2100F instrument operated at 200 kV. All STEM specimens were prepared by placing a drop of the solution on carbon-coated copper grids and allowed to dry in air (without staining). $NaBH_4$ was purchased from Wako Pure Chemical Company and recrystallized from diglyme by heating according to the literature[1] and stored in a glove box. It is important to manipulate all operations under Ar atmosphere during recrystallization. Activity of catalyst and reproducibility are highly influenced by the purity and condition of $NaBH_4$ in the course of catalyst preparation. Ketjen black (CARBON ECP) was purchased from Lion Corporation. Alcohols were purified by recrystallization or distillation following the normal procedures, and amines were purified by distillation.

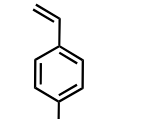
2. Catalysts Preparation[2]

Preparation of 2-(2-(2-(2-(4-vinylbenzyloxy)ethoxy)ethoxy)ethoxy)ethanol: To sodium hydride (60% in mineral oil, 3.34 g, 83.54 mmol, 1.50 equiv) suspended in THF (150 mL), tetraethyleneglycol (14.4 mL, 83.54 mmol, 1.50 equiv) was added at 0 °C. After the reaction mixture was stirred for 1 h at rt, 1-(chloromethyl)-4-vinylbenzene (7.9 mL, 55.69 mmol, 1 equiv) was added and the mixture was further stirred for 12 h. The mixture was diluted with ethyl acetate (200 mL). Saturated aqueous ammonium chloride (140 mL) was added to quench the reaction and the aqueous layer was extracted with ethyl acetate (3 x 100 mL). The combined organic layers were dried over sodium sulfate and the solvent was removed *in vacuo*. The residue was purified by flash chromatography to afford (2-(2-(2-(4-vinylbenzyloxy)ethoxy)ethoxy)ethoxy)ethanol ether (13.7 g, 78%). **^{1}H NMR (500 MHz, $CDCl_3$, 20 °C): δ** (ppm) 2.55-2.59 (m, 1H), 3.59-3.73 (m, 16H), 4.55 (s, 2H), 5.25 (d, 1H, J = 6.4 Hz), 5.53 (d, 1H, J = 18 Hz), 6.71 (dd, 1H, J = 11.0, 17.9 Hz), 7.22-7.27 (m, 3H), 7.31-7.39 (m, 2H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 61.8, 69.5, 70.5, 70.69, 70.74, 72.6, 73.0, 113.8, 126.3, 128.0, 136.0, 137.1, 138.0.

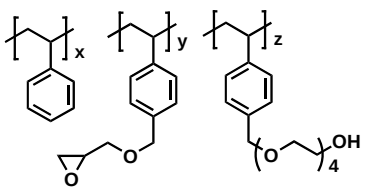


Chemical Formula: $C_{17}H_{26}O_5$
Exact Mass: 310,18

Preparation of 4-vinylbenzyl glycidyl ether: To sodium hydride (60% in mineral oil, 13.1 g, 327.60 mmol, 2.00 equiv) suspended in DMF (300 mL), glycidol (27.15 mL, 459.1 mmol, 2.50 equiv) and 1-(chloromethyl)-4-vinylbenzene (23.3 mL, 163.80 mmol, 1.00 equiv) were added at 0 °C. After the mixture was further stirred for 4 h, the mixture was diluted with ethyl acetate (400 mL). Saturated aqueous ammonium chloride (200 mL) was added to quench the reaction and the aqueous layer was extracted with ethyl acetate (3 x 100 mL). The combined organic layers were dried over sodium sulfate and the solvent was removed *in vacuo*. The residue was purified by flash chromatography to afford 4-Vinylbenzyl glycidyl ether (23 g, 74%). **^{1}H NMR (500 MHz, $CDCl_3$, 20 °C): δ** (ppm) 2.60 (dd, 1H, J = 2.8, 4.8 Hz), 2.78 (dd, 1H, J = 4.0, 4.8 Hz), 3.17 (m, 1H), 3.42 (dd, 1H, J = 5.6, 11.2 Hz), 3.74 (dd, 1H, J = 2.8, 11.2 Hz), 4.56 (dd, 2H, J = 10.8, 17.6 Hz), 7.30 (d, 2H, J = 8.0 Hz), 7.39 (d, 2H, J = 8.0 Hz); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 40.2, 50.7, 70.7, 72.9, 113.8, 126.2, 127.9, 136.4, 137.0, 137.4.


Chemical Formula: $C_{12}H_{14}O_2$
Exact Mass: 190,10

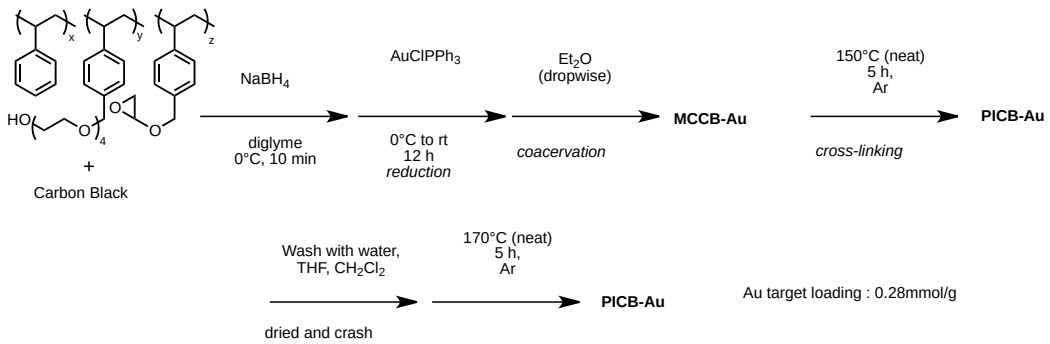
Preparation of copolymer A: Styrene (3 g, 28.80 mmol, 1.00 equiv), 4-vinylbenzyl glycidyl ether (5.50 g, 28.80 mmol, 1.00 equiv), 2-(2-(2-(2-(4-vinylbenzyloxy)ethoxy)ethoxy)ethoxy)ethanol (8.95 g, 28.80 mmol, 1.00 equiv) and 2,2'-azobis(4-methoxy)-2,4-dimethylvaleronitrile (242 mg, 0.864 mmol, 0.30 equiv) were combined in chloroform (16.0 mL). The mixture was stirred for 72 h at room temperature. The resulting polymer solution was slowly poured into diethyl ether. The solvent was removed by decantation and the residue was dissolved again in THF. The polymer solution was slowly poured into diethyl ether again. The same procedure was repeated a total of three times. The precipitated polymer was collected and washed with ether several times and dried *in vacuo* to



polymer 1 (x/y/z = 1:1:1)

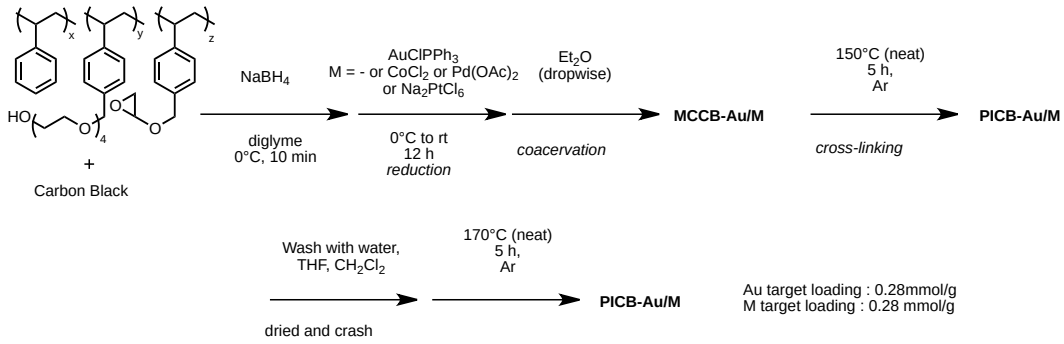
afford the desired copolymer (**1**, 10.52 g, 63% yield). The molar ratio of the components was determined by ^{1}H NMR analysis (x: y: z = 34: 32: 34).

Preparation of PICB-Au Catalyst:



To a solution of copolymer **1** (500 mg) in diglyme (32 mL) at rt were added at 0 °C Ketjen black (500 mg), $NaBH_4$ (32.1 mg, 0.85 mmol, 3.00 equiv) and a solution of PPh_3AuCl (140 mg, 0.28 mmol, 1.00 equiv) in 10 mL of diglyme. After stirring for over night at rt, diethyl ether (160 mL) was added dropwise with stirring for microencapsulation. The resulting MCCB catalyst was filtered, washed several times with diethyl ether and dried under *vacuo*. Next, the catalyst capsules were heated (no stirred) at 150 °C for 5 h under an argon atmosphere. The PICB catalyst was then washed with water, THF, and DCM. After dried *in vacuo*, the catalyst was heated (no stirred) at 170 °C for 5 h under an argon atmosphere to give polymer incarcerated carbon black gold (PICB-Au).

Preparation of PICB-Au/Co Catalyst:



To a solution of copolymer **1** (500 mg) in diglyme (32 mL) at rt were added at 0 °C Ketjen black (500 mg), $NaBH_4$ (96.4 mg or 160 mg, 2.55 or 4.25 mmol, 3 equiv for Au, 6 equiv for Co, Fe or Ni, 12 equiv for Pd or Pt) and a solution of metal sources PPh_3AuCl (140 mg, 0.28 mmol, 1.00 equiv) for Au, NiF_2 (27.4 mg, 0.28 mmol, 1.00 equiv) for Ni, $CoCl_2$ (36.8 mg, 0.28 mmol, 1.00 equiv) for Co, $FeCl_2$ (36 mg, 0.28 mmol, 1.00 equiv) for Fe, $Pd(OAc)_2$ (63.4 mg, 0.28 mmol, 1.00 equiv) for Pd, $Na_2PtCl_6 \bullet 6H_2O$ (160 mg, 0.28 mmol, 1.00 equiv) for Pt in 10 mL of diglyme. After stirring for over night at rt, diethyl ether (160 mL) was added dropwise with stirring for microencapsulation. The resulting MCCB catalyst was filtered, washed several times with diethyl ether and dried under *vacuo*. Next, the catalyst capsules were heated (no stirred) at 150 °C for 5 h under an argon atmosphere. The PICB catalyst was then washed with water, THF, and DCM. After dried *in vacuo*, the catalyst was heated (no stirred) at 170 °C for 5 h under an argon atmosphere to give polymer incarcerated carbon black gold / Metals (PICB-Au/M).

°C for for 5 h under an argon atmosphere to give polymer incarcerated carbon black gold / Metals (PICB-Au/Co).

Preparation of the sample for ICP-analysis: 10 mg of catalyst was dissolved in 1 ml of H_2SO_4 (conc.) and heated at 200°C, then HNO_3 was added slowly and the solution was heated until remove nitric acid (water mmq can be added). After 1ml of aqua regia was added for dissolved all metal species and the solutions was completed with water mmq (50 ml).

Table S1 : Loading of catalysts determined by ICP

Catalyst	Metals salts	Loading Au (mmol/mg)	Loading M (mmol/mg)
PICB-Au	-	0.251 or 0.254	-
PICB-Au/Co	$CoCl_2$	0.279 or 212	0.267 or 0.203
PICB-Au/Pd	$Pd(OAc)_2$	0.226 or 0.228	0.235 or 0.288
PICB-Au/Pt	$Na_2PtCl_66H_2O$	0.226	0.235

PICB-Au/Pd STEM-EDS analysis

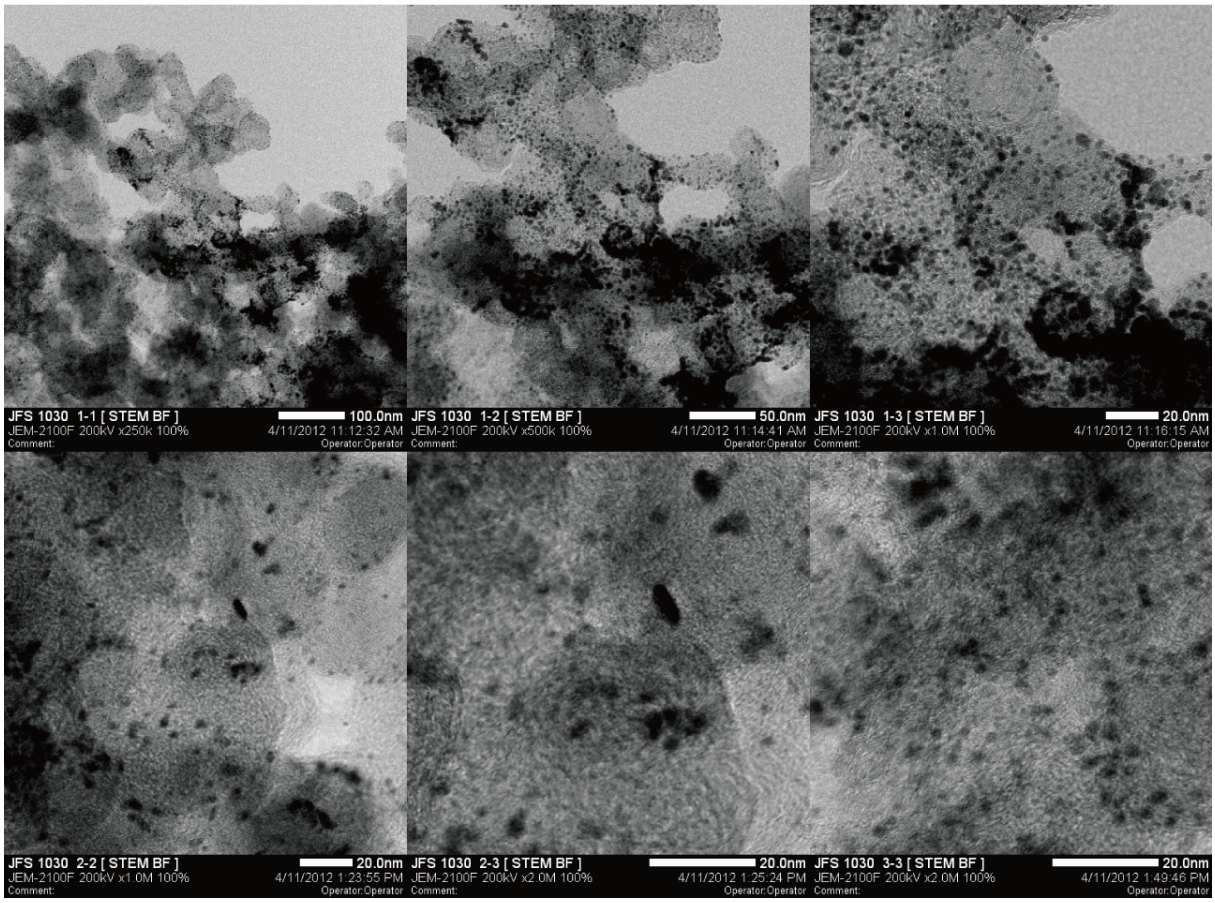


Figure S 1. Typical STEM Images of PICB-Au/Pd

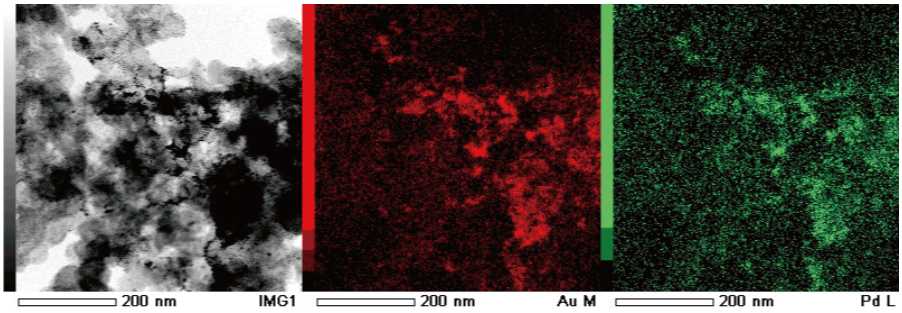


Figure S 2. EDS Mapping of PICB-Au/Pd

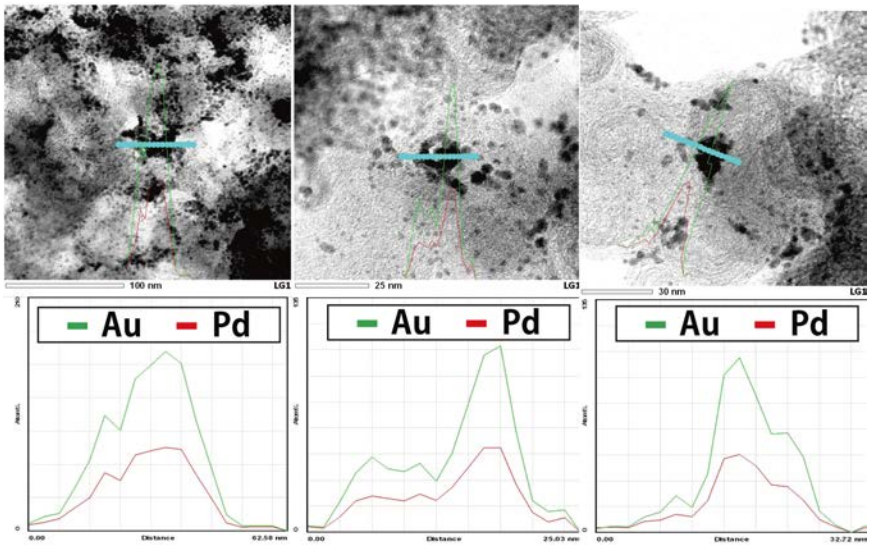


Figure S 3. EDS Line Analysis of PICB-Au/Pd

PICB-Au/Pd recovery after use STEM-EDS analysis

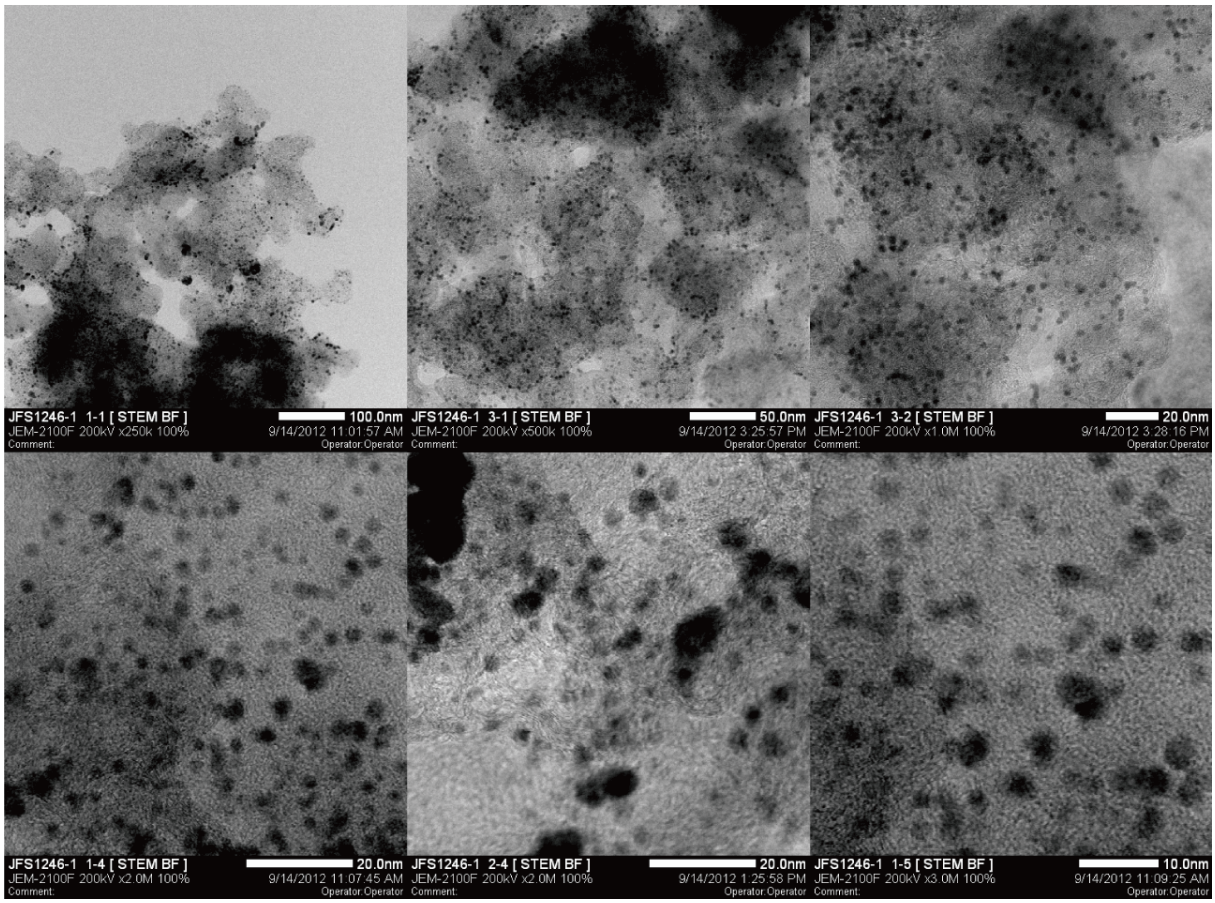


Figure S 4. Typical STEM Images of PICB-Au/Pd after use.

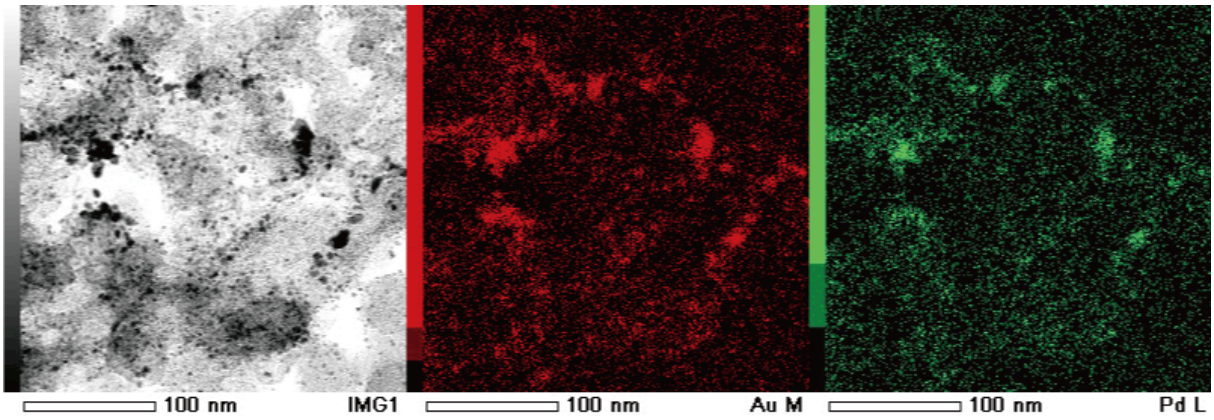


Figure S 5. EDS Mapping of PICB-Au/Pd after use.

PICB-Au/Pd recovery after treatment STEM-EDS analysis

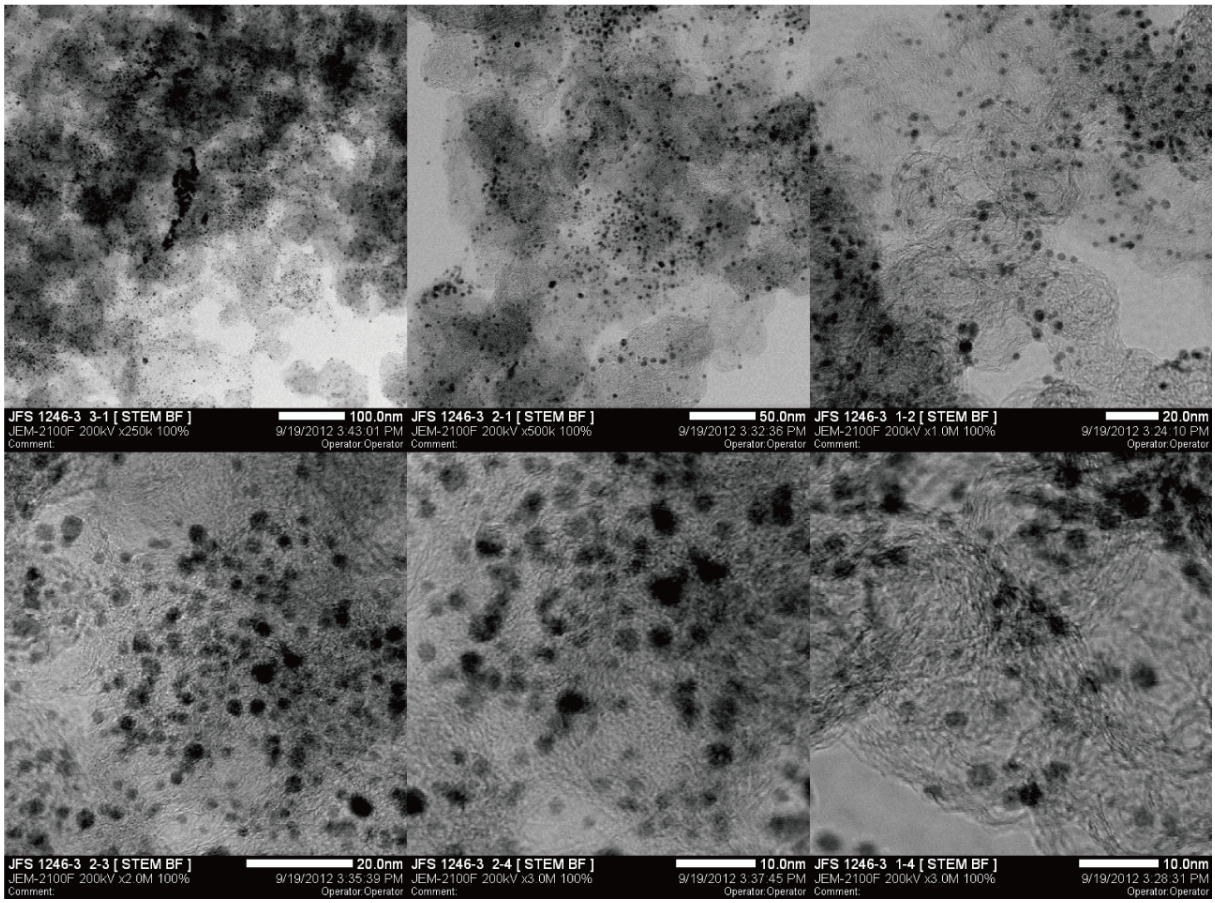


Figure S 6. Typical STEM Images of PICB-Au/Pd after treatment.

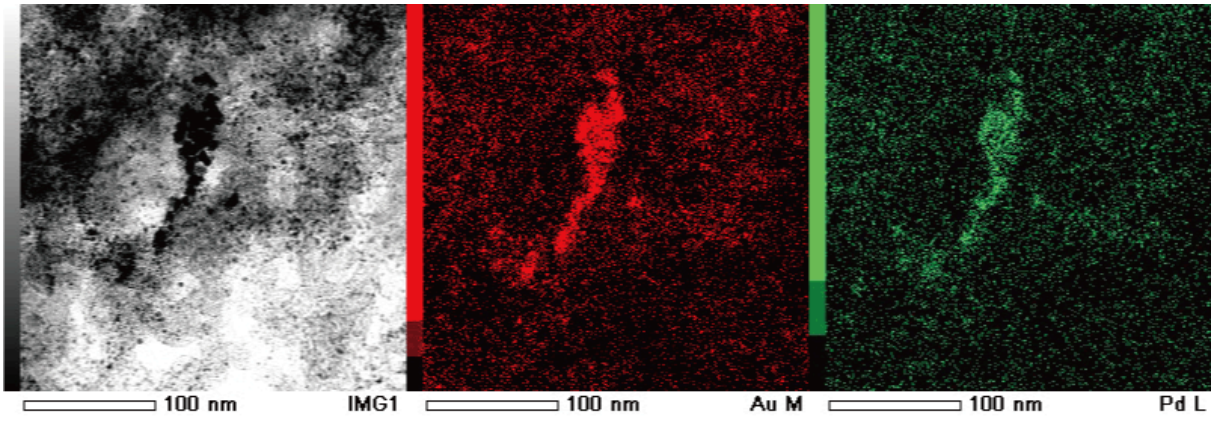


Figure S 7. EDS Mapping of PICB-Au/Pd after treatment.

3. General procedure

GP A: General procedure for imine formation from alcohol and amine using PI-CB Au/Pd and molecular oxygen as terminal oxidant

Alcohol (0.368 mmol, 1 equiv), amine (0.368 mmol, 1 equiv), sodium hydroxide (0.095 mmol, 0.25 equiv) and PICB-Au (0.0057 mmol, 1.5 mol%) were combined in mixture of THF/CF_3CH_2OH (9:1, 0.5 mL). The mixture was stirred at 30 °C for 12 h under oxygen atmosphere. The catalyst was separated by filtration and washed with CH_2Cl_2 (10 mL) and water. Aqueous NH_4Cl solution (sat, 10 mL) was added and the aqueous layer was extracted with dichloromethane (3 x 20 mL). The combined organic layers were dried over sodium sulfate and the solvent was removed *in vacuo* to afford the corresponded imine as a pure product.

Recovery and reuse catalyst.

4-methylbenzyl alcohol (200 mg, 1.64 mmol, 1 equiv), benzylamine (180 μL, 1.64 mmol, 1 equiv), NaOH (16.4 mg, 0.41 mmol, 0.25 equiv), PICB-Au/Co (0.21 mmol/g, 78.0 mg, 1.5 mol%), THF/CF_3CH_2OH (9:1, 2.2 ml) were combined in a round-bottomed flask. After the mixture was stirred by a stirring bar and a magnetic stirrer for 12 h under O_2 atmosphere at room temperature, the catalyst was collected by filtration and washed with THF and water using KIRIYAMAROHTO® funnel. The aqueous layer was washed with diethylether (20 mL). The yield was determined by ^{1}H NMR analysis with reference to an internal standard (IS = 1,1,2,2-tetracholoroethane). After determining the yield, the solvents of both aqueous and organic layers were removed *in vacuo.*

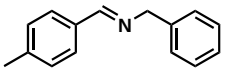
Reactivation process: The filtered catalysts was dried *in vacuo* and heated at 170 °C for 5 h without solvent under argon conditions and 71 mg of catalyst was collected. (About 6-7 mg of catalyst was trapped by filtration paper in every use.)

Leaching Test.

To crude sample (after filtration on celite pad to remove the catalyst) were added sulfuric acid and aqua regia then the volume of the residue was adjusted to 50 mL using water to give a sample for ICP analyses for the measurement of the leaching of gold and palladium.

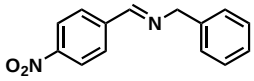
4. Imine Products – Table 2

- ***N*-Benzyl-4-methylbenzaldehyde imine 3aa:** Following the above general procedure A with 4-methylbenzylalcohol (45 mg, 0.368 mmol, 1 equiv) and benzylamine (40 μL, 0.368 mmol, 1 equiv). Imine **3aa** (89 mg, 95%) can be isolated as pure product and as colorless oil. **^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** 8.21 (s, 1H), 7.55 (d, *J* = 8 Hz, 2H), 7.23-7.16 (m, 4H), 7.15-7.11 (m, 1H), 7.08 (d, *J* = 8 Hz, 2H), 4.67 (s, 2H), 2.24 (s, 3H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 162.1, 141.2, 139.6, 129.5, 128.6, 128.4, 128.1, 127.1, 65.1, 21.7. This is a known compound and the spectral data are identical to those reported in literature.[3]



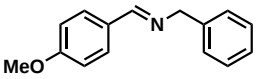
Chemical Formula: $C_{15}H_{15}N$
Exact Mass: 209,1204

- ***N*-Benzyl-4-nitrobenzaldehyde imine 3ba:** Following the above general procedure A with 4-nitrobenzylalcohol (58 mg, 0.368 mmol, 1 equiv) and benzylamine (40 μL, 0.368 mmol, 1 equiv). Imine **3ba** (59 mg, 65%) can be isolated as pure product and as colorless oil (*E*/*Z* = 85:15). **^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** 8.39 (s, 1H, *Z*), 8.32 (s, 1H, *E*), 7.81 (d, *J* = 7 Hz, 2H, *Z*), 7.72 (d, *J* = 7 Hz, 2H, *E*), 7.35-7.30 (m, 4H, *Z*&*E*), 7.26-7.23 (m, 1H, *Z*&*E*), 6.92 (d, *J* = 7 Hz, 2H, *E*), 4.82 (s, 2H, *Z*), 4.78 (s, 2H, *E*); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 161.8, 130.3, 129.6, 128.9, 128.9, 128.4, 127.8, 127.3, 114.4, 55.8. This is a known compound and the spectral data are identical to those reported in literature.[4]



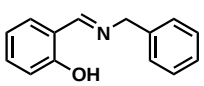
Chemical Formula: $C_{14}H_{12}N_2O_2$
Exact Mass: 240,0899

- ***N*-Benzyl-4-methoxybenzaldehyde imine 3ca:** Following the above general procedure A with 4-methoxybenzylalcohol (52 mg, 0.368 mmol, 1 equiv) and benzylamine (40 μL, 0.368 mmol, 1 equiv). Imine **3ca** (78 mg, 92%) can be isolated as pure product and as colorless oil (*E*/*Z* = 85:25).**^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** 8.29 (*Z,* s, 1H), 8.22 (*E*, s, 1H) 7.73 (*Z,* d, *J* = 8.5 Hz, 2H), 7.64 (*E,* d, *J* = 8.5 Hz, 2H), 7.27-7.21 (*E*&*Z*, m, 3H), 7.19-7-11 (*E*&*Z*, m, 1H), 6.89 (*Z,* d, *J* = 7 Hz, 2H), 6.83 (*E,* d, *J* = 7 Hz, 2H), 4.73 (*Z,* s, 2H), 4.69 (*E*, s, 2H), 3.76 (*Z*, s, 3H), 3.73 (*E*, s, 3H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 161.4, 139.7, 130.9, 123.0, 129.3, 128.6, 128.1, 127.0, 114.1, 65.1, 55.5. This is a known compound and the spectral data are identical to those reported in literature.[4]



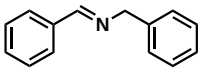
Chemical Formula: $C_{15}H_{15}NO$
Exact Mass: 225,1154

- ***N*-Benzyl-2-hydroxybenzaldehyde imine 3da:** Following the above general procedure A with 2-hydroxybenzylalcohol (47 mg, 0.379 mmol, 1 equiv) and benzylamine (41 μL, 0.379 mmol, 1 equiv). Imine **3da** (74 mg, 93%) can be isolated as pure product and as a yellow oil. **^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** 8.34 (s, 1H), 7.37-7.33 (m, 1H), 7.30-7.26 (m, 2H), 2.26-2.21 (m, 3H), 7.21-7.17 (m, 1H), 6.90 (bd, *J* = 8 Hz, 1H), 6.81 (bt, *J* = 7 Hz, 1H), 4.75 (s, 2H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 166.0, 161.5, 138.6, 132.7, 131.8, 129.1, 129.0, 128.1, 127.7, 119.2, 119.0, 117.4, 63.6. This is a known compound and the spectral data are identical to those reported in literature.[5]



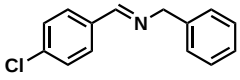
Chemical Formula: $C_{14}H_{13}NO$
Exact Mass: 211,0997

- *N*-Benzyl-benzaldehyde imine 3ea: Following the above general procedure A with benzylalcohol (41 μL, 0.368 mmol, 1 equiv) and benzylamine (40 μL, 0.368 mmol, 1 equiv). Imine **3ea** (67 mg, 91%) can be isolated as pure product and as colorless oil.[1] **^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** δ (ppm) 8.30 (s, 1H), 7.68 (d, *J* = 7 Hz, 2H), 7.35-7.29 (m, 3H), 7.26-7.22 (m, 3H), 7.18-7.12 (m, 1H), 4.72 (s, 2H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 162.4, 139.7, 136.5, 132.5, 129.5, 129.0, 128.9, 128.7, 128.4, 127.4, 65.4. This is a known compound and the spectral data are identical to those reported in literature.[6]



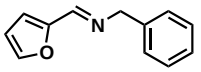
Chemical Formula: $C_{14}H_{13}N$
Exact Mass: 195,1048

- *N*-Benzyl-4-chlorobenzaldehyde imine 3fa: Following the above general procedure A with 4- chlorobenzyl alcohol (54 mg, 0.379 mmol, 1 equiv) and benzylamine (41 μL, 0. 379 mmol, 1 equiv). Imine **3fa** (84 mg, 97%) can be isolated as pure product and as colorless oil.**^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** δ (ppm) 8.30 (s, 1H), 7.71-7.67 (m, 1H), 7.34-7.29 (m, 3H), 7.36-7.23 (m, 3H), 7.28-7.14 (m, 1H), 4.73 (s, 1H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 162.4, 139.7, 131.2, 129.0, 128.9, 128.7, 128.4, 127.4, 65.5. This is a known compound and the spectral data are identical to those reported in literature.[4]



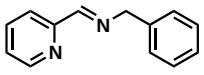
Chemical Formula: $C_{14}H_{12}ClN$
Exact Mass: 229,0658

- *N*-benzyl-furfuryladehyde imine 3ha: Following the above general procedure A with furfurylalcohol (37 mg, 0.377 mmol, 1 equiv) and benzylamine (40 μL, 0.377 mmol, 1 equiv). Imine **3ha** (59 mg, 84%) can be isolated as pure product and as a yellow oil (*E*/*Z* = 80:20). **^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** 8.31 (s, 1H, *Z*), 8.01 (s, 1H, *E*), 7.70 (d, *J* = 6 Hz, 2H, *Z*), 7.43 (s, 1H, *E*), 7.33 (d, *J* = 6.5 Hz, 2H, *E*), 7.27-7.22 (m, 4H, *Z*&*E*), 7.19-7.15 (m, 1H, *Z*&*E*), 6.69 (d, *J* = 4 Hz, 1H), 6.40-6.38 (m, 1H), 4.74 (s, 2H, *Z*), 4.70 (s, 2H, *E*); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 150.8, 145.2, 139.0, 129.0, 128.7, 127.5, 114.6, 112.1, 65.5. This is a known compound and the spectral data are identical to those reported in literature.[4]



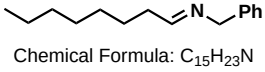
Chemical Formula: $C_{12}H_{11}NO$
Exact Mass: 185,0841

- *N*-benzyl-picolinaldehyde imine 3ia: Following the above general procedure A with pyridin-2-ylmethanol (41 mg, 0.377 mmol, 1 equiv) and benzylamine (40 μL, 0.377 mmol, 1 equiv). Imine **3ia** (68 mg, 92%) can be isolated as pure product and as a yellow oil (*E*/*Z* = 93:7). **^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** 8.44 (d, *J* = 4 Hz, 1H), 8.30 (bs, 1H), 8.15 (d, *J* = 8.5 Hz, 1H), 7.77 (td, *J* = 7.5 and 1 Hz, 1H), 7.35-7.32 (m, 1H), 7.30-7.24 (m, 4H), 7.22-7.17 (m, 1H), 4.59 (d, *J* = 6 Hz, 2H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 164.7, 150.3, 148.5, 138.6, 137.8, 129.1, 128.3, 127.9, 126.6, 122.8, 43.9. This is a known compound and the spectral data are identical to those reported in literature.[4]



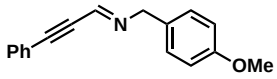
Chemical Formula: $C_{13}H_{12}N_2$
Exact Mass: 196,1000

- *N*-Benzyl-octanal imine 3ga: Following the above general procedure A with octanal (49 mg, 0.373 mmol, 1 equiv) and benzylamine (40 μL, 0. 373 mmol, 1 equiv). The reaction was performed at 40°C during 12h. Imine **3ga** (44 mg, 58%) can be isolated as pure product and as colorless oil. **^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** 7.78 (s, 1 H), 7.30-7.14 (m, 5 H), 4.62 (s, 2 H), 2.34 (t, J = 8 Hz, 2 H), 1.60-1.40 (m, 2 H), 1.25 (m, 8 H), 0.88 (t, 3 H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 166.3, 139.3, 128.4, 127.8, 126.8, 65.1, 35.9, 31.5, 29.4, 29.2, 26.0, 22.5, 14.0. This is a known compound and the spectral data are identical to those reported in literature.[7]



Chemical Formula: $C_{15}H_{23}N$
Exact Mass: 217,1830

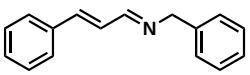
- *N*-4-methoxybenzyl- 3-phenylpropiolaldehyde imine 3ji: Following the above general procedure A with 3-



Chemical Formula: $C_{17}H_{15}NO$
Exact Mass: 249,1154

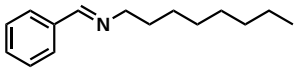
phenylpropiolaldehyde (50 mg, 0.378 mmol, 1 equiv) and benzylamine (50 mg, 0.378 mmol, 1 equiv) in THF/CF_3CH_2OH (4:1, 0,5 mL) at 60°C. Imine **3ji** (79 mg, 79%) can be isolated as pure product and as a yellow oil. **^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** 7.81 (t, *J* = 1.5 Hz, 1H), 7.54–7.51 (m, 2 H), 7.45–7.21 (m, 5 H), 6.90–6.87 (m, 2 H), 4.88 (d, *J* = 1.5 Hz, 2H), 3.80 (s, 3 H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 158.8, 145.3, 143.0, 132.1, 131.2, 130.0, 129.7, 129.4, 129.2, 128.5, 121.4, 113.9, 97.6, 86.6, 65.1, 59.5 ppm. This is a known compound and the spectral data are identical to those reported in literature.[8]

- *N*-benzyl-cinnamaldehyde imine 3ka: Following the above general procedure A with cinnamyl alcohol (52 mg, 0.388 mmol, 1 equiv) and benzylamine (41 μL, 0.388 mmol, 1 equiv). Imine **3ka** (84 mg, 98%) can be isolated as pure product and as a yellow oil. **^{1}H NMR (500 MHz, $CDCl_3$, 20 °C):** 8.12 (s, 1H), 7.46 (d, *J* = 8.5 Hz, 2H), 7.36-7.28 (m, 7H), 7.26-7.24 (m, 1H), 6.96 (d, *J* = 3 Hz, 2H), 4.70 (s, 2H); **^{13}C NMR (125 MHz, $CDCl_3$, 20 °C):** δ (ppm) 163.6, 142.2, 139.3, 135.9, 129.4, 129.1, 129.0, 128.7, 128.3, 128.2, 127.4, 127.2, 100.0, 65.4. This is a known compound and the spectral data are identical to those reported in literature.[4]



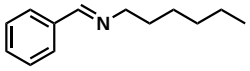
Chemical Formula: $C_{16}H_{15}N$
Exact Mass: 221,1204

- *N*-octyl-benzaldehyde imine 3eb: Following the above general procedure A with benzylalcohol (41 μL, 0.379 mmol, 1 equiv) and octylamine (49 mg, 0. 379 mmol, 1 equiv). Imine **3eb** (76 mg, 92%) can be isolated as pure product and as colorless oil.**^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** 8.25 (s, 1H), 7.73-7.68 (m, 2H), 7.40-7.36 (m, 3H), 3.58 (t, *J* = 7 Hz, 2H), 1.36-1.21 (m, 12H), 0.86 (t, *J* = 7 Hz, 3H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 161.0, 136.6, 130.7, 128.8, 128.3, 62.1, 32.1, 31.2, 29.7, 29.5, 27.6, 22.9, 14.3. This is a known compound and the spectral data are identical to those reported in literature.[9]



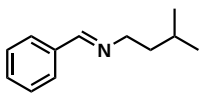
Chemical Formula: $C_{15}H_{23}N$
Exact Mass: 217,1830

- *N*-hexyl-benzaldehyde imine 3ec: Following the above general procedure A with benzylalcohol (41 μL, 0.379 mmol, 1 equiv) and hexylamine (38.4 mg, 0. 379 mmol, 1 equiv). Imine **3ec** (76 mg, 92%) can be isolated as pure product and as colorless oil. **^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** 8.25 (s, 1H), 7.73-7.68 (m, 2H), 7.40-7.36 (m, 3H), 3.59 (t, *J* = 7 Hz, 2H), 1.68 (quint, *J* = 7 Hz, 2H), 1.36-1.27 (m, 6H), 0.88 (t, *J* = 5 Hz, 2H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 161.1, 136.8, 130.8, 128.9, 128.4, 62.2, 32.3, 31.3, 29.8, 29.7, 27.8, 23.1, 14.5. This is a known compound and the spectral data are identical to those reported in literature.[10]



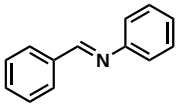
Chemical Formula: $C_{13}H_{19}N$
Exact Mass: 189,1517

- *N*-isopentyl-benzaldehyde imine 3ed: Following the above general procedure A with benzylalcohol (41 μL, 0.379 mmol, 1 equiv) and hexylamine (33.0 mg, 0.379 mmol, 1 equiv). Imine **3ed** (56 mg, 84%) can be isolated as pure product and as colorless oil.**^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** 8.24 (s, 1H), 7.69 (bs, 2H), 7.36 (bs, 3H), 3.59 (t, *J* = 7 HZ, 2H), 1.65 (non, *J* = 6.5 Hz, 1H), 1.60-1.53 (m, 2H), 0.91 (s, 6H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 160.9, 136.5, 130.7, 130.6, 128.7, 128.2, 60.0, 40.1, 26.1, 22.8. This is a known compound and the spectral data are identical to those reported in literature.[10]



Chemical Formula: $C_{12}H_{17}N$
Exact Mass: 175,1361

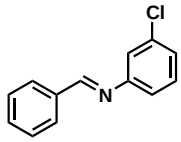
- *N*-phenyl-benzaldehyde imine 3ee: Following the above general procedure A with benzylalcohol (41 μL, 0.379 mmol, 1 equiv) and aniline (35.3 mg, 0.379 mmol, 1 equiv) in THF/CF_3CH_2OH (4:1, 0,5 mL) at 60°C. Imine **3ee** (48 mg, 70%) can be isolated as pure product and



Chemical Formula: $C_{13}H_{11}N$
Exact Mass: 181,0891

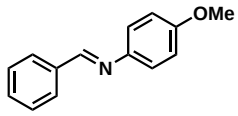
as yellow oil. **^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** 8.40 (s, 1H), 7.85 (dd, *J* = 5 and 2 Hz, 2H), 7.43-7.40 (m, 3H), 7.33 (td, *J* = 8 and 2 Hz, 2H), 7.18 (td, *J* = 2 and 9 Hz, 1H), 7.17-7.14 (m, 2H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 160.6, 152.4, 136.4, 131.6, 129.4, 129.0, 129.0, 126.1, 121.1. This is a known compound and the spectral data are identical to those reported in literature.[11]

- *N*-3-chlorophenyl-benzaldehyde imine 3ef: Following the above general procedure C with benzyl alcohol (41 μL, 0.379 mmol, 1 equiv) and 3-chloroaniline (48.4 mg, 0.379 mmol, 1 equiv) in THF/CF_3CH_2OH (4:1, 0,5 mL) at 60°C. Imine **3ef** (65 mg, 79%) can be isolated as pure product and as yellow oil. **^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** 8.42 (s, 1H), 7. 90-7.88 (m, 2H), 7.50-7.46 (m, 3H), 7.32-7.29 (m, 1H), 7.21-7.19 (m, 2H), 7.09-7.07 (m, 1H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 161.3, 153.3, 135.8, 134.7, 131.7, 130.0, 128.9, 128.8, 125.8, 120.9, 119.4. This is a known compound and the spectral data are identical to those reported in literature.[12]



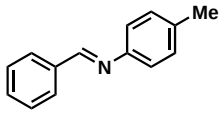
Chemical Formula: $C_{13}H_{10}ClN$
Exact Mass: 215,0502

- *N*-4-methoxyphenyl-benzaldehyde imine 3eg: Following the above general procedure A with benzylalcohol (41 μL, 0.379 mmol, 1 equiv) and 4-methoxyaniline (46.7 mg, 0.379 mmol, 1 equiv) in THF/CF_3CH_2OH (4:1, 0,5 mL) at 60°C. Imine **3eg** (76 mg, 95%) can be isolated as pure product and as red amorphous solid. **^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** 8.47 (s, 1H), 7.87 (dd, *J* = 4 and 5 Hz, 2H), 7.45 (q, *J* = 3.5 Hz, 3H), 7.22 (d, *J* = 9 Hz, 2H), 6.92 (d, *J* = 9 Hz, 2H), 3.81 (s, 3H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 158.9, 158.7, 145.4, 136.9, 131.5, 129.2, 129.0, 122.6, 114.8, 55.9. This is a known compound and the spectral data are identical to those reported in literature.[11]



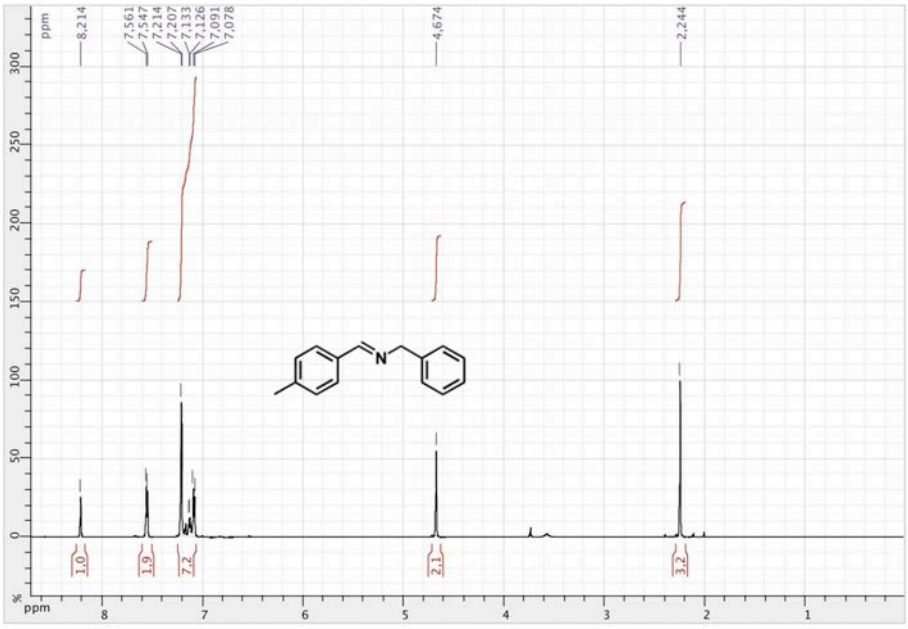
Chemical Formula: $C_{14}H_{13}NO$
Exact Mass: 211,0997

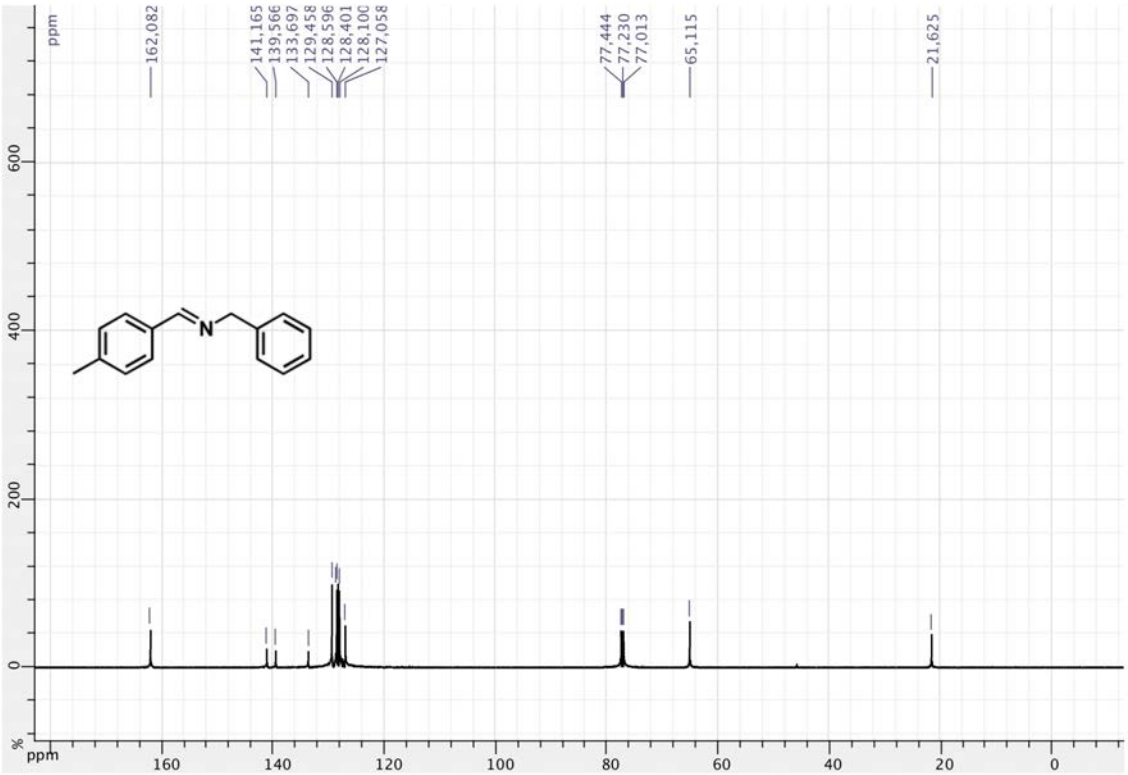
- *N*-4-methylphenyl-benzaldehyde imine 3eh: Following the above general procedure C with benzylalcohol (41 μL, 0.379 mmol, 1 equiv) and 4-methylaniline (40.6 mg, 0.379 mmol, 1 equiv) in THF/CF_3CH_2OH (4:1, 0,5 mL) at 60°C. Imine **3eh** (65 mg, 88%) can be isolated as pure product and as red amorphous solid. **^{1}H NMR (600 MHz, $CDCl_3$, 20 °C):** 8.45 (s, 1H), 7.88 (q, *J* = 4 Hz, 2H), 7.45 (d, *J* = 5 Hz, 3H), 7.19-7.16 (m, 2H), 7.14-7.11 (m, 2H), 2.35 (s, 3H); **^{13}C NMR (150 MHz, $CDCl_3$, 20 °C):** δ (ppm) 160.0, 149.9, 136.8, 136.3, 131.6, 130.2, 129.1, 127.4, 121.2, 21.4. This is a known compound and the spectral data are identical to those reported in literature.[11]

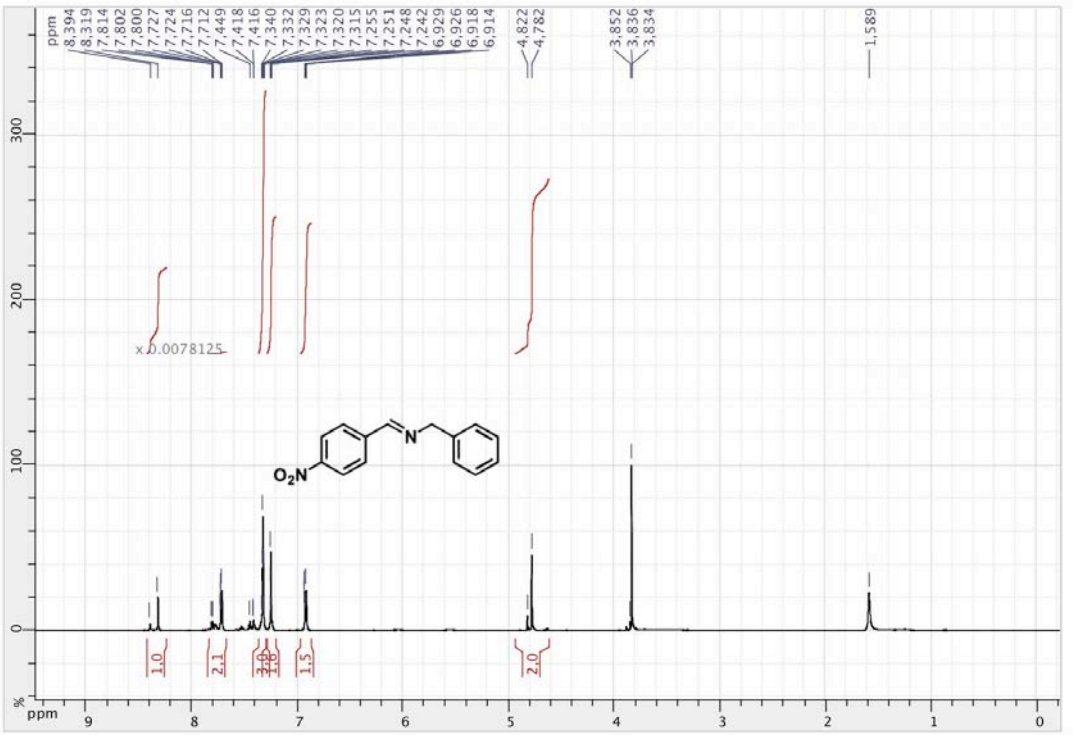


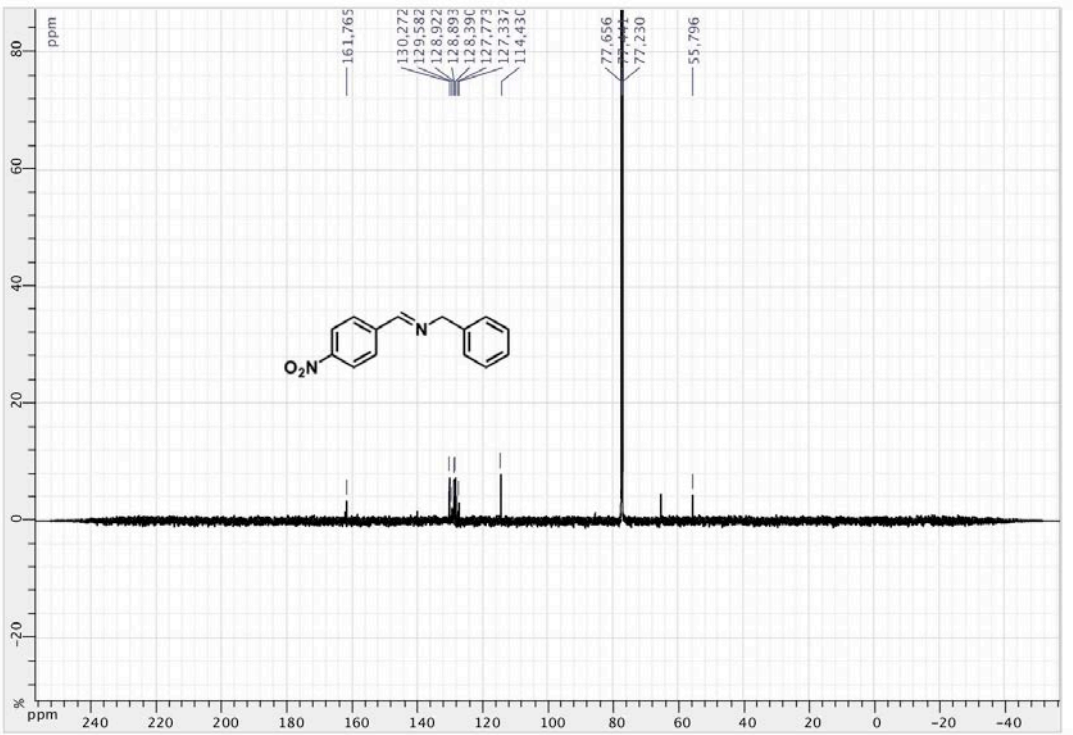
Chemical Formula: $C_{14}H_{13}N$
Exact Mass: 195,1048

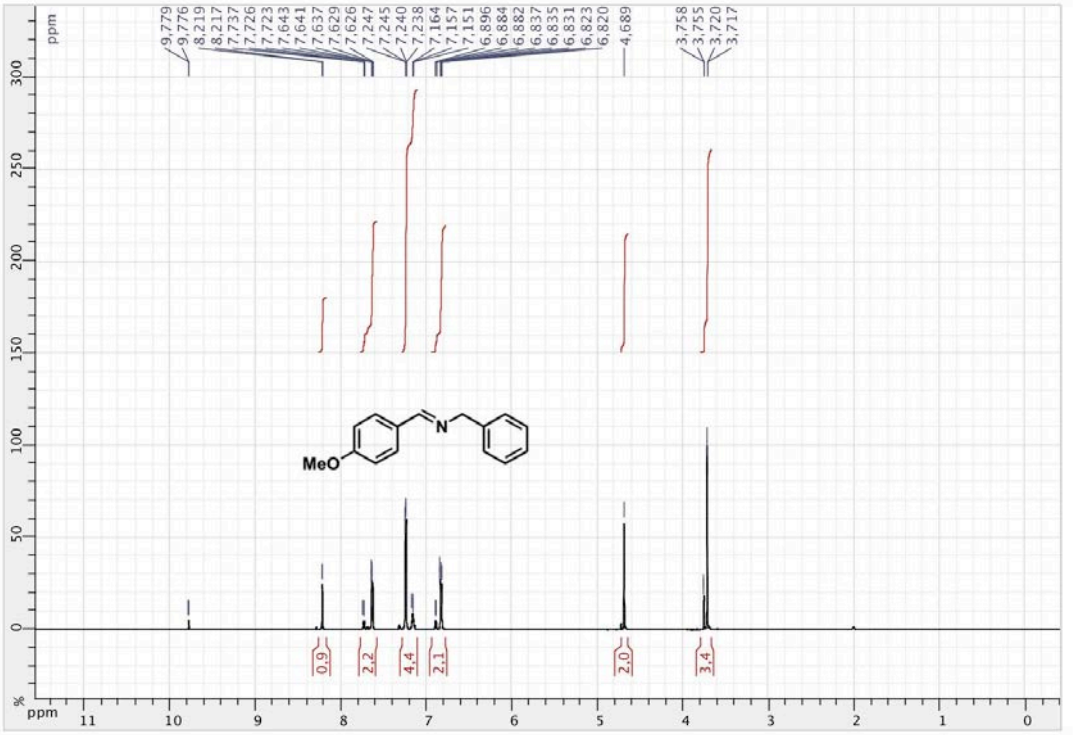
5. NMR Charts

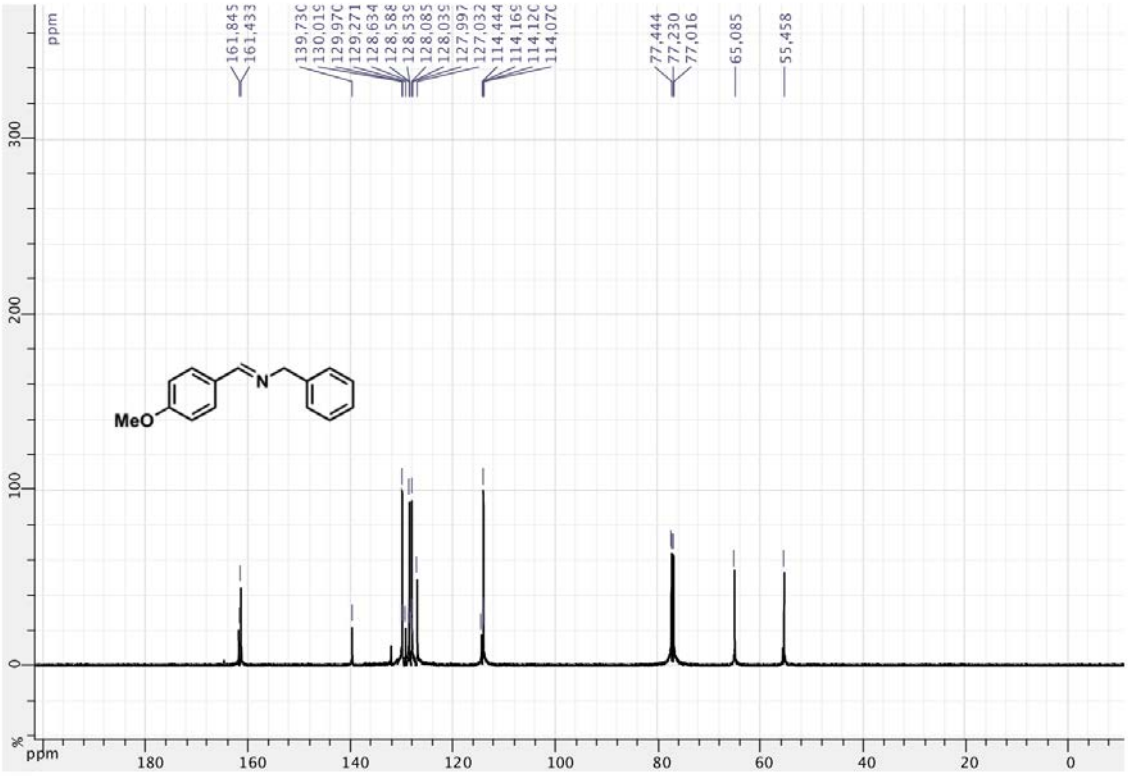


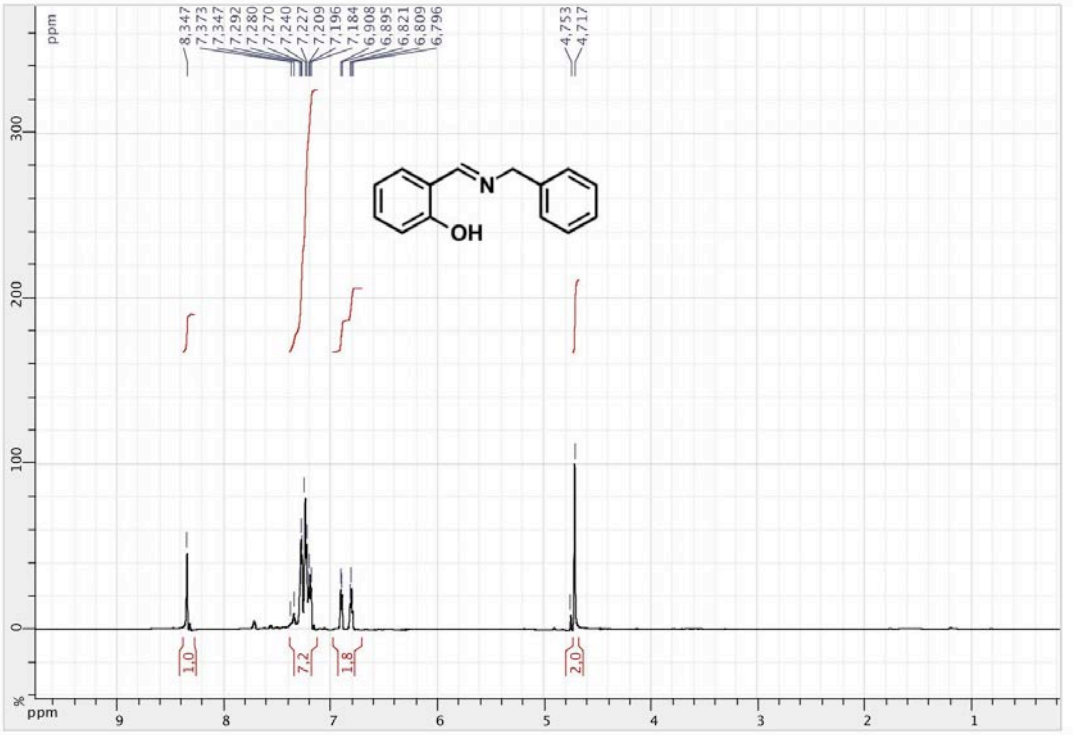


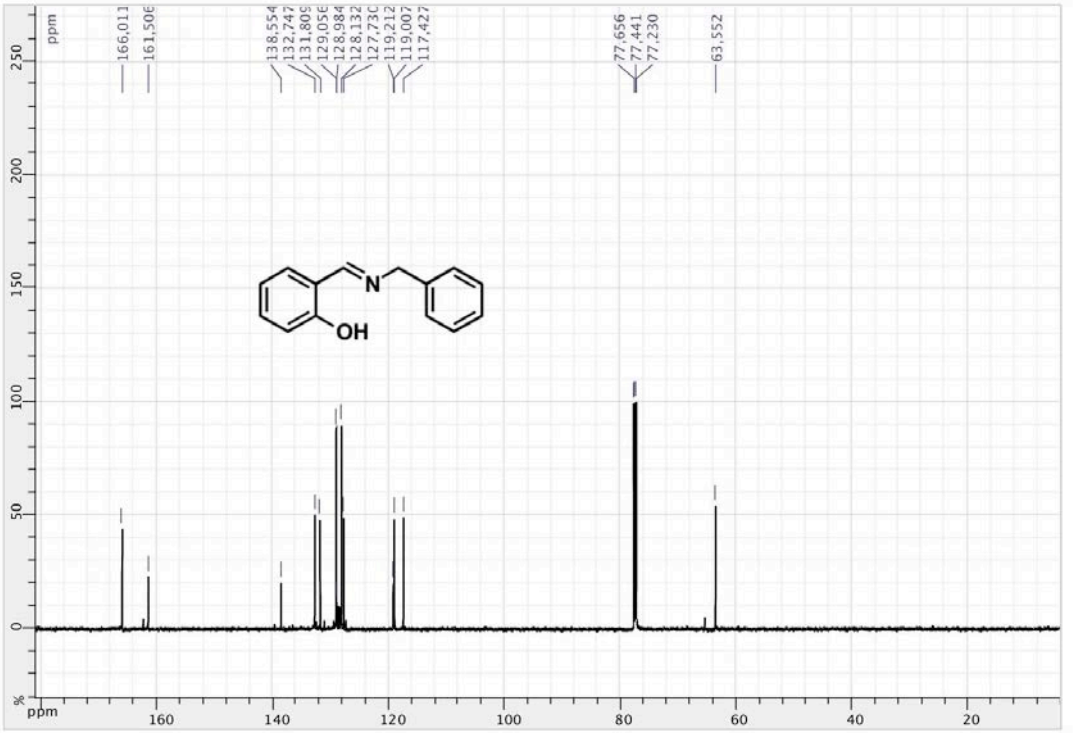


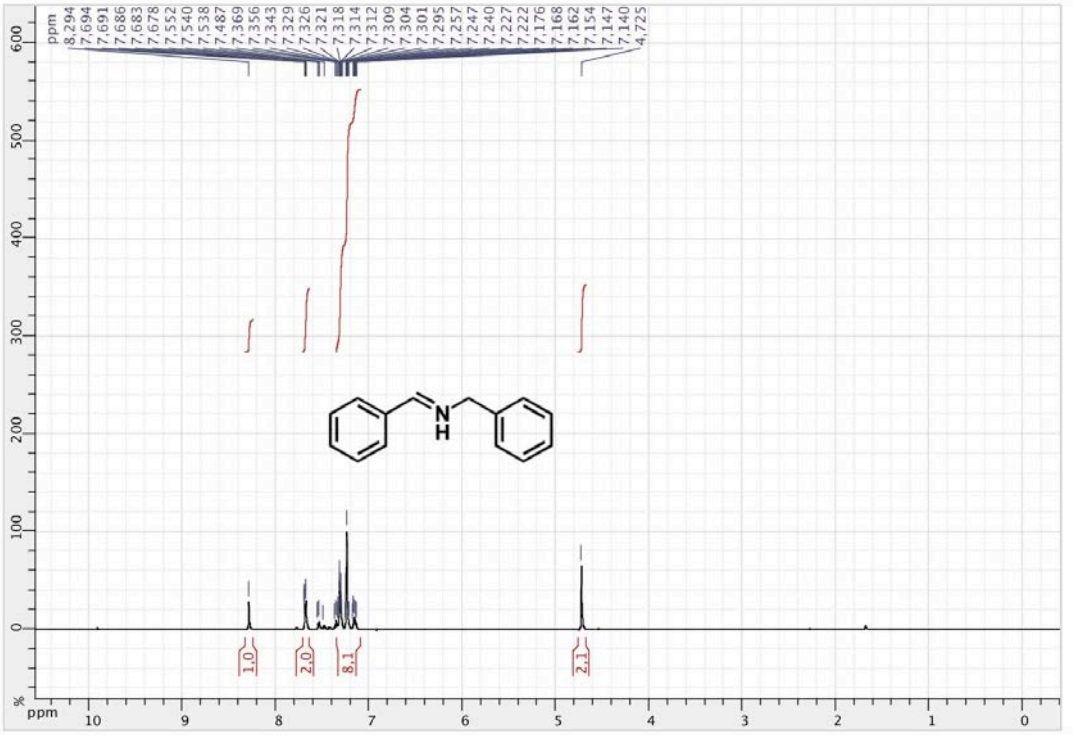


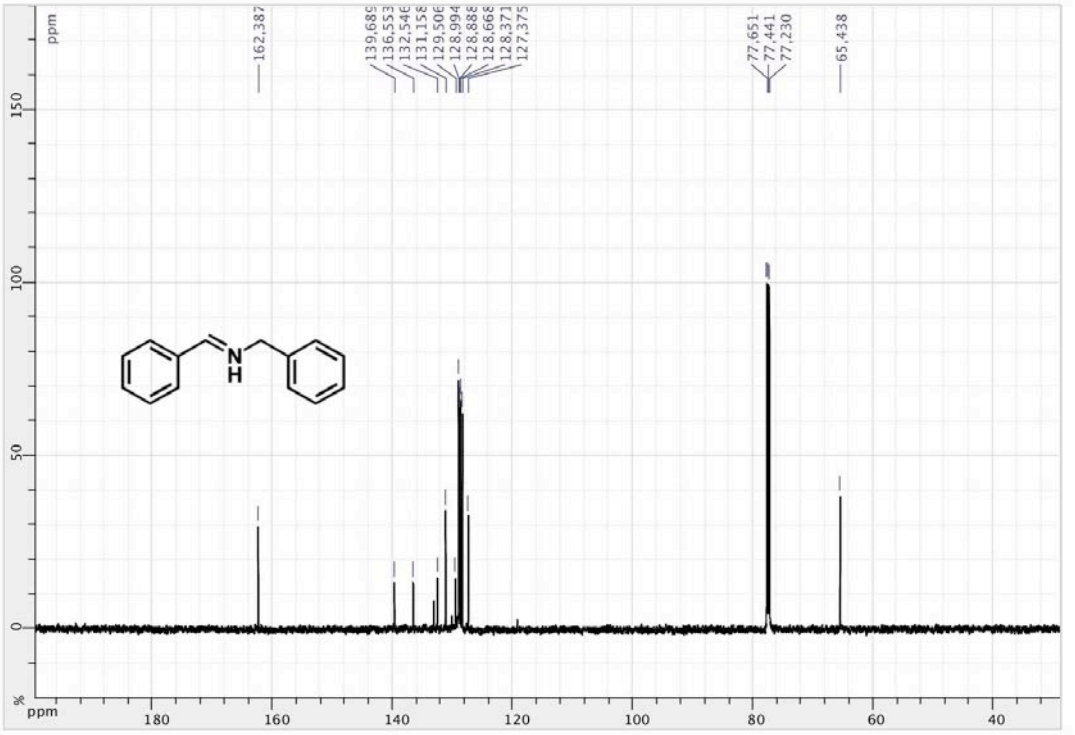


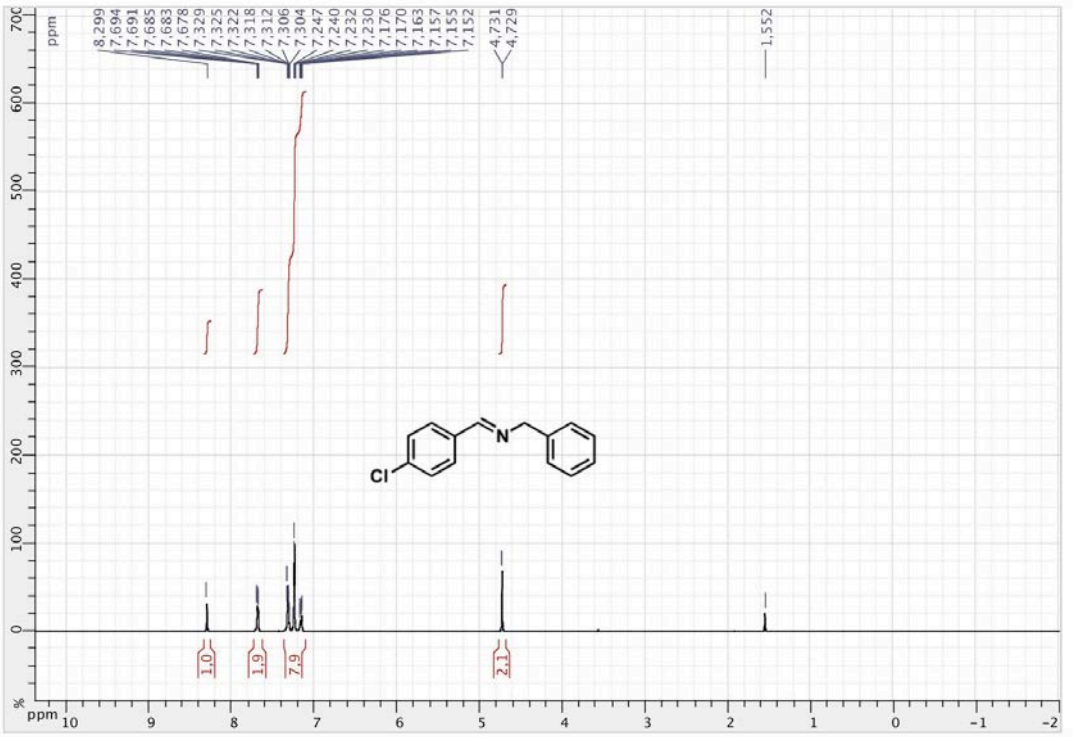


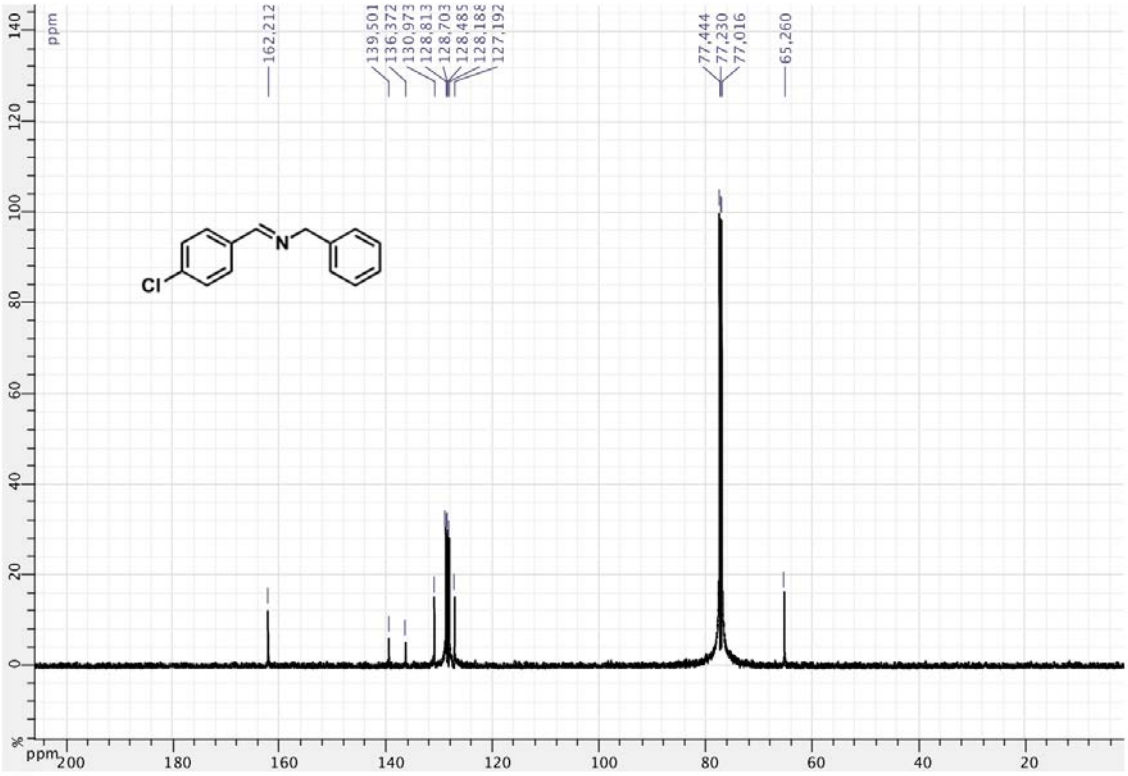


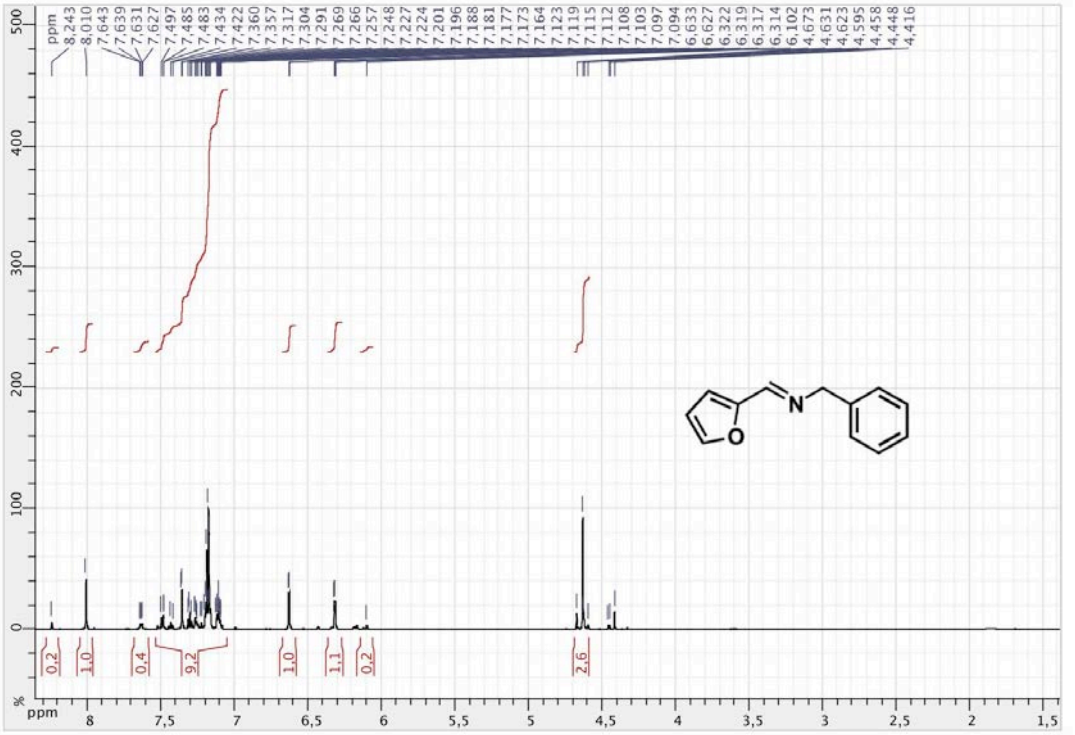


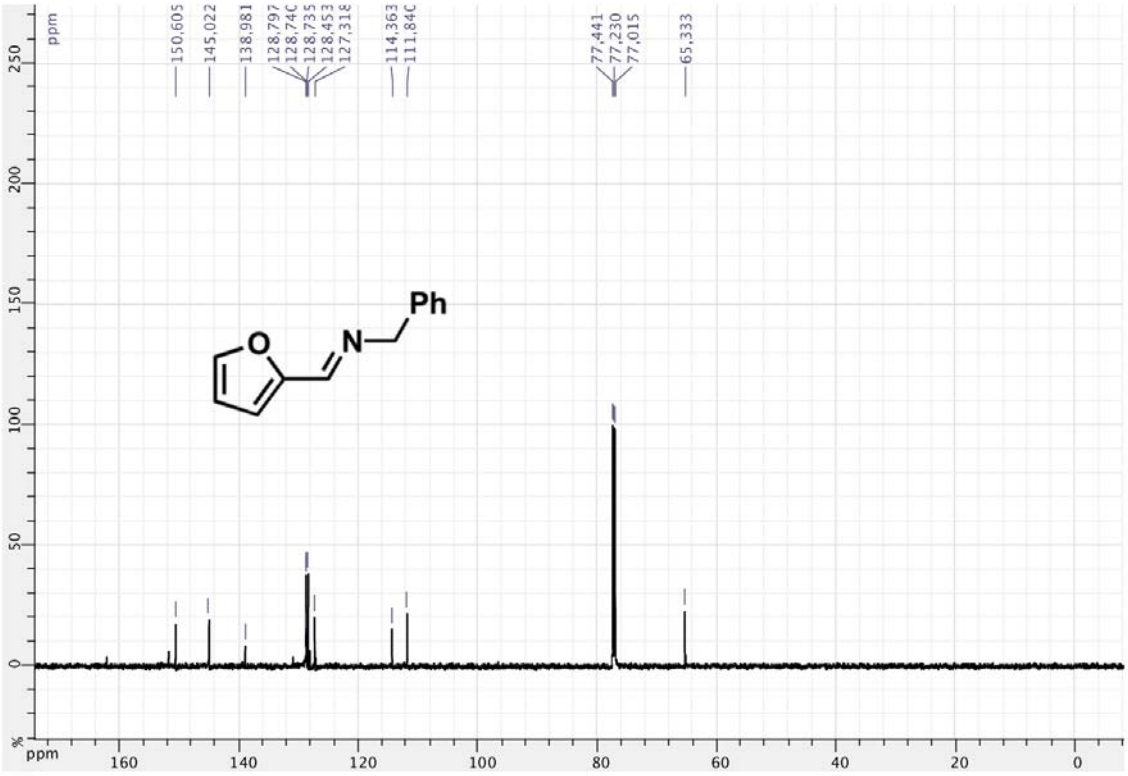


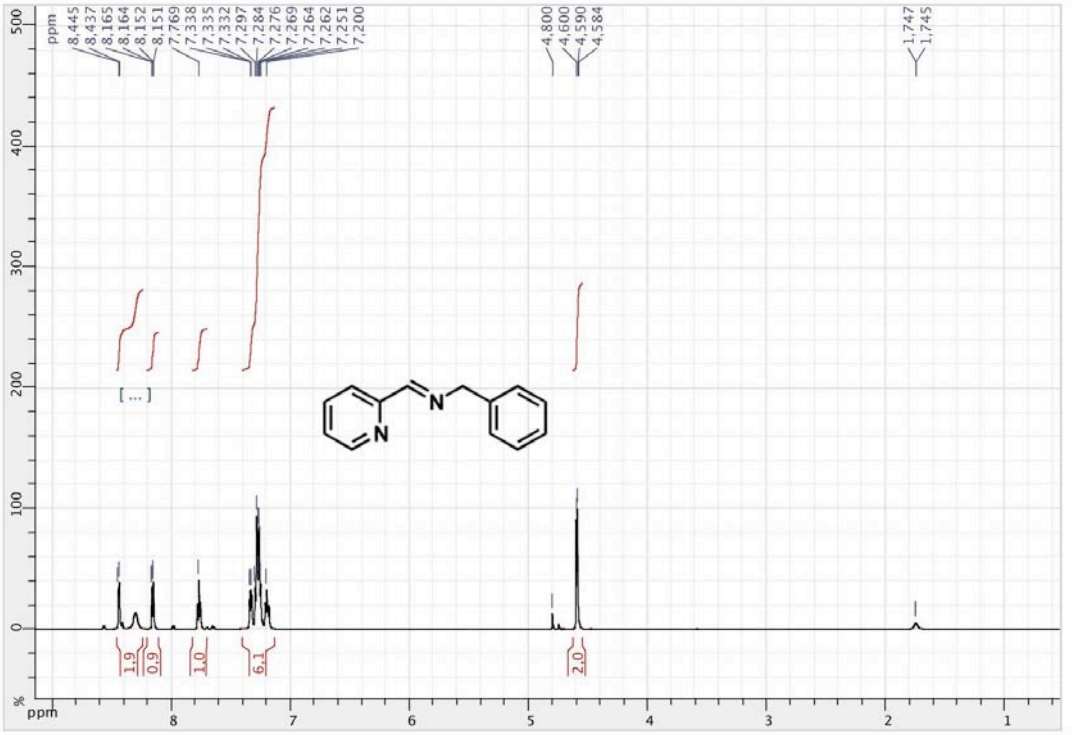


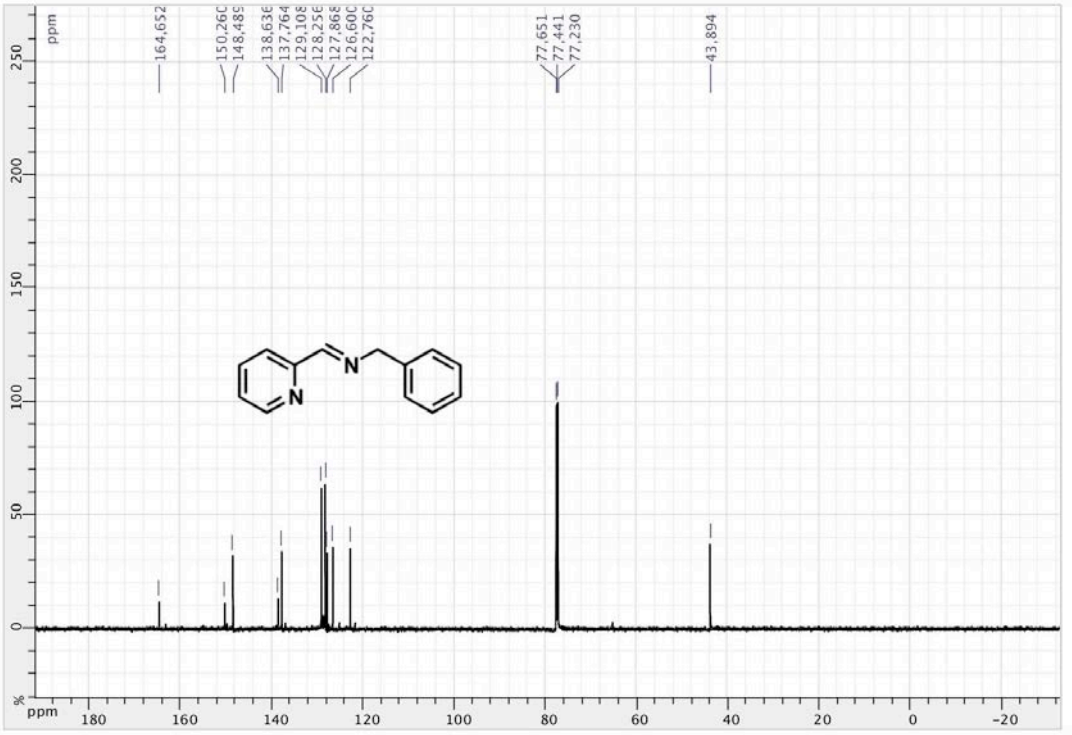


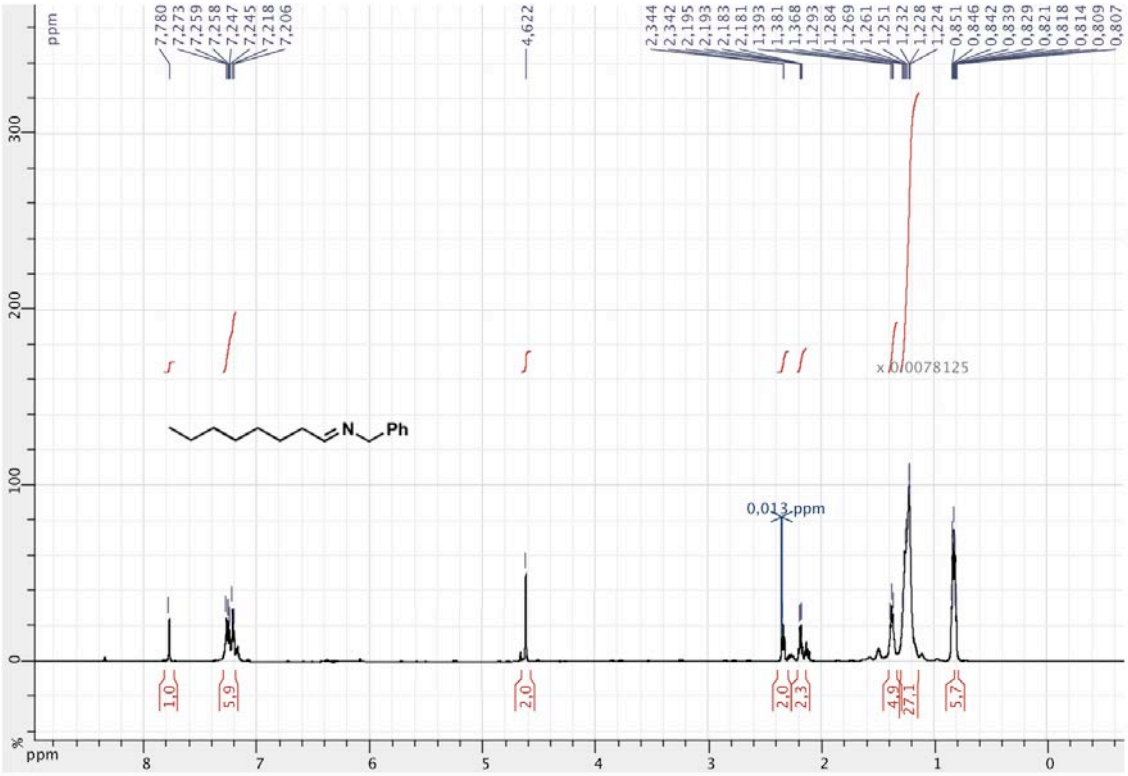








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0,814
0,809
0,807
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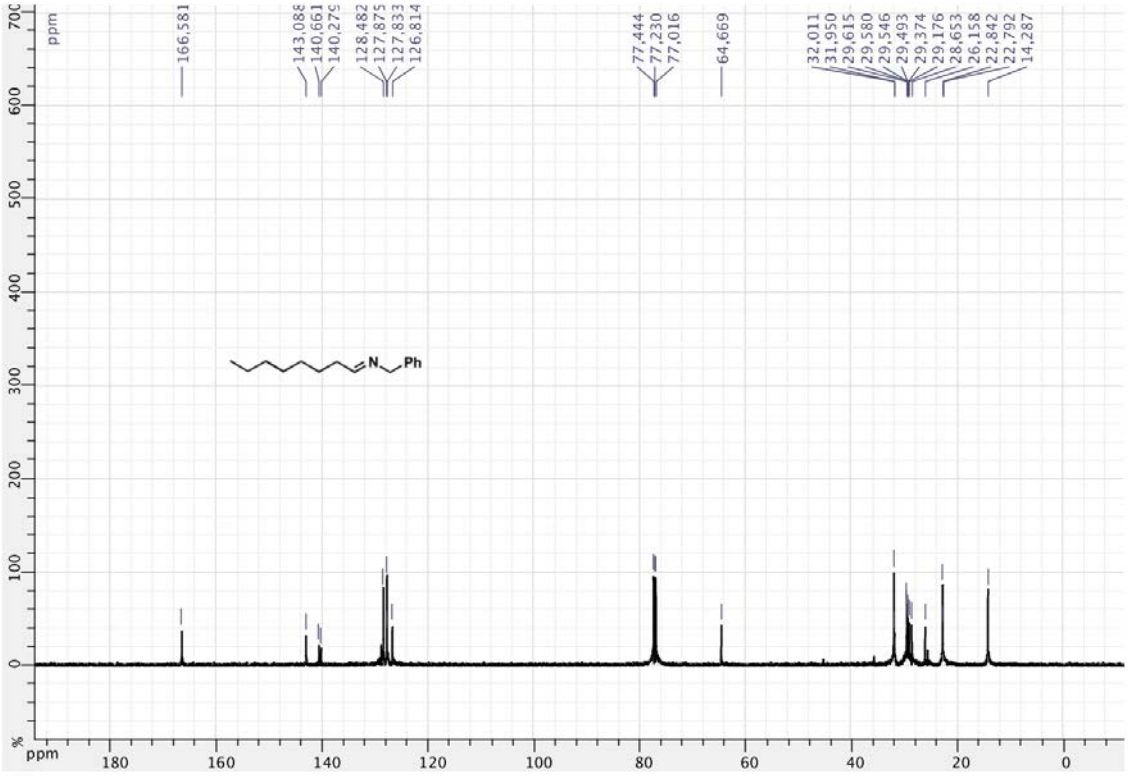
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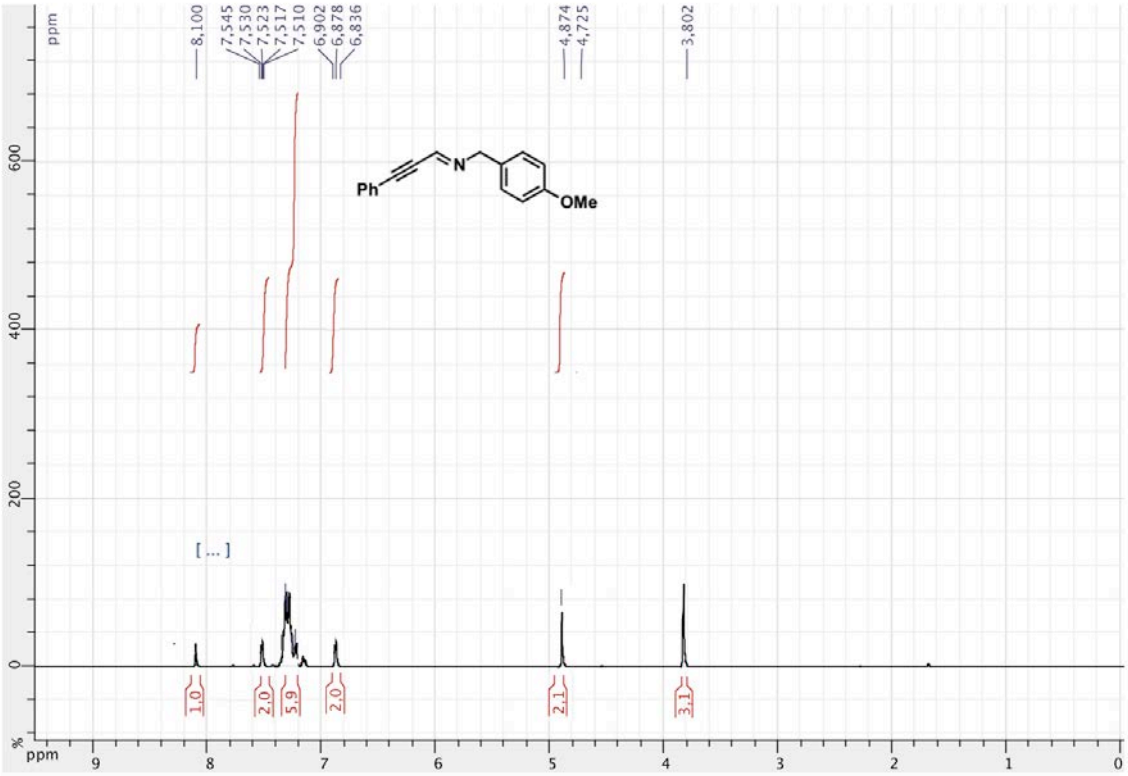

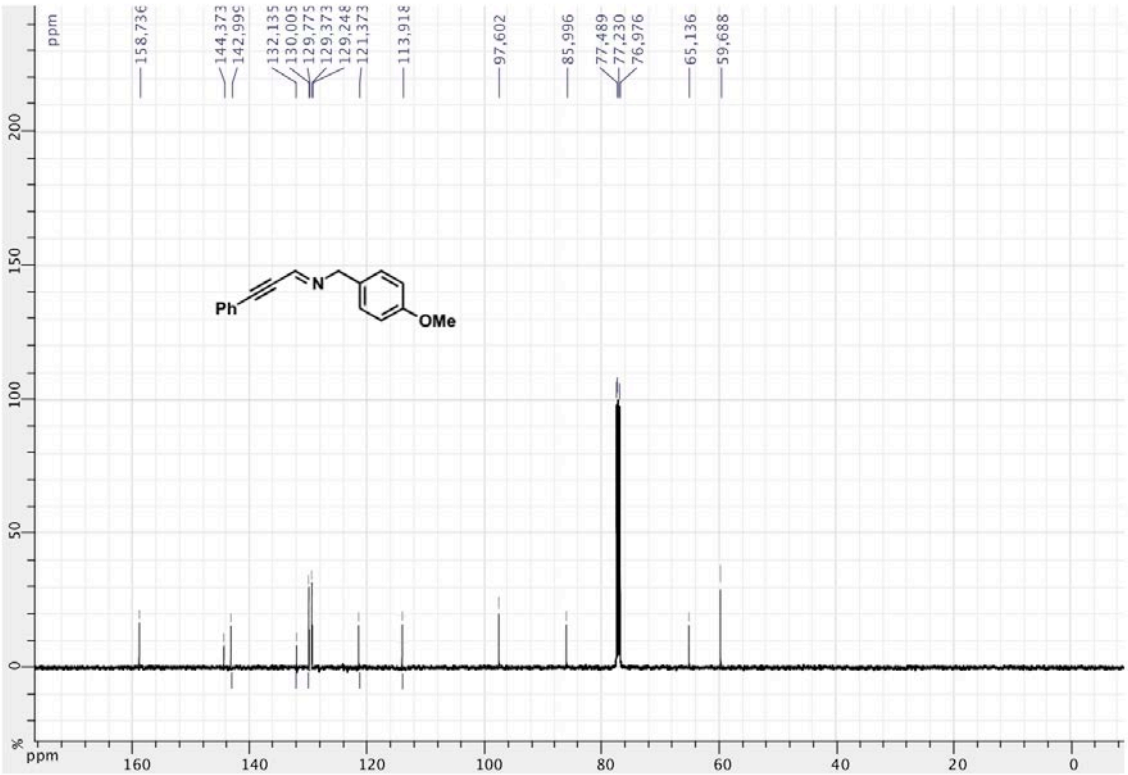

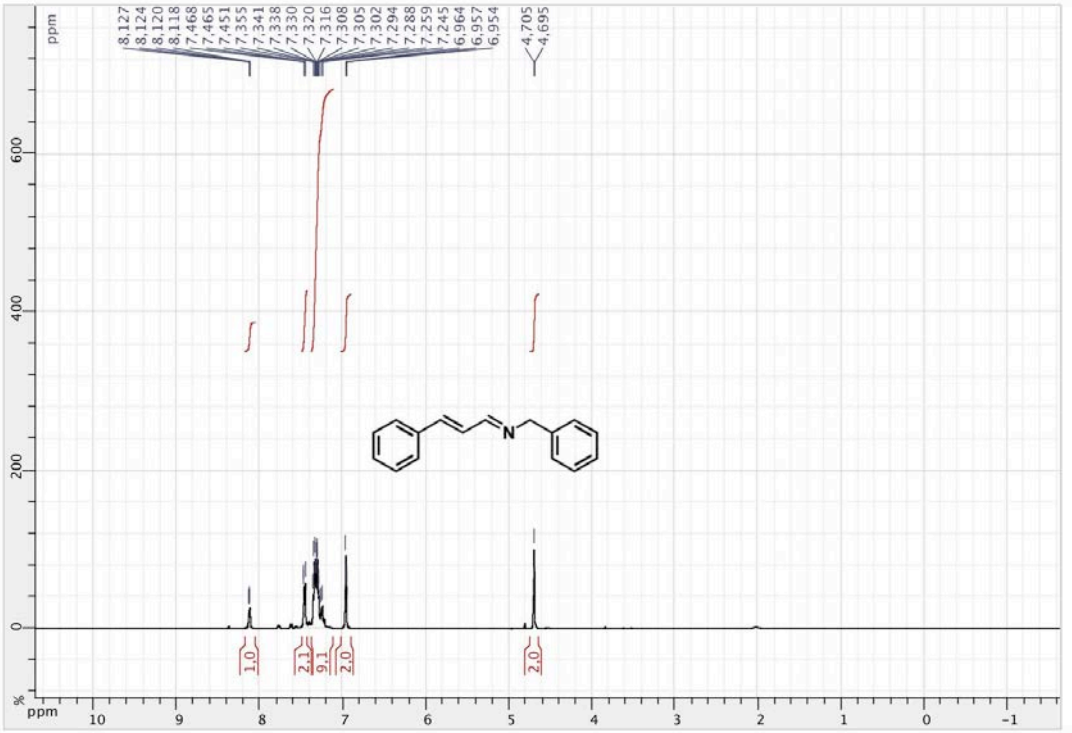

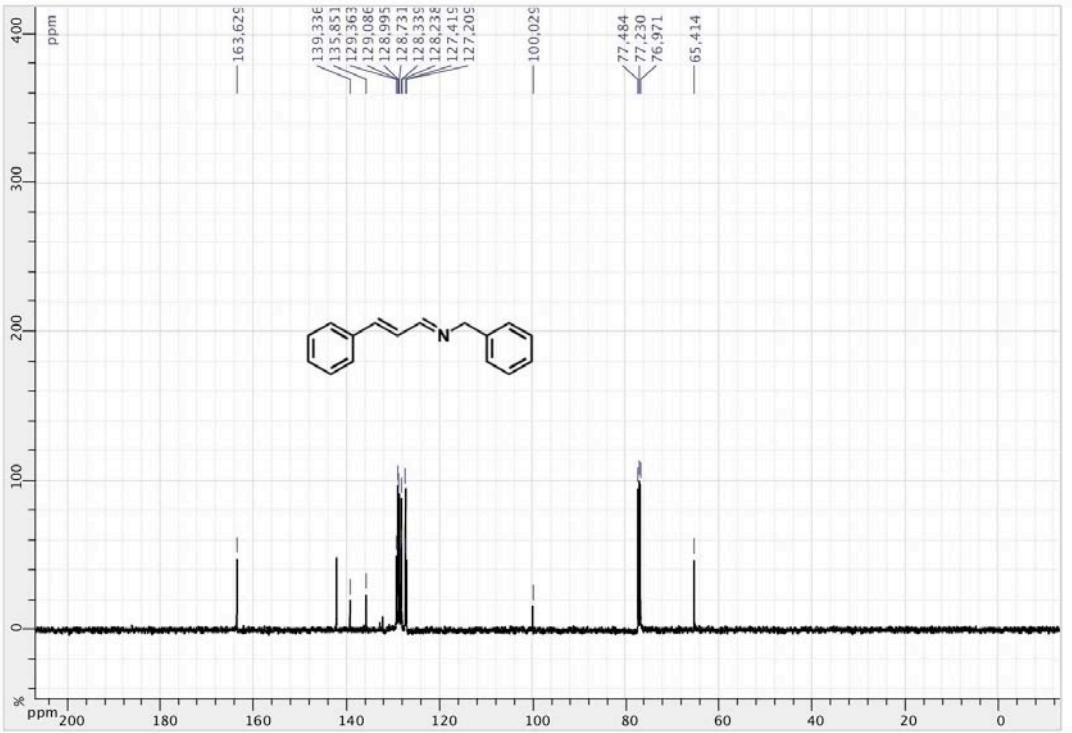

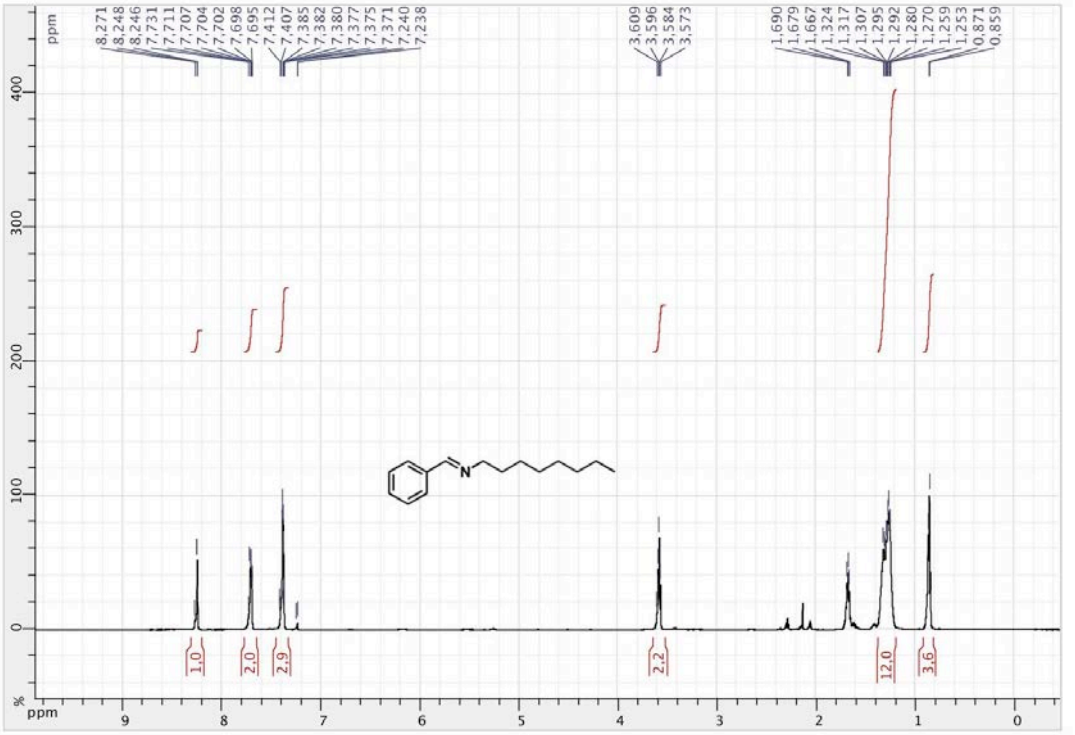

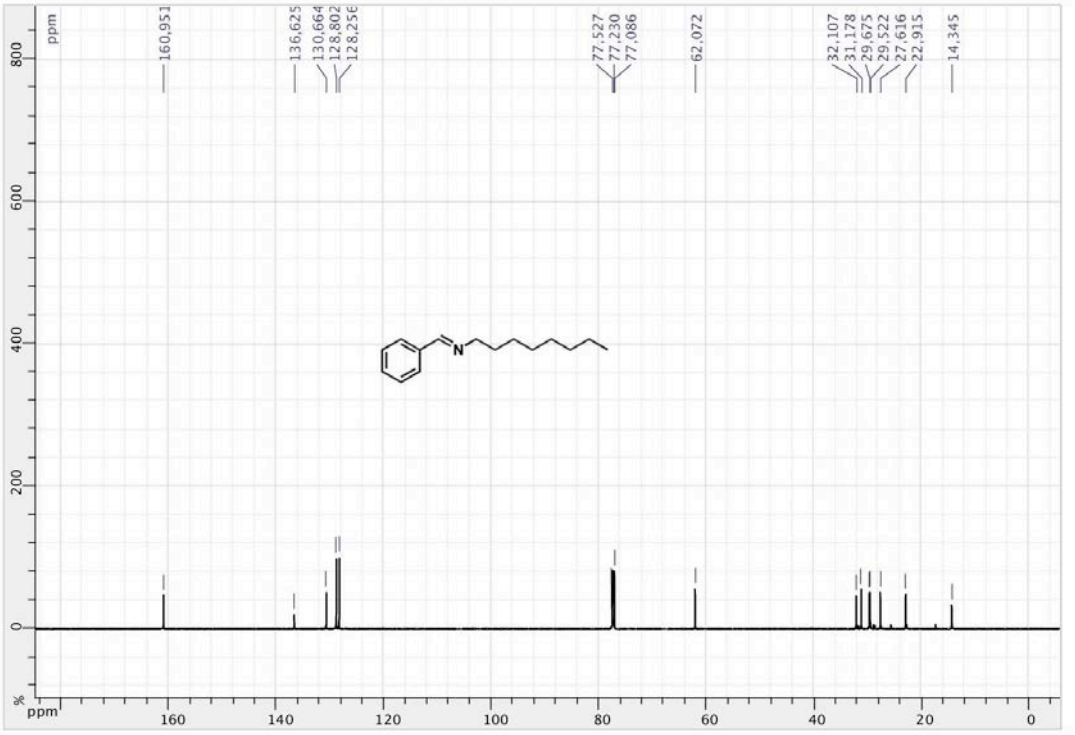

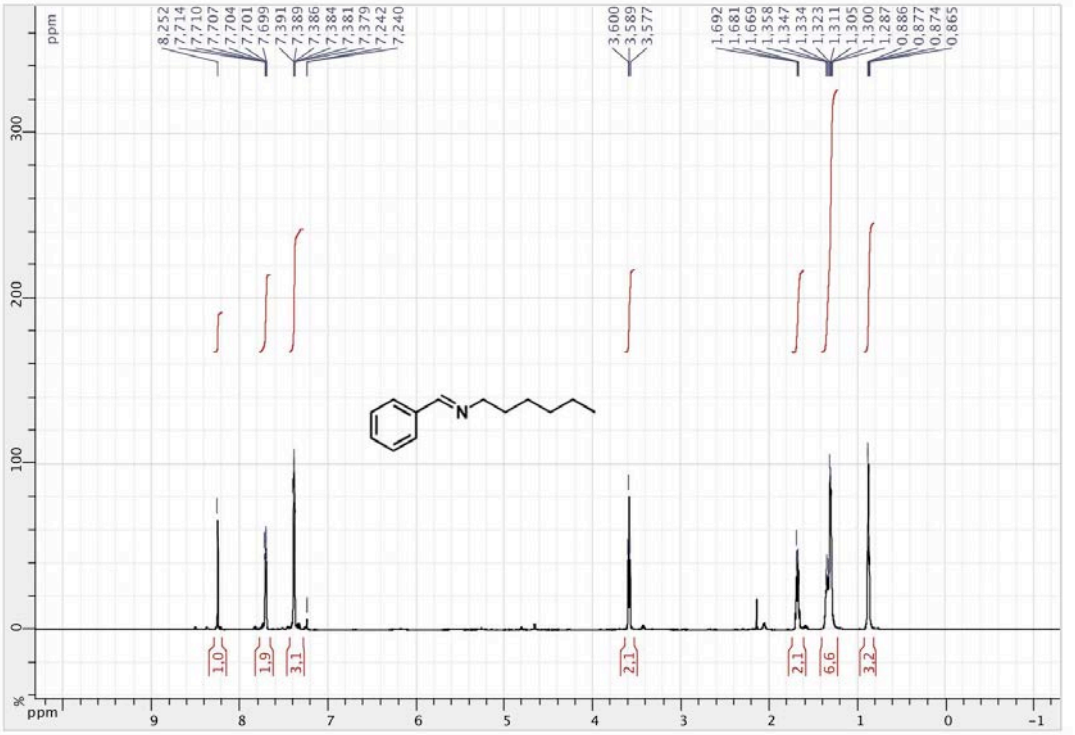

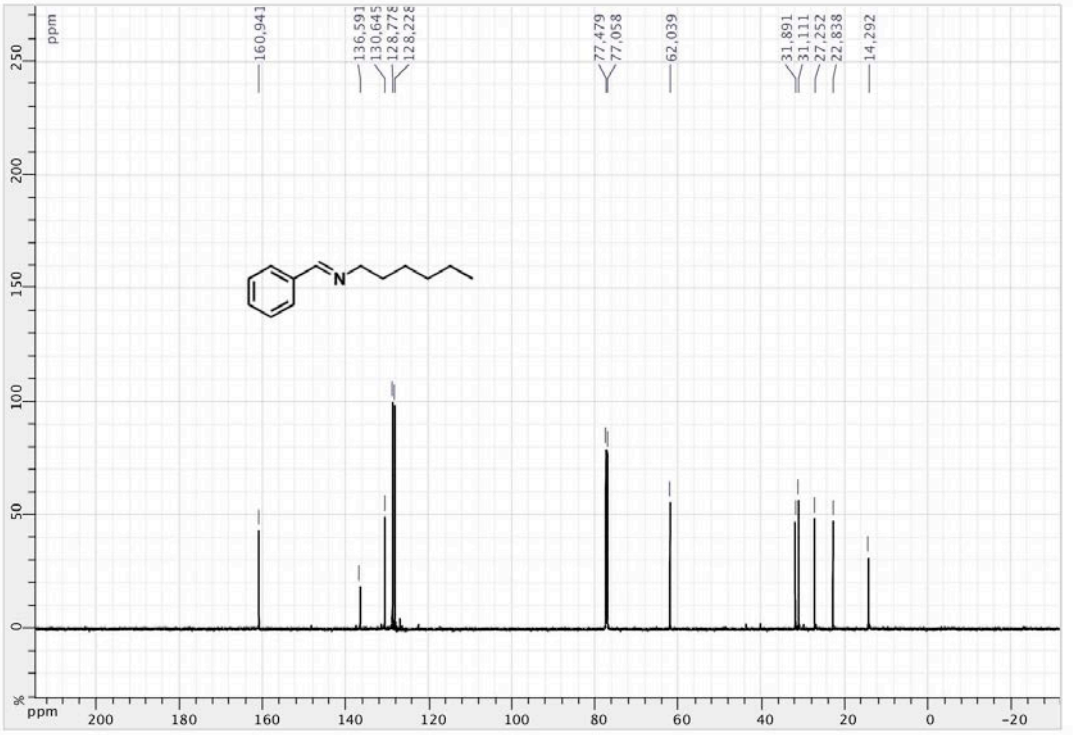

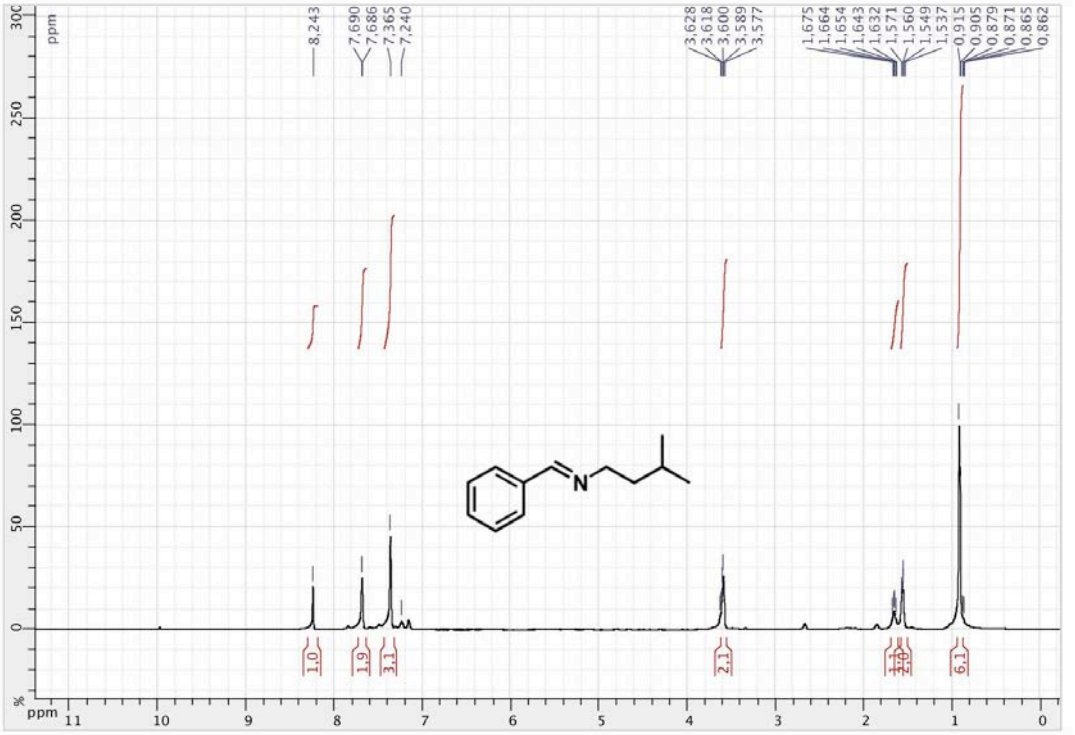

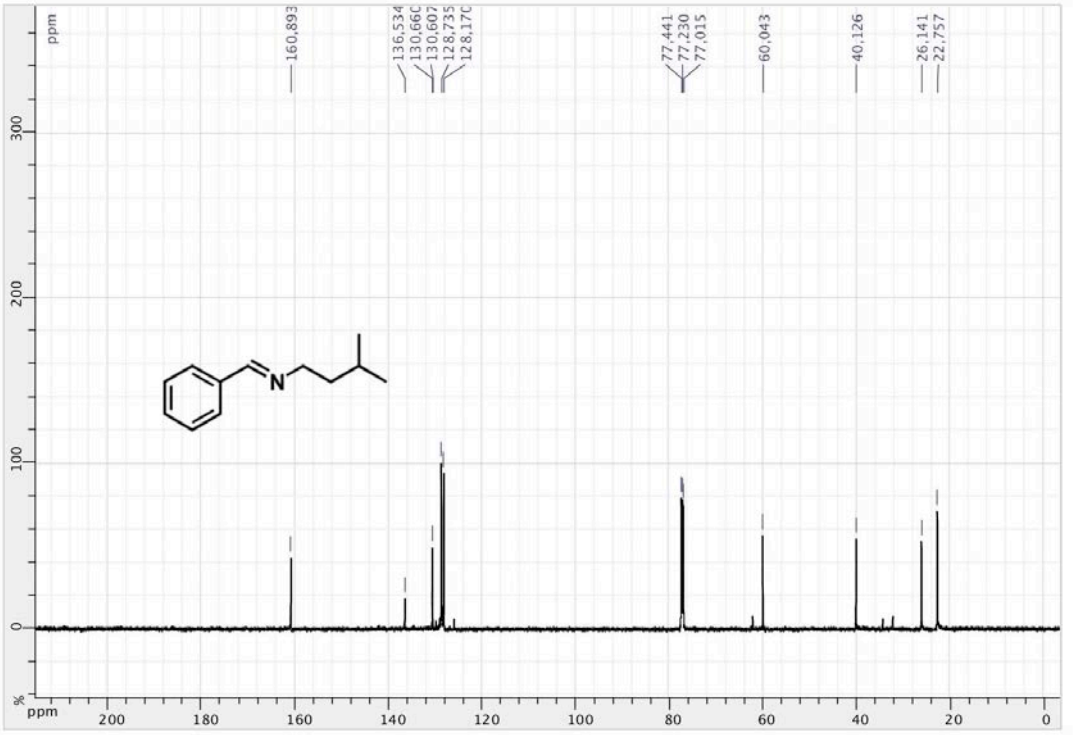

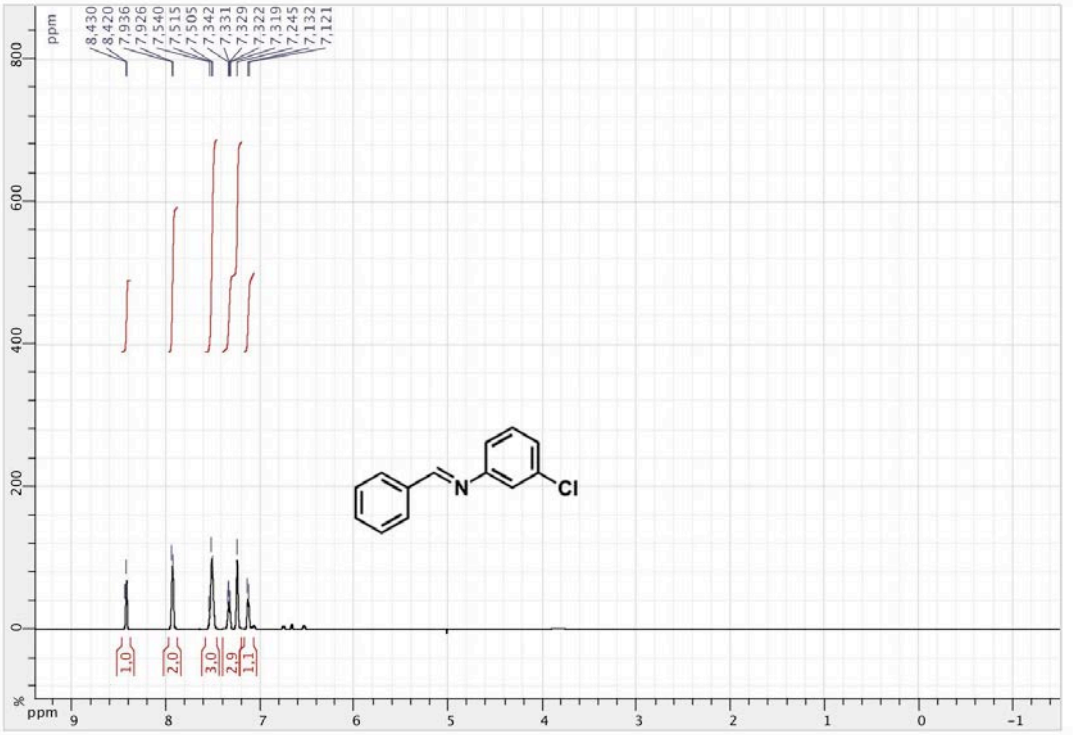

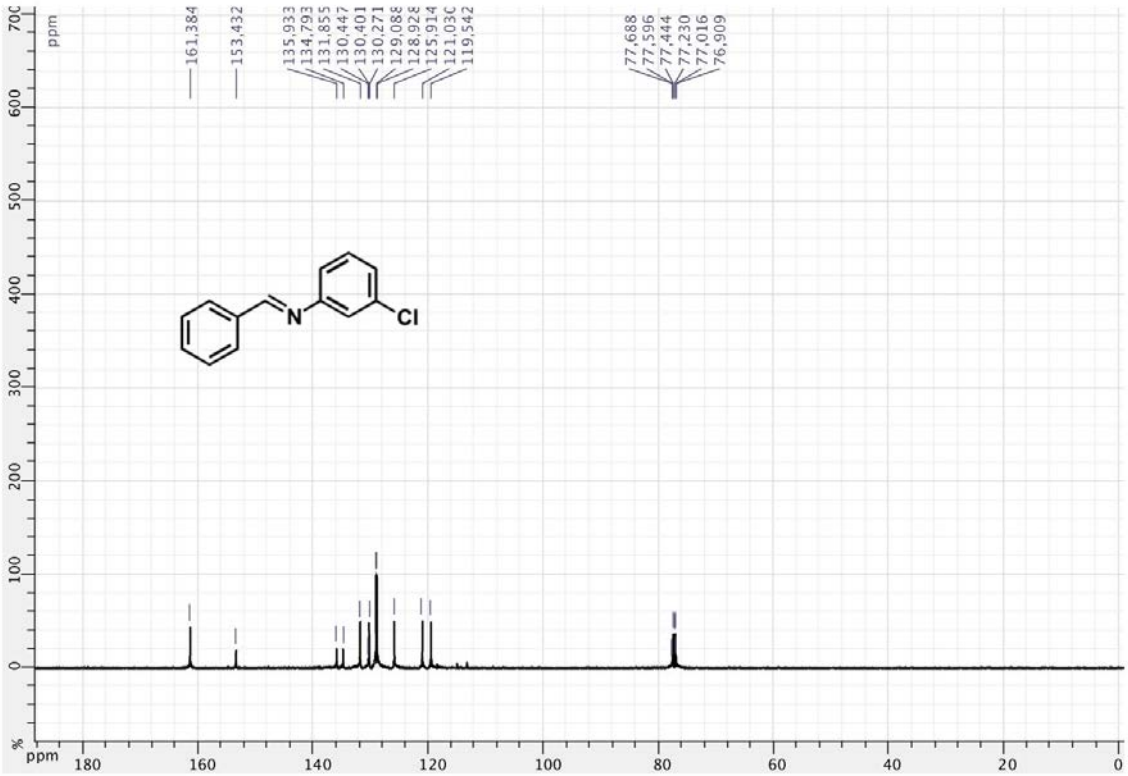

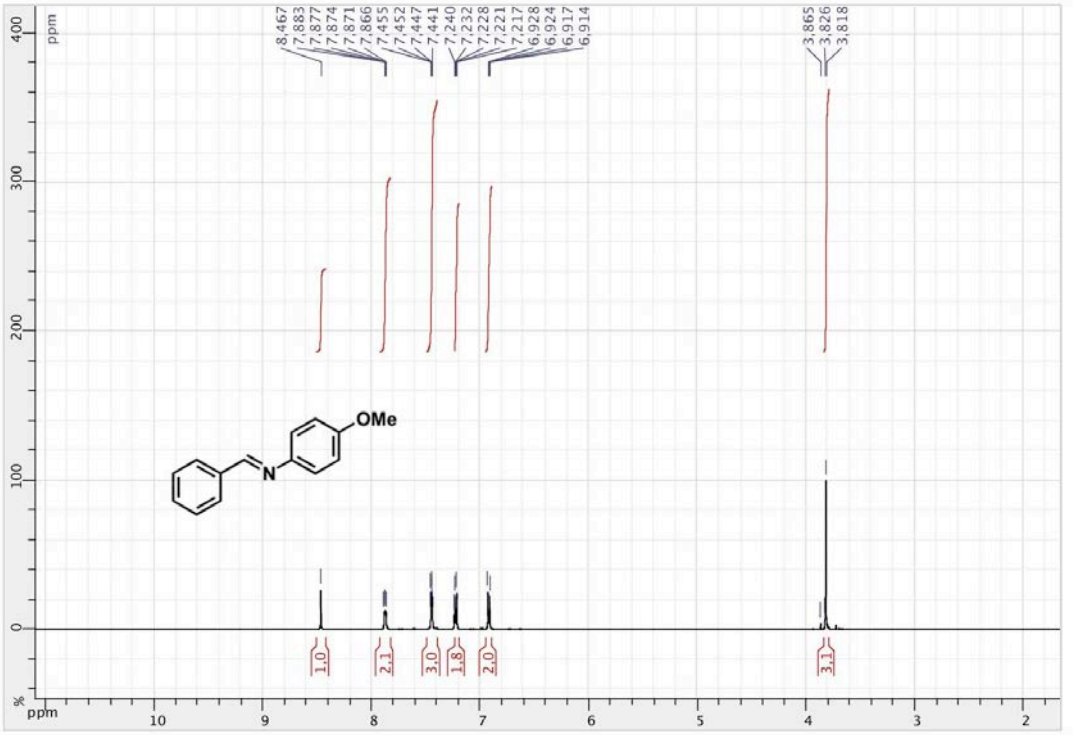

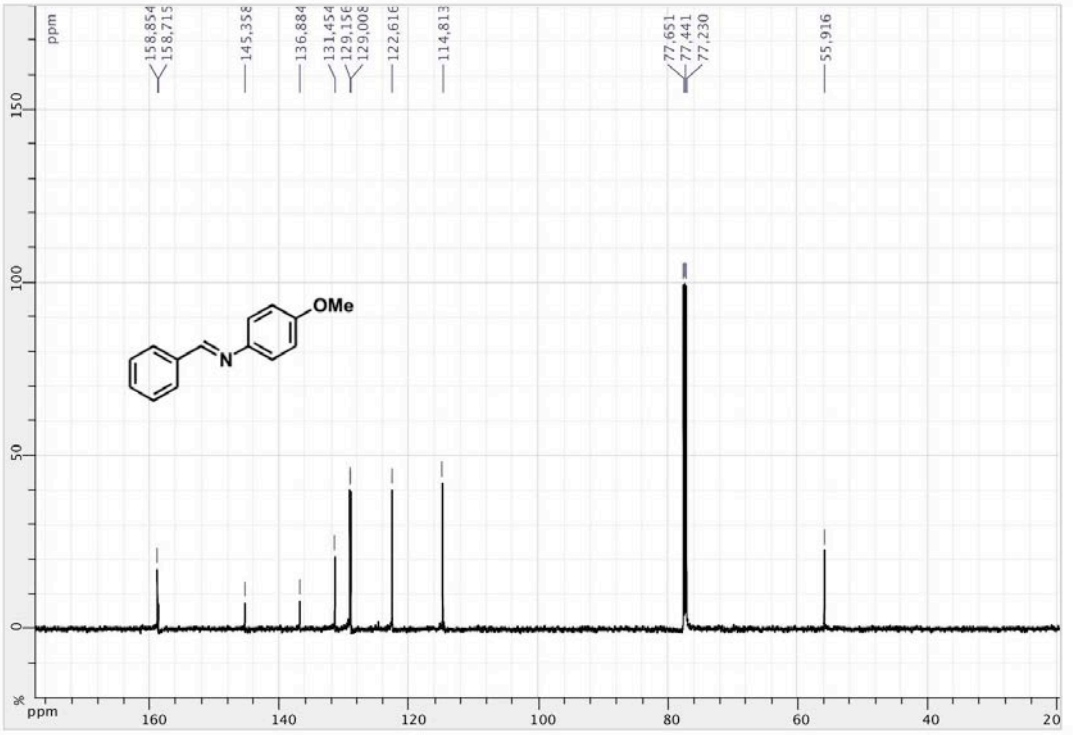

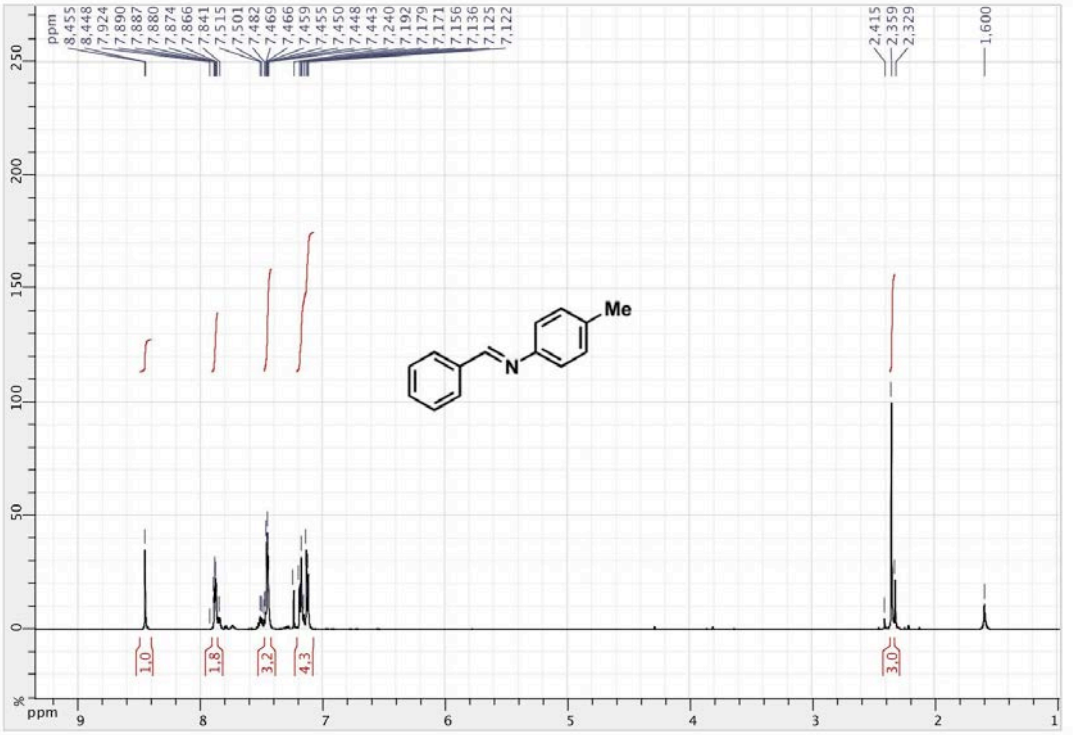

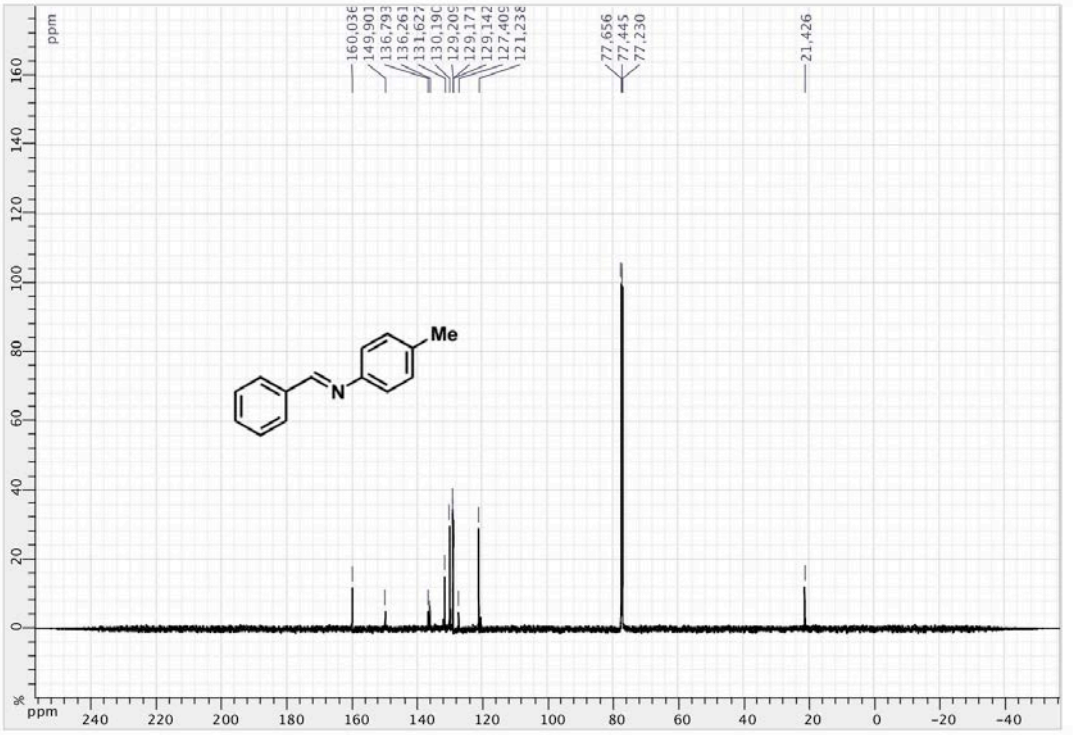

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