

**Wacker-Type Oxidation and Dehydrogenation of Terminal Olefins Using Molecular Oxygen
as the Sole Oxidant without Adding Ligand**

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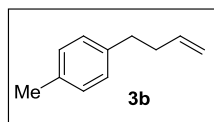
1. General Information

All commercial reagents and solvents were used as received without further purification. Reactions were followed with TLC (0.254 mm silica gel 60-F plates). Visualization was accomplished with UV light. Flash chromatographies were carried out on silica gel 200-300 mesh. All NMR spectra were obtained at ambient temperature using Varian INOVA-400MHz spectrometer. ^1H NMR and ^{13}C NMR spectra were recorded using CDCl_3 as solvent. Spectra were referenced internally to the residual proton resonance in CDCl_3 (δ 7.26 ppm), or with tetramethylsilane (TMS, δ 0.00 ppm) as the internal standard. Chemical shifts (δ) were reported as part per million (ppm) in δ scale downfield from TMS. Multiplicities are reported as follows: s = singlet, d = doublet, t = triplet, m = multiplet, br. s = broad singlet. The known products were characterized by comparing to the corresponding ^1H -NMR and ^{13}C -NMR from the literatures. Infrared (IR) spectra were recorded using KBr Solid on a Bruker EQUINOX55 instrument. High resolution mass spectrometry data were obtained on a micro TOF-Q II (hybrid quadrupolar/time-of-flight) API US system by electrospray ionization (ESI) in the positive ion mode using a Bruker instrument.

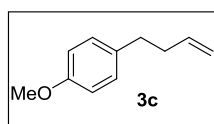
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2. Representative Procedure for the Synthesis of Butenylated Arenes: 3b-3f¹

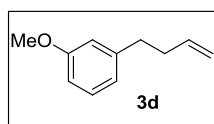
To a flame-dried 100 mL flask was added benzyl bromide (1.0 equiv) and 20 mL anhydrous THF. The reaction flask was under an atmosphere of nitrogen and allylmagnesium bromide was added dropwise (1.0 M in Et₂O, 2.0 equiv). The reaction was stirred for 4h at RT and then quenched with saturated aqueous NH₄Cl. The aqueous layer was extracted with CH₂Cl₂ and the combined organics were dried over MgSO₄, filtered through celite, and concentrated in vacuo. The crude product was purified by flash chromatography (eluent: hexane), affording the desired product as a clear, colorless oil.



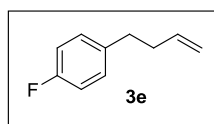
4-(4-tolyl)-1-butene (3b)² Prepared from 4-methylbenzyl bromide according to the representative procedure as light yellow oil; Yield 96% (eluent: hexane); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.08 (s, 4H), 5.80-5.89 (m, 1H), 5.06-4.96 (m, 2H), 2.65-2.68 (m, 2H), 2.31-2.36 (m, 5H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 138.9, 138.3, 135.3, 129.1, 128.4, 114.9, 35.8, 35.1, 21.1.



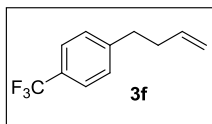
4-(4-methoxyphenyl)-1-butene (3c)¹ Prepared from 4-methoxybenzyl chloride according to the representative procedure as light yellow oil; Yield 95% (eluent: hexane); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.08 (d, *J* = 8.0 Hz, 2H), 6.81 (d, *J* = 8.0 Hz, 2H), 5.80-5.87 (m, 1H), 4.95-5.04 (m, 2H), 3.75 (s, 3H), 2.63 (t, *J* = 8.0 Hz, 2H), 2.33 (t, *J* = 8.0 Hz, 2H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 157.9, 138.3, 134.1, 129.4, 115.0, 113.8, 55.3, 36.0, 34.6.



3-(*m*-methoxyphenyl)-1-butene (3d)² Prepared from 3-methoxybenzyl chloride according to the representative procedure as light yellow oil; Yield 93% (eluent: hexane); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.18 (t, *J* = 8.0 Hz, 1H), 6.71-6.76 (m, 3H), 5.80-5.84 (m, 1H), 4.96-5.06 (m, 2H), 3.77 (s, 3H), 2.67 (t, *J* = 8.0 Hz, 2H), 2.37 (t, *J* = 8.0 Hz, 2H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 159.8, 143.7, 138.2, 129.4, 121.0, 115.1, 114.4, 111.2, 55.2, 35.6.

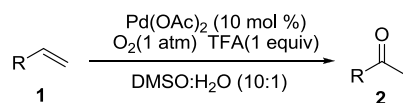


4-(4-Fluorophenyl)-1-butene (3e)³ Prepared from 4-Fluorophenyl bromide according to the representative procedure as light yellow oil; Yield 90% (eluent: hexane); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.12 (d, *J* = 8.0 Hz, 2H), 6.97 (d, *J* = 8.0 Hz, 2H), 5.80-5.84 (m, 1H), 4.97-5.05 (m, 2H), 2.67 (d, *J* = 8.0 Hz, 2H), 2.35 (d, *J* = 4.0 Hz, 2H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 161.5 (d, *J* = 240.0 Hz), 138.0, 137.6, 130.0, 129.94 (d, *J* = 7.0 Hz), 115.33 (d, *J* = 5.0 Hz), 115.1, 35.9, 34.8.



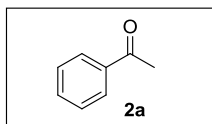
4-(4-Trifluoromethylphenyl)-1-butene (3f)¹ Prepared from 4-Trifluoromethylphenyl bromide according to the representative procedure as light yellow oil; Yield 93% (eluent: hexane); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.53 (d, *J* = 8.0 Hz, 2H), 7.28 (d, *J* = 8.0 Hz, 2H), 5.78-5.88 (m, 1H), 4.98-5.06 (m, 2H), 2.76 (t, *J* = 8.0 Hz, 2H), 2.38 (q, *J* = 8.0 Hz, 2H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 146.1, 137.5, 128.9, 128.3 (q, *J* = 32.0 Hz), 125.4 (q, *J* = 4.0 Hz), 124.5 (q, *J* = 271.0 Hz), 115.6, 35.29, 35.26.

3. General Procedure for Palladium-Catalyzed Wacker Oxidant Using Oxygen as the Oxidant

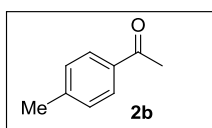


A seal tube containing terminal olefins **1** (1 mmol) and Pd(OAc)₂ (10 mol %), was evacuated and filled with dioxygen gas using an oxygen containing balloon. Then, DMSO (2.5 mL), H₂O (0.25 mL) and trifluoroacetic acid (TFA) (1 mmol) were sequentially added to the system via syringe under an oxygen atmosphere. The reaction mixture was stirred at 70 °C until completion of the reaction (TLC). Then the reaction was cooled to RT and partitioned between water and ethyl acetate. The layers were separated and the organic layer was washed with aqueous saturated brine solution, dried over MgSO₄, filtered and concentrated under reduced pressure. Purification by flash chromatography afforded the methyl ketones **2**.

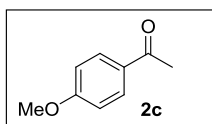
4. Characterization Data of Methyl Ketones



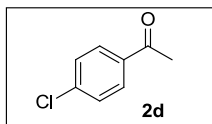
1-Phenylethanone (2a)⁴ as light yellow oil; Yield 86% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.94 (d, *J* = 8.0 Hz, 2H), 7.55-7.52 (m, 1H), 7.43 (t, *J* = 8 Hz, 2H), 2.57 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 26.6, 128.3, 128.5, 133.1, 137.0, 198.1.



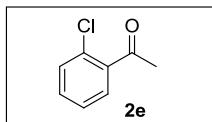
1-p-Tolyleanone (2b)⁵ as light yellow oil; Yield 94% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.85 (d, *J* = 8.0 Hz, 2H), 7.25 (d, *J* = 8.0 Hz, 2H), 2.57 (s, 3H), 2.40 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 198.0, 144.0, 134.8, 129.4, 128.6, 26.7, 21.8.



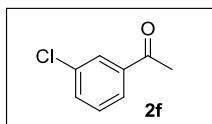
1-(4-Methoxyphenyl)ethanone (2c)⁵ as light yellow oil; Yield 92% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.92 (d, *J* = 8.0 Hz, 2H), 6.92 (d, *J* = 8.0 Hz, 2H), 3.85 (s, 3H), 2.54 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 196.9, 163.6, 130.7, 113.8, 55.5, 26.4.



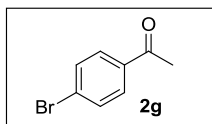
1-(4-Chlorophenyl)ethanone (2d)⁵ as light yellow oil; Yield 88% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.84 (d, *J* = 8.0 Hz, 2H), 7.38 (d, *J* = 8.0 Hz, 2H), 2.54 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 197.0, 139.7, 135.6, 130.0, 129.1, 26.8.



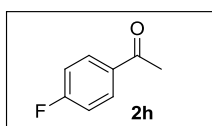
1-(2-Chlorophenyl)ethanone (2e)⁶ as light yellow oil; Yield 95% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.54 (d, *J* = 8.0 Hz, 1H), 7.36-7.39 (m, 2H), 7.31 (t, *J* = 8.0 Hz, 1H), 2.63 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 200.3, 139.0, 132.0, 131.2, 130.6, 129.3, 126.9, 30.6.



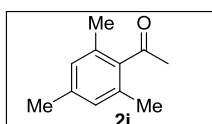
1-(3-Chlorophenyl)ethanone (2f)⁶ as light yellow oil; Yield 85% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.86 (d, *J* = 8.0 Hz, 1H), 7.77 (t, *J* = 8.0 Hz, 1H), 7.47 (t, *J* = 8.0 Hz, 1H), 7.36 (t, *J* = 8.0 Hz, 1H), 2.55 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 196.9, 138.8, 133.2, 130.1, 128.6, 126.6, 26.9.



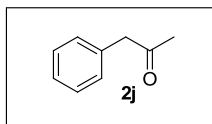
1-(4-Bromophenyl)ethanone (2g)⁵ as light yellow oil; Yield 86% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.82 (d, *J* = 8.0 Hz, 2H), 7.61 (d, *J* = 8.0 Hz, 2H), 2.59 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 197.2, 136.0, 132.1, 130.0, 128.5, 26.8.



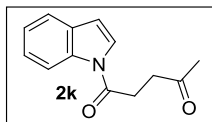
1-(4-Fluorophenyl)ethanone (2h)⁶ as light yellow oil; Yield 91% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.97-8.00 (m, 2H), 7.10-7.15 (m, 2H), 2.59 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 196.6, 165.9 (d, *J* = 250.0 Hz), 133.7 (d, *J* = 3.0 Hz), 131.1 (d, *J* = 9.0 Hz), 115.7 (d, *J* = 22.0 Hz), 26.6.



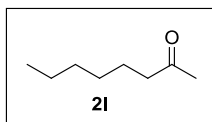
1-(2,4,6-Trimethylphenyl)ethanone (2i)⁷ as light yellow oil; Yield 76% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 6.84 (s, 1H), 2.46 (s, 3H), 2.27 (s, 3H), 2.22 (s, 6H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 19.3, 21.2, 32.4, 128.7, 132.5, 138.5, 140.0, 208.9.



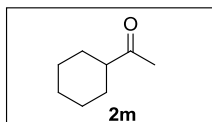
1-Phenylpropan-2-one (2j)⁸ as light yellow oil; Yield 69% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.31 (t, *J* = 8.0 Hz, 2H), 7.25 (d, *J* = 8.0 Hz, 1H), 7.18 (d, *J* = 8.0 Hz, 2H), 3.66 (s, 2H), 2.12 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 206.7, 134.4, 129.6, 128.9, 127.3, 51.2, 29.5.



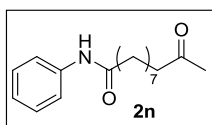
1-(1-indol-1-yl)pentane-1,4-dione (2k)⁹ as white solid; mp = 75-77 °C; Yield 89% (eluent: ethyl acetate/hexane = 1/10); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 8.40 (d, *J* = 8.0 Hz, 1H), 7.51-7.57 (m, 2H), 7.34-7.26 (m, 2H), 6.66 (d, *J* = 4.0 Hz, 1H), 3.23 (t, *J* = 6.1 Hz, 2H), 2.97 (t, *J* = 6.1 Hz, 2H), 2.29 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 206.9, 170.4, 135.8, 130.5, 125.3, 124.7, 123.9, 121.1, 116.7, 109.6, 37.5, 30.3, 29.8; HRMS (ESI): Exact mass calcd for C₁₃H₁₃O₂N [M+Na]⁺: 238.0839. Found: 238.0849.



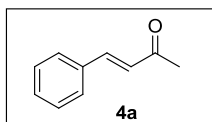
Nonan-2-one (2l)¹⁰ as light yellow oil; Yield 83% (eluent: ethyl acetate/hexane = 1/50); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 2.40 (t, *J* = 8.0 Hz, 2H), 2.11 (s, 3H), 1.55 (t, *J* = 4.0 Hz, 2H), 1.26 (s, 8H), 0.86 (t, *J* = 4.0 Hz, 3H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 209.6, 44.0, 31.8, 30.0, 29.0, 24.0, 22.7, 14.2.



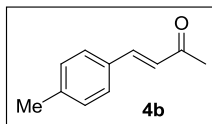
1-Cyclohexylethanone (2m)¹¹ as light yellow oil; Yield 78% (eluent: ethyl acetate/hexane = 1/50); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 2.33-2.25 (m, 1H), 2.13 (s, 3H), 1.77-1.85 (m, 4H), 1.18-1.34 (m, 6H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 212.5, 51.6, 28.6, 28.0, 26.0, 25.8.



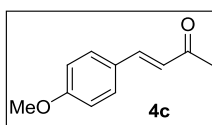
10-oxo-N-phenylundecanamide (2n) as white solid; mp = 52-54 °C; Yield 81% (eluent: ethyl acetate/hexane = 1/20); IR (KBr, cm⁻¹): 3345, 2929, 2851, 1704, 1687, 1600, 1543, 1499, 1440, 1310, 1161, 950, 897, 756, 699, 507; ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.52-7.58 (m, 2H), 7.30 (t, *J* = 8.0 Hz, 2H), 7.09 (t, *J* = 8.0 Hz, 1H), 2.41 (t, *J* = 8.0 Hz, 2H), 2.34 (t, *J* = 8.0 Hz, 2H), 2.13 (s, 3H), 1.70 (d, *J* = 8.0 Hz, 2H), 1.55 (s, 2H), 1.29 (s, 8H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 209.7, 171.7, 138.2, 129.0, 124.2, 119.9, 43.9, 37.8, 30.0, 29.24, 29.21, 29.1, 25.7, 23.8; HRMS (ESI): Exact mass calcd for C₁₇H₂₅O₂N [M+Na]⁺: 298.1778. Found: 298.1780.



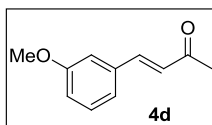
4-Phenylbut-3-en-2-one (4a)¹ as light yellow oil; Yield 87% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.53 (s, 2H), 7.49 (s, 1H), 7.40 (s, 3H), 6.72 (d, *J* = 16.0 Hz, 1H), 2.38 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 198.6, 143.6, 134.6, 130.7, 129.1, 128.4, 127.3, 27.7; HRMS (ESI): Exact mass calcd for C₁₁H₁₄O₂ [M+Na]⁺: 201.0886. Found: 201.0880.



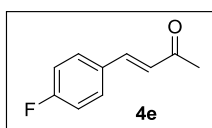
4-*p*-Tolylbut-3-en-2-one (4b)¹² as light yellow oil; Yield 72% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.51-7.43 (m, 3H), 7.26-7.20 (m, 2H), 6.68 (d, *J* = 16.0 Hz, 1H), 2.38 (s, 6H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 198.6, 143.6, 141.1, 131.8, 129.8, 128.4, 126.4, 27.5, 21.6; HRMS (ESI): Exact mass calcd for C₁₁H₁₂O [M+Na]⁺: 183.0780. Found: 183.0787.



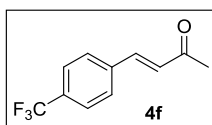
4-(4-MethoxyPhenyl)but-3-en-2-one (4c)¹ as light yellow oil; Yield 74% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.45-7.50 (m, 3H), 6.91 (d, *J* = 8.0 Hz, 2H), 6.60 (d, *J* = 16.0 Hz, 1H), 3.83 (s, 3H), 2.36 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 198.5, 161.6, 143.3, 130.0, 127.1, 125.0, 114.5, 55.4, 27.5.



4-(3-MethoxyPhenyl)but-3-en-2-one (4d)¹³ as light yellow oil; Yield 77% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.47 (dd, *J* = 16.0, 8.0 Hz, 1H), 7.31 (dd, *J* = 16.0, 8.0 Hz, 1H), 7.14 (d, *J* = 8.0 Hz, 1H), 7.06 (d, *J* = 8.0 Hz, 1H), 6.95 (d, *J* = 8.0 Hz, 1H), 6.69 (dd, *J* = 16.0, 8.0 Hz, 1H), 3.84 (s, 3H), 2.39 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 160.1, 143.6, 136.0, 130.2, 127.6, 121.2, 116.6, 113.2, 55.5, 27.7; HRMS (ESI): Exact mass calcd for C₁₁H₁₂O₂ [M+Na]⁺: 199.0730. Found: 199.0733.

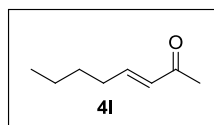


4-(4-FluoroPhenyl)but-3-en-2-one (4e)¹² as white solid; mp = 70-72 °C; Yield 77% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.55-7.43 (m, 3H), 7.09 (t, *J* = 8.0 Hz, 2H), 6.65 (d, *J* = 16.0 Hz, 1H), 2.38 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ (ppm) 198.4, 164.2 (d, *J* = 251.0 Hz), 142.3, 130.9, 130.3 (d, *J* = 9.0 Hz), 127.0, 116.3 (d, *J* = 22.0 Hz), 27.8; HRMS (ESI): Exact mass calcd for C₁₀H₉OF [M+Na]⁺: 187.0530. Found: 187.0532.

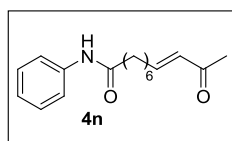


4-(4-TrifluoromethylPhenyl)but-3-en-2-one (4f)¹ as white solid; mp = 57-59 °C; Yield 75% (eluent: ethyl acetate/hexane = 1/100); ¹H NMR (400 MHz, CDCl₃): δ (ppm) 7.65 (s, 4H), 7.52 (d, *J* = 16.0 Hz, 1H), 6.78

(d, $J = 16.0$ Hz, 1H), 2.41 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 198.1, 141.5, 138.0, 132.1, (q, $J = 32.0$ Hz), 129.3, 128.5, 126.1 (q, $J = 3.0$ Hz), 124.0 (q, $J = 270.0$ Hz) 28.0; HRMS (ESI): Exact mass calcd for $\text{C}_{11}\text{H}_9\text{OF}_3$ $[\text{M}+\text{Na}]^+$: 237.0498. Found: 237.0508.



Non-3-en-2-one (4l)¹⁴ as light yellow oil; Yield 53% (eluent: ethyl acetate/hexane = 1/40); ^1H NMR (400 MHz, CDCl_3): δ (ppm) 6.77-6.85 (m, 1H), 6.07 (d, $J = 16.0$ Hz, 1H), 2.21-2.25 (m, 1H), 1.46 (t, $J = 8.0$ Hz, 2H), 1.37 (dd, $J = 16.0, 8.0$ Hz, 2H), 0.92 (t, $J = 8.0$ Hz, 3H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 199.0, 148.9, 131.5, 32.3, 30.3, 27.0, 22.4, 14.0; HRMS (ESI): Exact mass calcd for $\text{C}_8\text{H}_{14}\text{O}$ $[\text{M}+\text{Na}]^+$: 149.0937. Found: 149.0941.



10-oxo-N-phenylundec-8-enamide (4n) as white solid; mp = 90-92 °C; Yield 55% (eluent: ethyl acetate/hexane = 1/4); IR (KBr, cm^{-1}): 3320, 2930, 2855, 1667, 1599, 1531, 1500, 1443, 1366, 1260, 1184, 980, 759, 694, 501; ^1H NMR (400 MHz, CDCl_3): δ (ppm) 7.63 (s, 1H), 7.53 (d, $J = 8.0$ Hz, 2H), 7.30 (t, $J = 8.0$ Hz, 2H), 7.10 (d, $J = 8.0$ Hz, 1H), 6.76-6.83 (m, 1H), 6.06 (d, $J = 16.0$ Hz, 1H), 2.35 (t, $J = 8.0$ Hz, 2H), 2.21-2.24 (m, 5H), 1.72 (s, 2H), 1.47 (s, 2H), 1.37 (s, 4H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 199.1, 171.5, 148.7, 138.2, 131.4, 129.0, 124.2, 119.9, 37.7, 32.5, 29.1, 29.0, 27.9, 27.0, 25.5; HRMS (ESI): Exact mass calcd for $\text{C}_{17}\text{H}_{23}\text{O}_2\text{N}$ $[\text{M}+\text{Na}]^+$: 296.1621. Found: 296.1631.

5. References

- (1) Bigi, M. A.; White, M. C. *J. Am. Chem. Soc.* **2013**, *135*, 7831-7834.
- (2) Elings, J. A.; Downing, R. S.; Sheldon, R. A. *Eur. J. Org. Chem.* **1999**, *4*, 837-846.
- (3) Olah, G. A.; Krishnamurthy, V. V.; Singh, B. P.; Iyer, P. S. *J. Org. Chem.* **1983**, *48*, 955-63.
- (4) Zhang, J.; Jiang, Z.; Zhao, D.; He, G.; Zhou, S.; Han, S. *Chin. J. Chem.* **2013**, *31*, 794-798.
- (5) Wang, X.; Wang, X.; Liu, M.; Ding, J.; Chen, J.; Wu H. *Synthesis* **2013**, *45*, 2241-2244.
- (6) Zhang, G.; Han, X.; Luan, Y.; Wang, Y.; Wen, X.; Ding, C. *Chem. Commun.* **2013**, *49*, 7908-7910.
- (7) Sharghi, H.; Jokar, M.; Doroodmand, M. M.; Khalifeh, R. *Adv. Synth. Catal.* **2010**, *352*, 3031-3044.
- (8) Hamada, S.; Furuta, T.; Wada, Y.; Kawabata, T. *Angew. Chem., Int. Ed.* **2013**, *52*, 8093-8097.
- (9) Beemelmans, C.; Blot, V.; Gross, S.; Lentz, D.; Reissig, H.-U. *Eur. J. Org. Chem.* **2010**, 2716-2732.
- (10) Miranda, P. O.; D áz, D. D.; Padr ón, J. I.; Ram íez, M. A.; Mart ín, V. S. *J. Org. Chem.* **2005**, *70*, 57-62.
- (11) Chen, Z.; Ye, D.; Qian, Y.; Ye, M.; Liu, L. *Tetrahedron* **2013**, *69*, 6116-6120.
- (12) Du, Z.; Zhang, L.; Fan, X.; Wu, F.; Da, C. *Tetrahedron Lett.* **2013**, *54*, 2828-2832.
- (13) Sharma, N.; Sharma, U. K.; Kumar, R.; Katoch, N.; Kumar, R.; Sinha, A. K. *Adv. Synth. Catal.* **2011**, *353*, 871-878.
- (14) Abbas, M.; Leitgeb, A.; Slugovc, C. *Synlett* **2013**, *24*, 1193-1196.

6. Copies of ^1H NMR and ^{13}C NMR Spectra

