## Synthesis, Characterization, and Catalytic Activity of a Series of Aluminium-Amidate Complexes

Kevin P. Yeagle,<sup>*A*</sup> Darryl Hester,<sup>*A*</sup> Nicholas A. Piro,<sup>*B*</sup> William G. Dougherty,<sup>*B*</sup> W. Scott Kassel,<sup>*B*</sup> and Christopher R. Graves<sup>*A*,*C*</sup> <sup>A</sup>Department of Chemistry & Biochemistry, Albright College, 13th & Bern St., Reading, PA 19612, United

States

<sup>B</sup>Department of Chemistry, Villanova University, 800 Lancaster Ave., Villanova, PA 19085, United States <sup>C</sup>Corresponding author. Email: cgraves@alb.edu

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**Figure S4.** <sup>1</sup>H NMR spectra of { $[\kappa^2-N, O-(t-BuNCOPh)]AlMe_2$ }<sub>2</sub> (2). Taken in CDCl<sub>3</sub> at 25 °C.



**Figure S5.** <sup>13</sup>C NMR spectra of { $[\kappa^2-N, O-(t-BuNCOPh)]AlMe_2$ }<sub>2</sub> (**2**). Taken in CDCl<sub>3</sub> at 25 °C.





**Figure S7.** <sup>13</sup>C NMR spectra of  $[\kappa^2 - N, O - (t - BuNCOPh)]_2AlMe(3)$ . Taken in CDCl<sub>3</sub> at 25 °C.



**Figure S8.** <sup>1</sup>H NMR spectra of  $[\kappa^2 - N, O - (t - BuNCOPh)]_3A1(4)$ . Taken in CDCl<sub>3</sub> at 25 °C.





**Figure S10.** Gas chromatograph of the acetophenone and *sec*-phenethyl alcohol mixture. Initial Temperature = 40 °C, Initial time = 2 min; Ramp Rate 1 = 5 °C/min to 100 °C Hold Time = 0 min; Ramp Rate 2 = 25 °C/min to 250 °C, Final Time = 2 min.



**Figure S11.** Gas chromatograph of the 2-napthanone and 1-(2-naphthyl)ethanol mixture. Initial Temperature = 50 °C, Initial time = 4 min; Ramp Rate 1 = 20 °C/min to 150 °C Hold Time = 5 min; Ramp Rate 2 = 5 °C/min to 250 °C, Final Time = 2 min.



**Figure S12.** Gas chromatograph of the 4-phenyl-3-buten-2-one and 4-phenyl-3-buten-2ol mixture. Initial Temperature = 40 °C, Initial time = 2 min; Ramp Rate 1 = 5 °C/min to 100 °C Hold Time = 0 min; Ramp Rate 2 = 25 °C/min to 250 °C, Final Time = 2 min.



**Figure S13.** Gas chromatograph of the 4-phenyl-3-butyne-2-one and 4-phenyl-3-butyne-2-ol mixture. Initial Temperature = 40 °C, Initial time = 2 min; Ramp Rate 1 = 5 °C/min to 100 °C Hold Time = 0 min; Ramp Rate 2 = 25 °C/min to 250 °C, Final Time = 2 min.



**Figure S14.** Gas chromatograph of the cyclohexanone and cyclohexanol mixture. Initial Temperature = 40 °C, Initial time = 2 min; Ramp Rate 1 = 5 °C/min to 100 °C Hold Time = 0 min; Ramp Rate 2 = 25 °C/min to 250 °C, Final Time = 2 min.



**Figure S15.** Gas chromatograph of the benzaldehyde and benzyl alcohol mixture. Initial Temperature = 40 °C, Initial time = 2 min; Ramp Rate 1 = 5 °C/min to 100 °C Hold Time = 0 min; Ramp Rate 2 = 25 °C/min to 250 °C, Final Time = 2 min.



**Figure S16.** Gas chromatograph of the cyclohexanecarboxaldehyde and cyclohexanemethanol mixture. Initial Temperature = 40 °C, Initial time = 2 min; Ramp Rate 1 = 5 °C/min to 100 °C Hold Time = 0 min; Ramp Rate 2 = 25 °C/min to 250 °C, Final Time = 2 min.



**Figure S17.** <sup>1</sup>H NMR spectra of *sec*-phenethyl alcohol obtained in the large scale reduction of acetophenone. Taken in CDCl<sub>3</sub> at 25  $^{\circ}$ C.



**Figure S18.** <sup>1</sup>H NMR spectra of acetophenone obtained in the large scale oxidation of *sec*-phenethyl alcohol. Taken in  $CDCl_3$  at 25 °C.