

Supporting Information

Synthesis and Characterization of Primary Aluminum Parent

Amides and Phosphides

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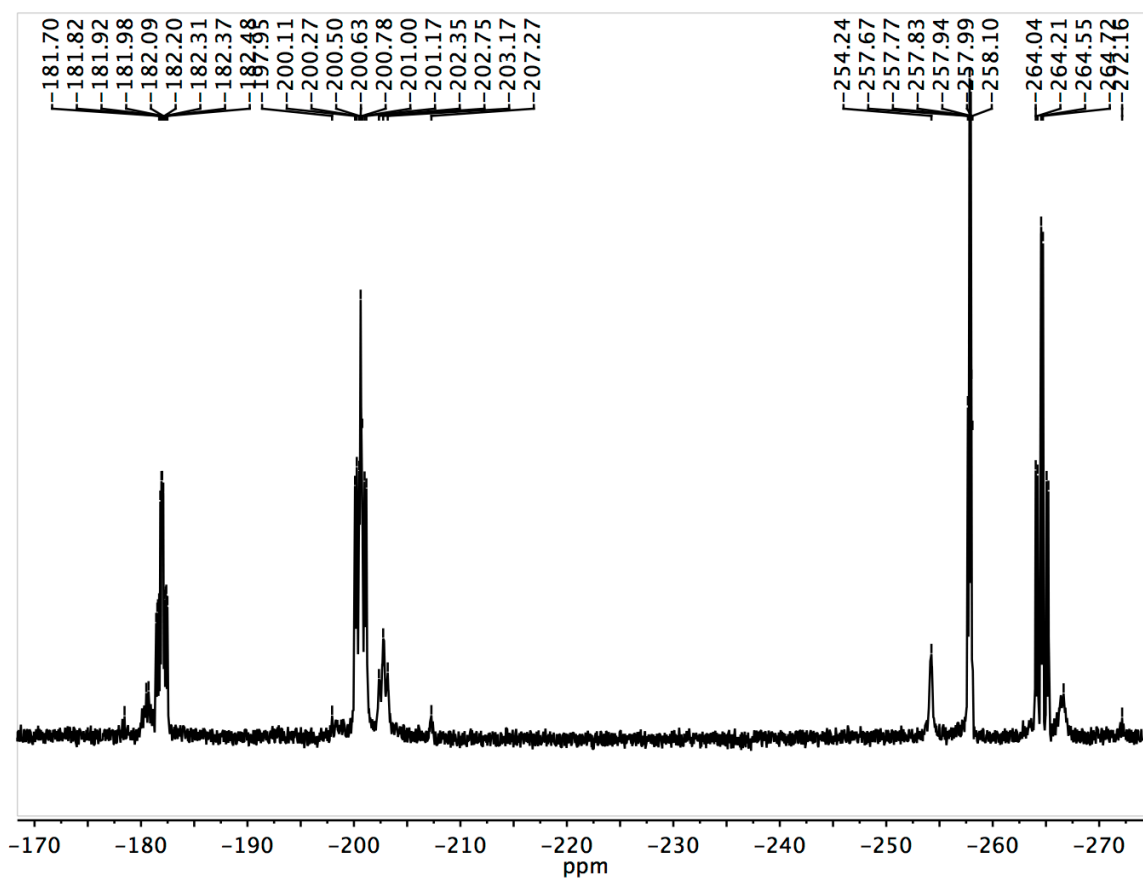
S14: ^1H NMR spectrum $\text{LiAlH}_3\text{Ar}^{\text{Pr}^i4}$ in C_6D_6 at 25 °C.

S15: ^1H NMR spectrum of $\text{LiAlH}_3\text{Ar}^{\text{Pr}^i8}$ in C_6D_6 at 25 °C.

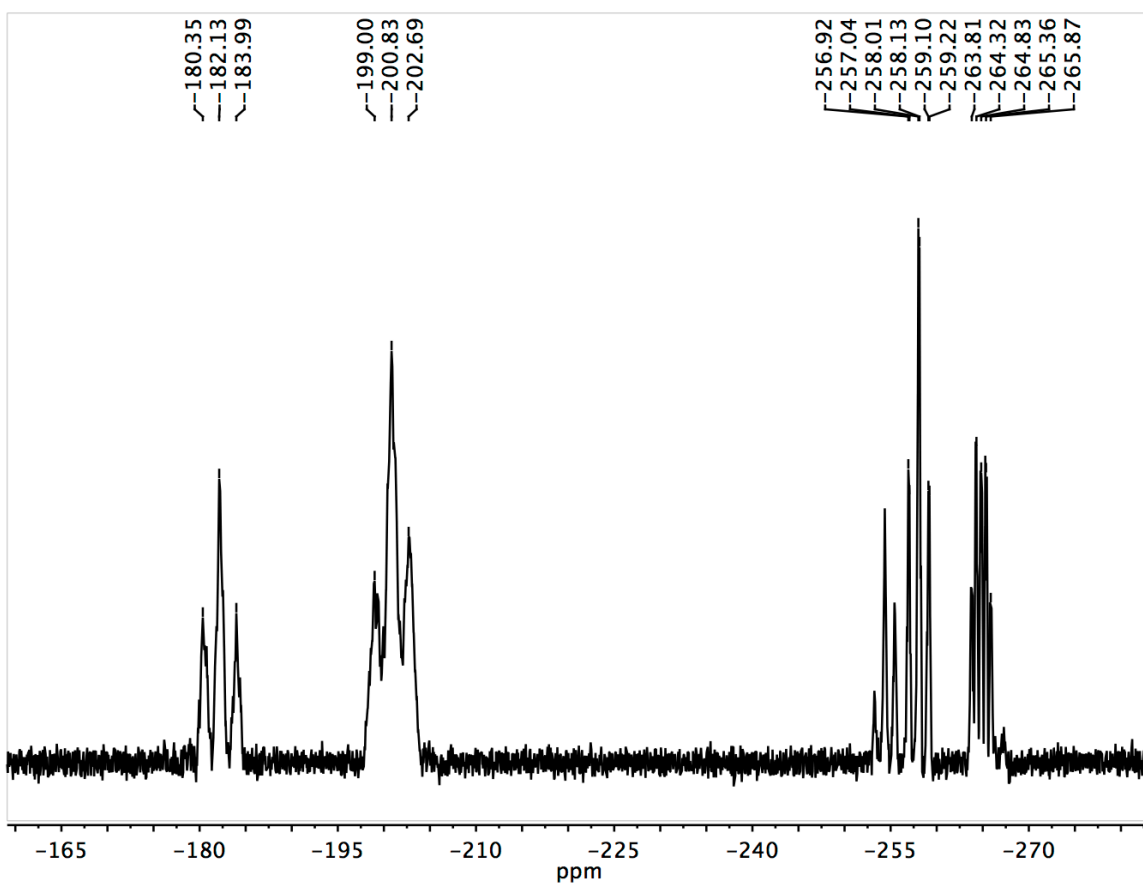
S16: ^1H NMR spectrum of **1** in C_6D_6 at 25 °C.

S17: ^1H NMR spectrum of **3** in C_6D_6 at 25 °C.

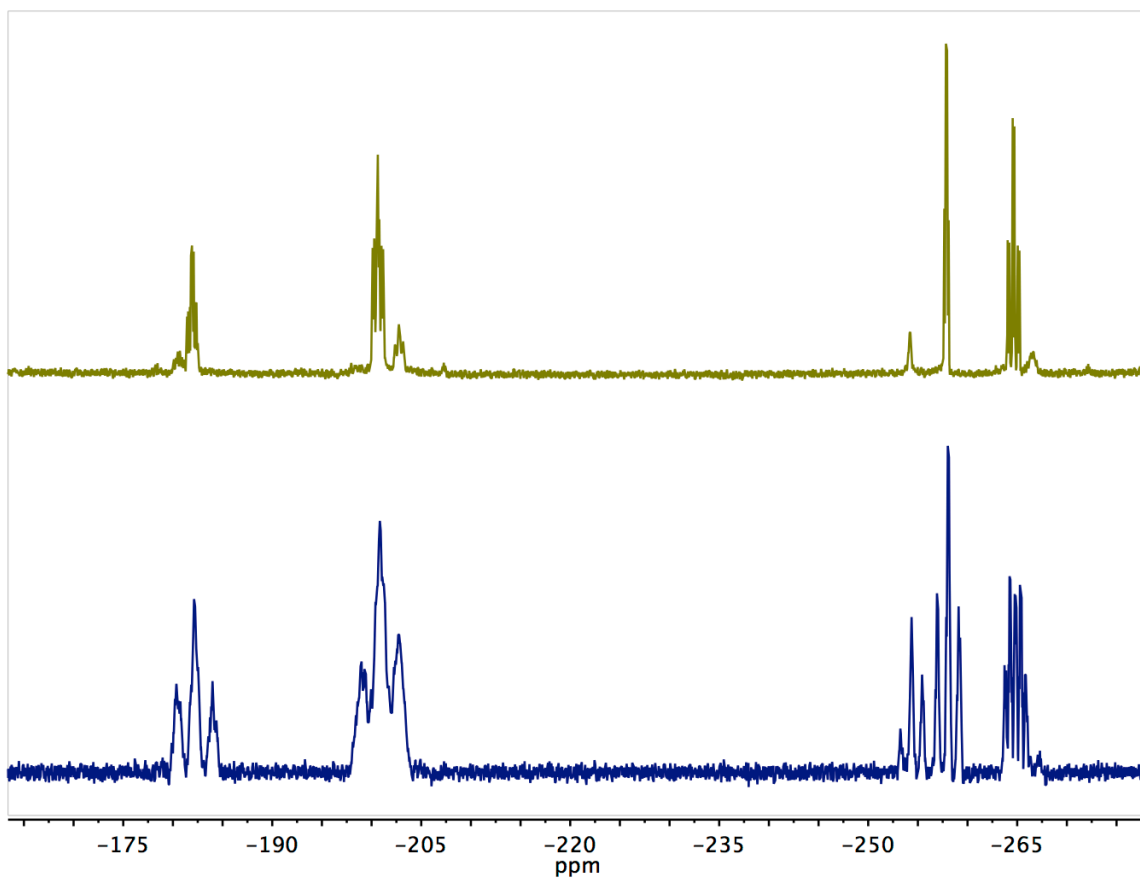
S18: ^1H NMR spectrum of **2** in C_6D_6 at 25 °C.



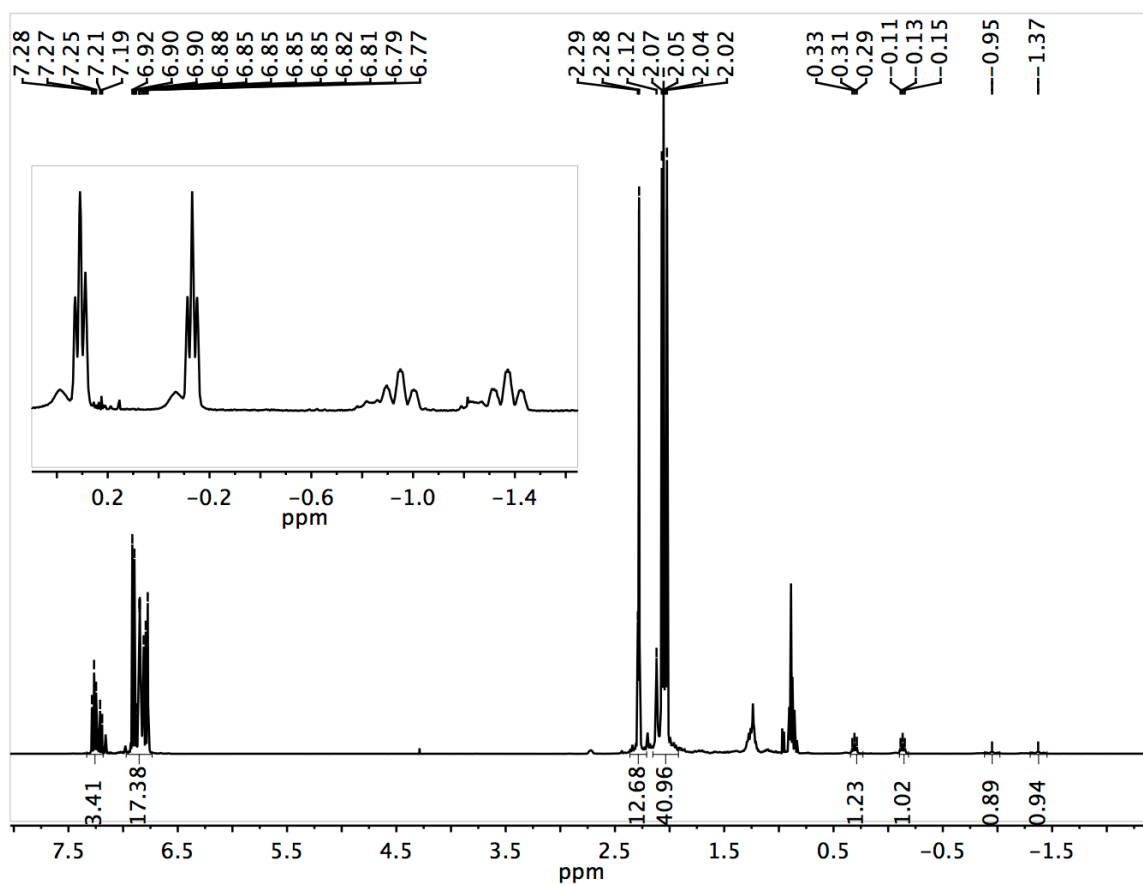
S1: Full $^{31}\text{P}\{^1\text{H}\}$ NMR spectrum of the redissolved powder from the crude solid compound **4**.



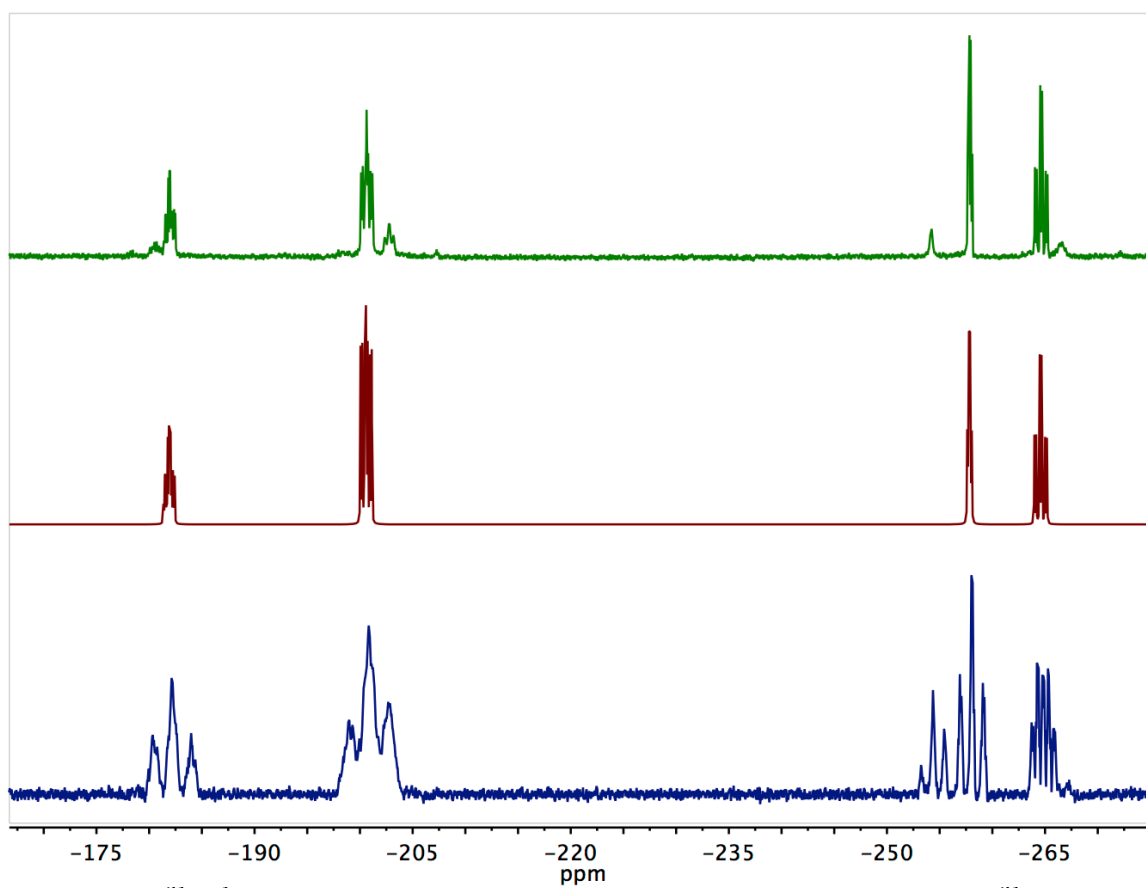
S2: Full ^{31}P NMR spectrum of the redissolved powder from the crude solid of compound **4**.



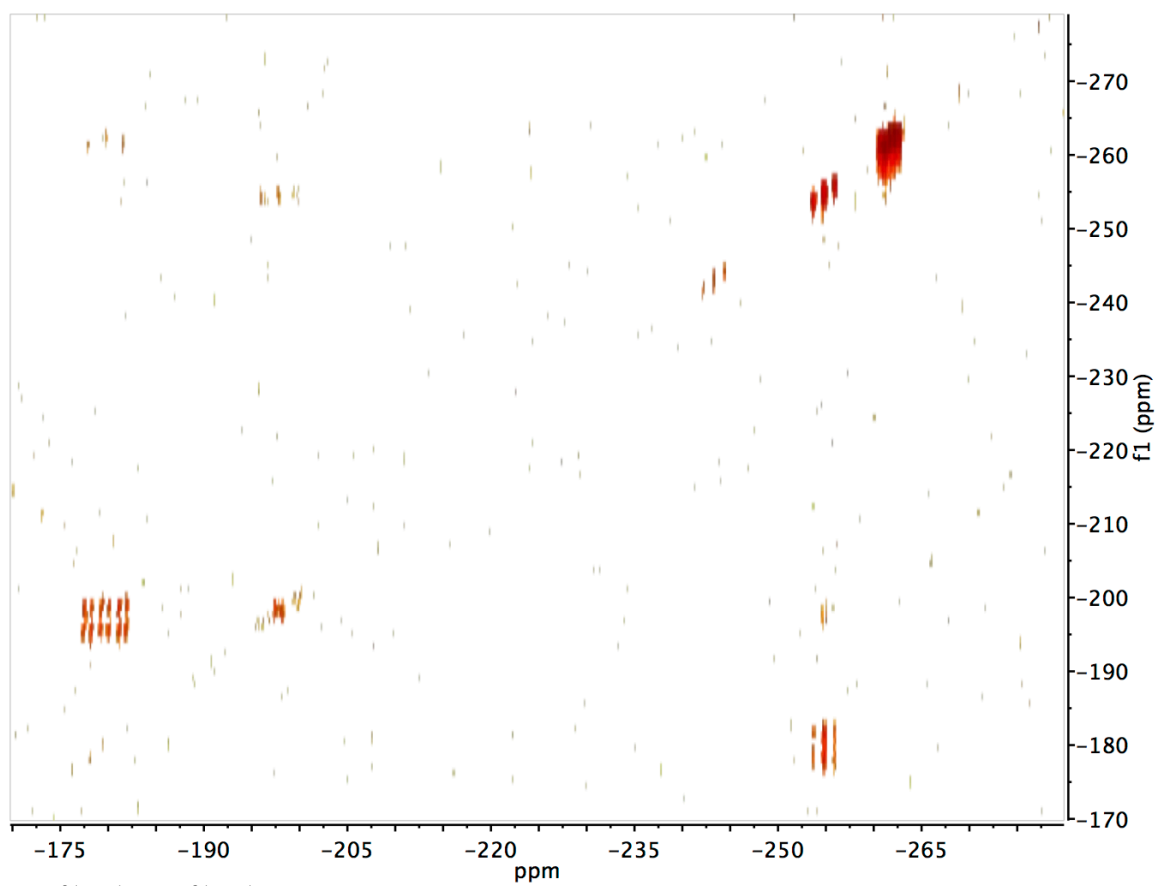
S3: Plots of the $^{31}\text{P}\{^1\text{H}\}$ NMR spectrum (top) and ^{31}P NMR spectrum (bottom) for compound **4**.



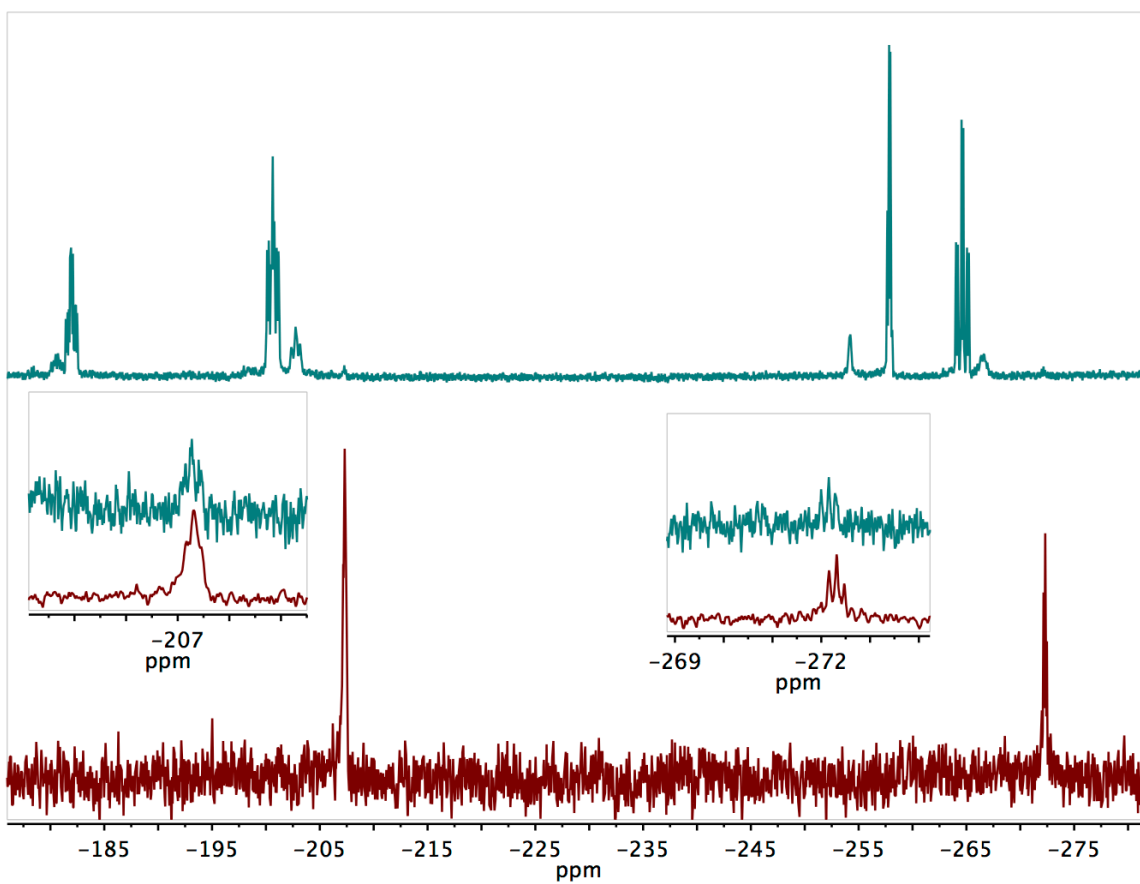
S4: ^1H NMR spectrum for crystallized compound **4**.



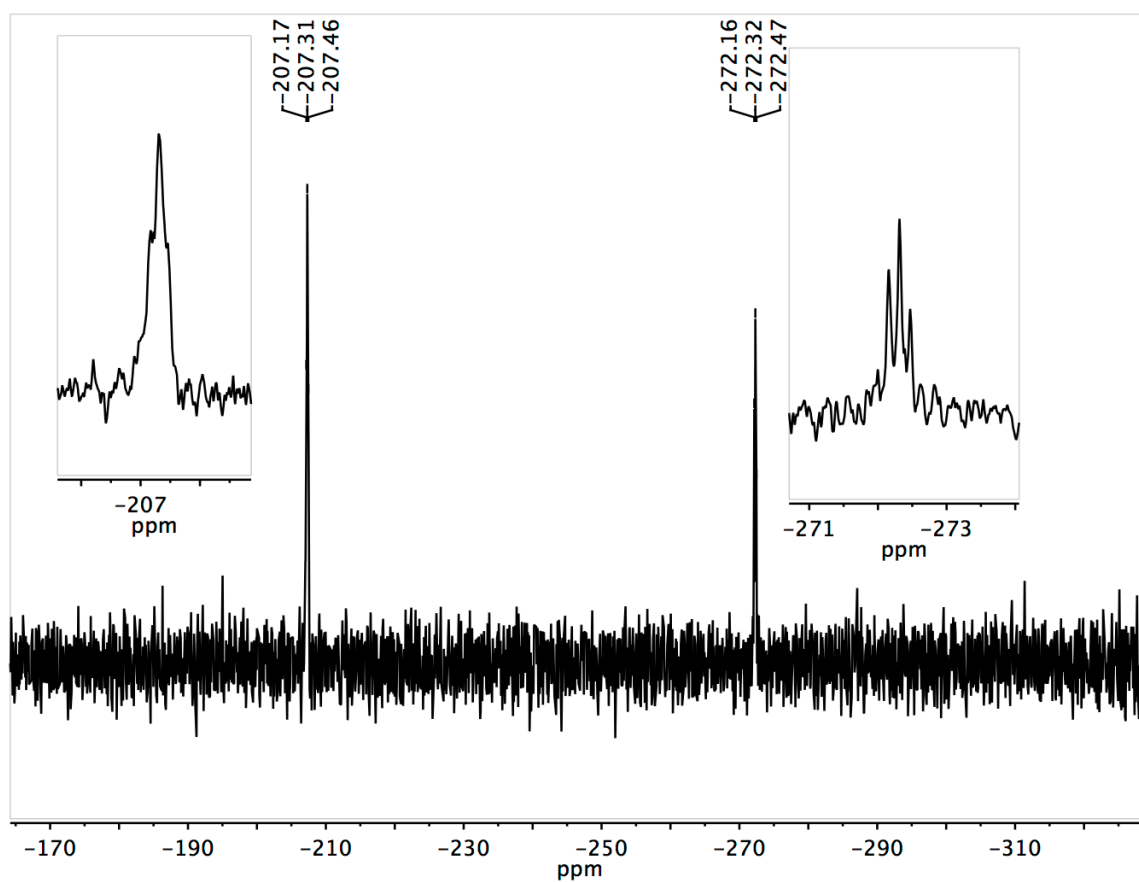
S5: Plots of $^{31}\text{P}\{^1\text{H}\}$ NMR spectrum (top), simulated spectrum (middle), and ^{31}P NMR spectrum of compound **4** (bottom).



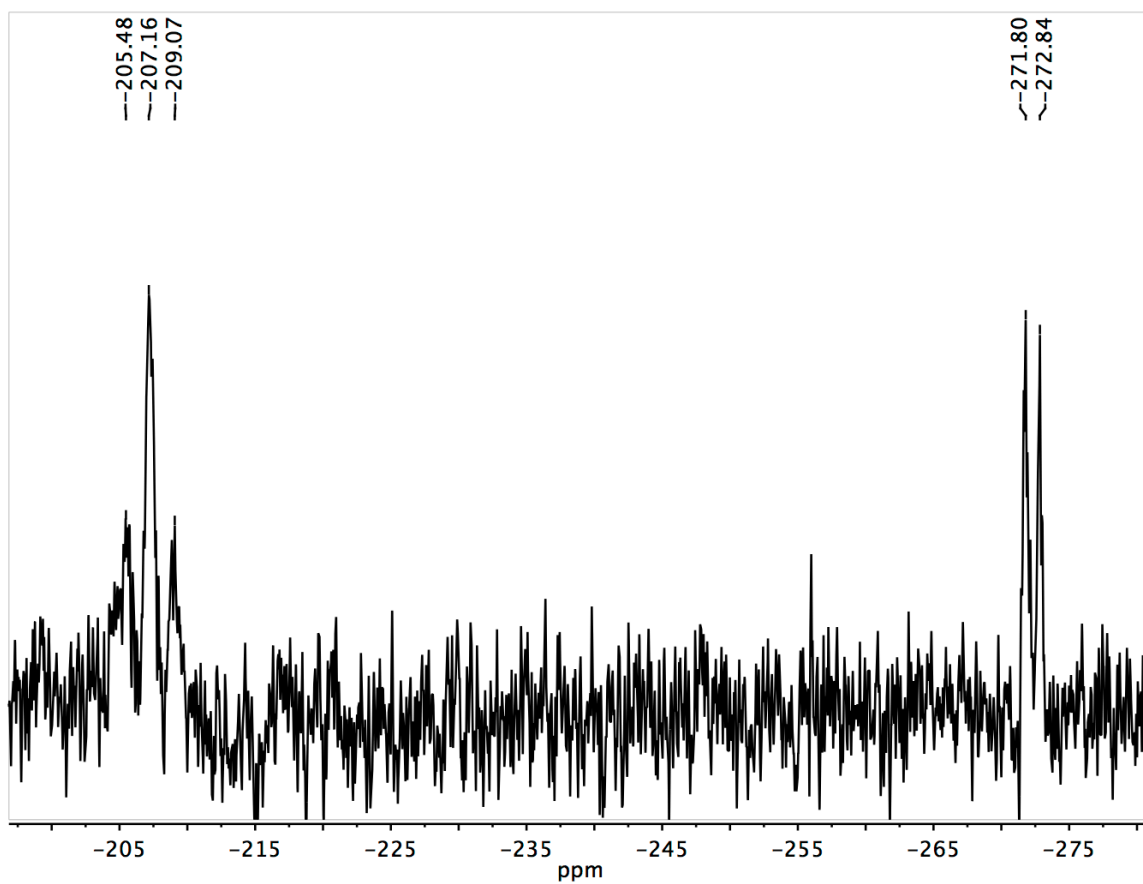
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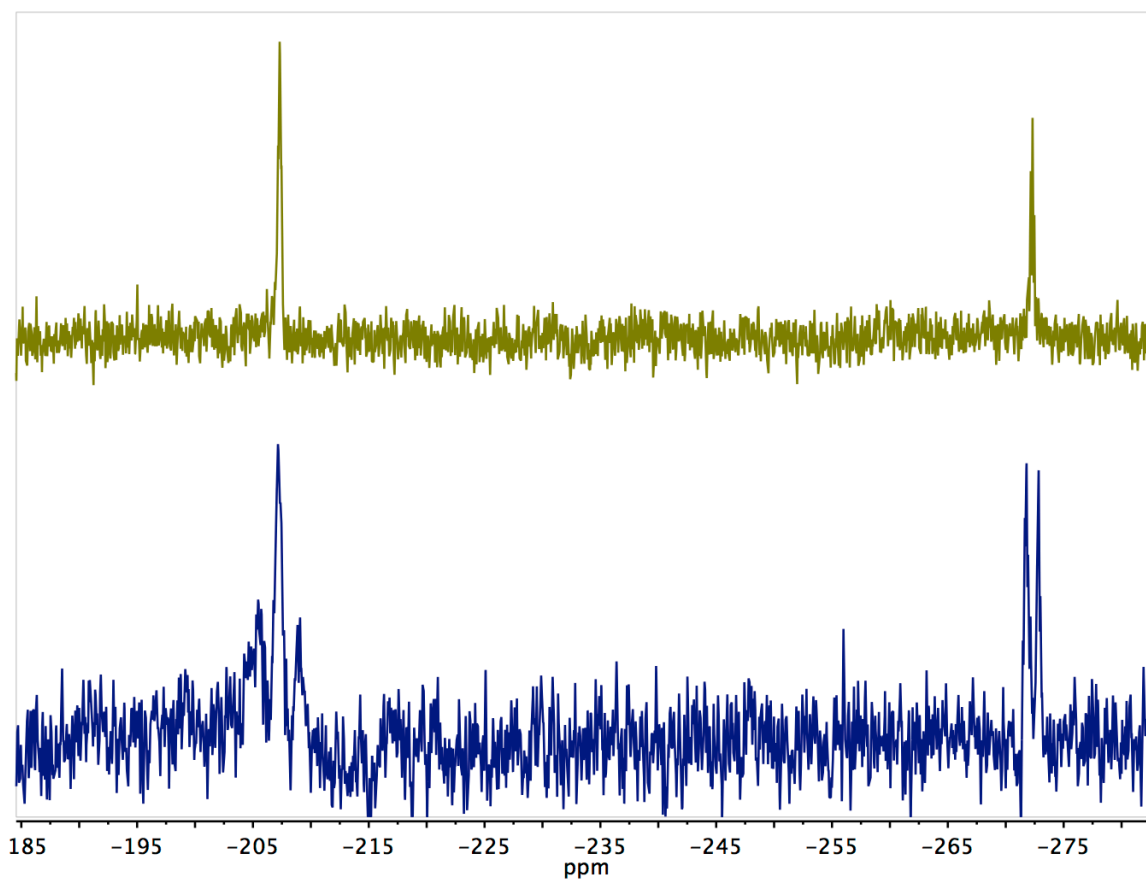
S7: Plots of the $^{31}\text{P}\{^1\text{H}\}$ NMR spectra of the crude solid compound **4** and **5** (top) and small amount of isolated compound **5** (bottom).



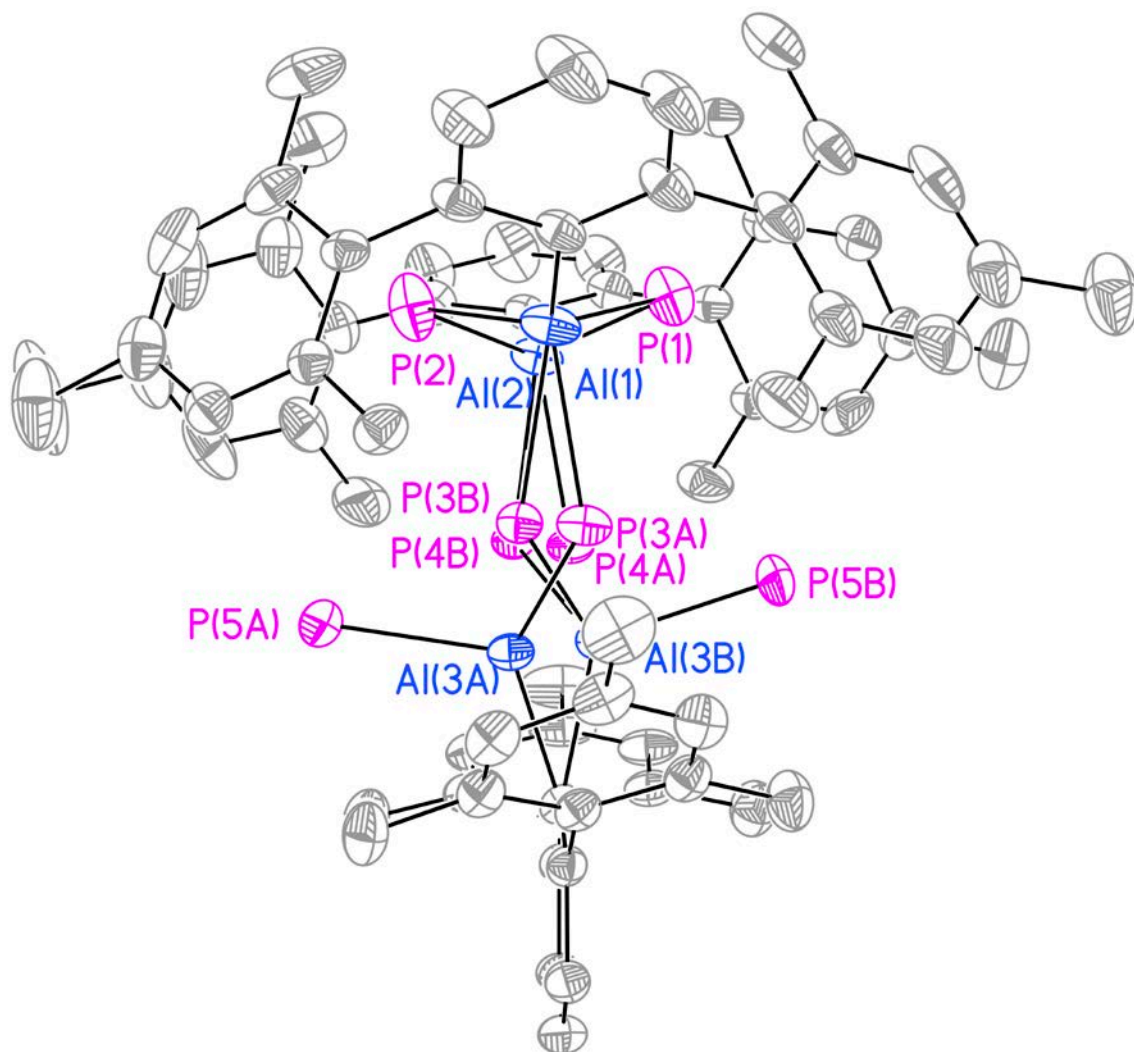
S8: $^{31}\text{P}\{^1\text{H}\}$ NMR spectrum of small amount of isolated compound **5**.



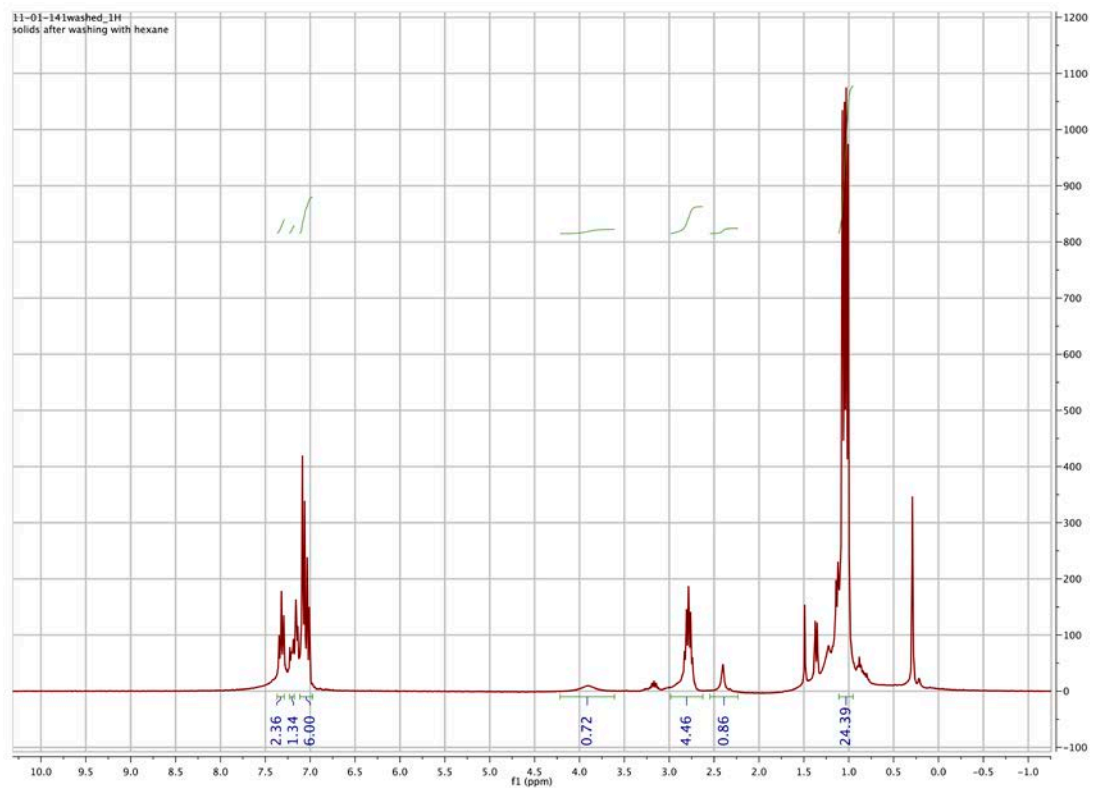
S9: ^{31}P NMR spectrum of small amount of isolated compound **5**.



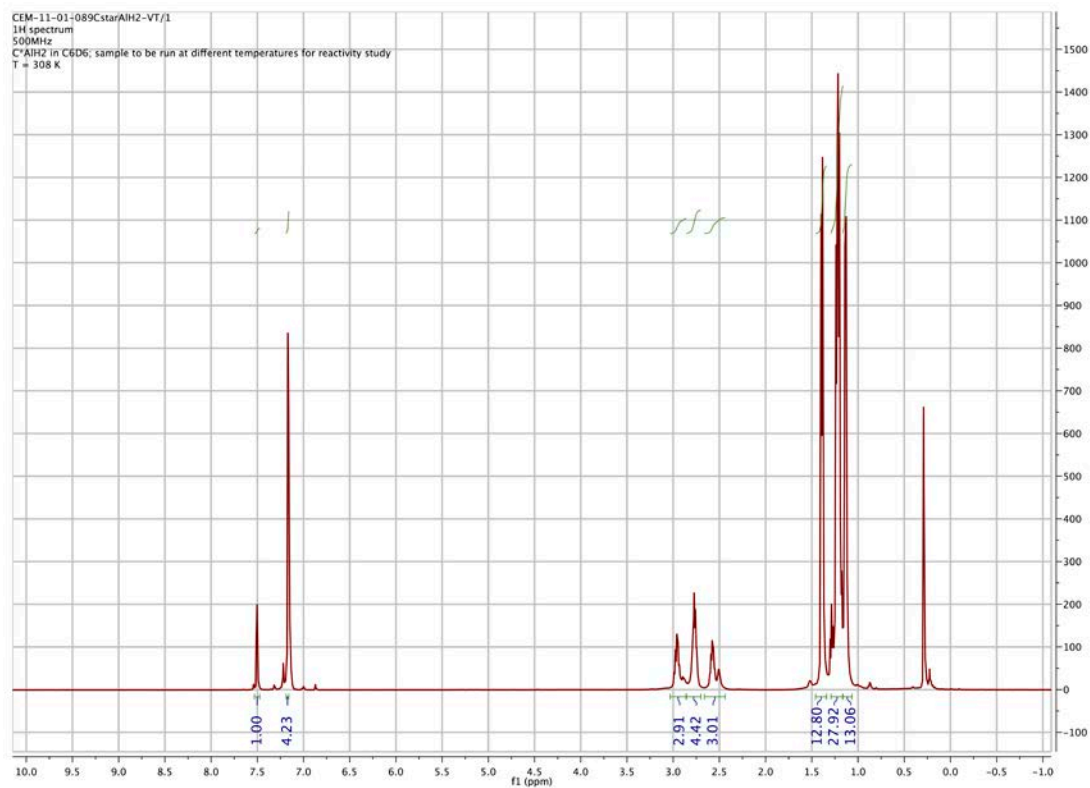
S10: Plots of the $^{31}\text{P}\{^1\text{H}\}$ NMR spectrum (top) and ^{31}P NMR spectrum (bottom) for compound **5**.



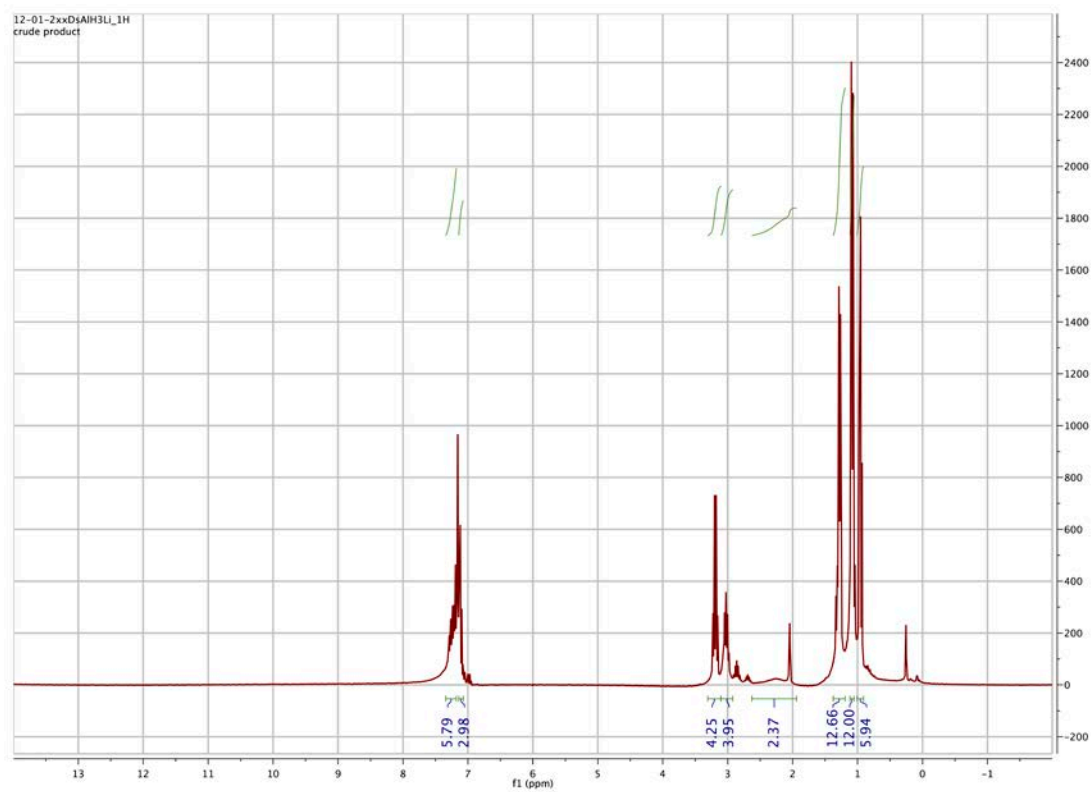
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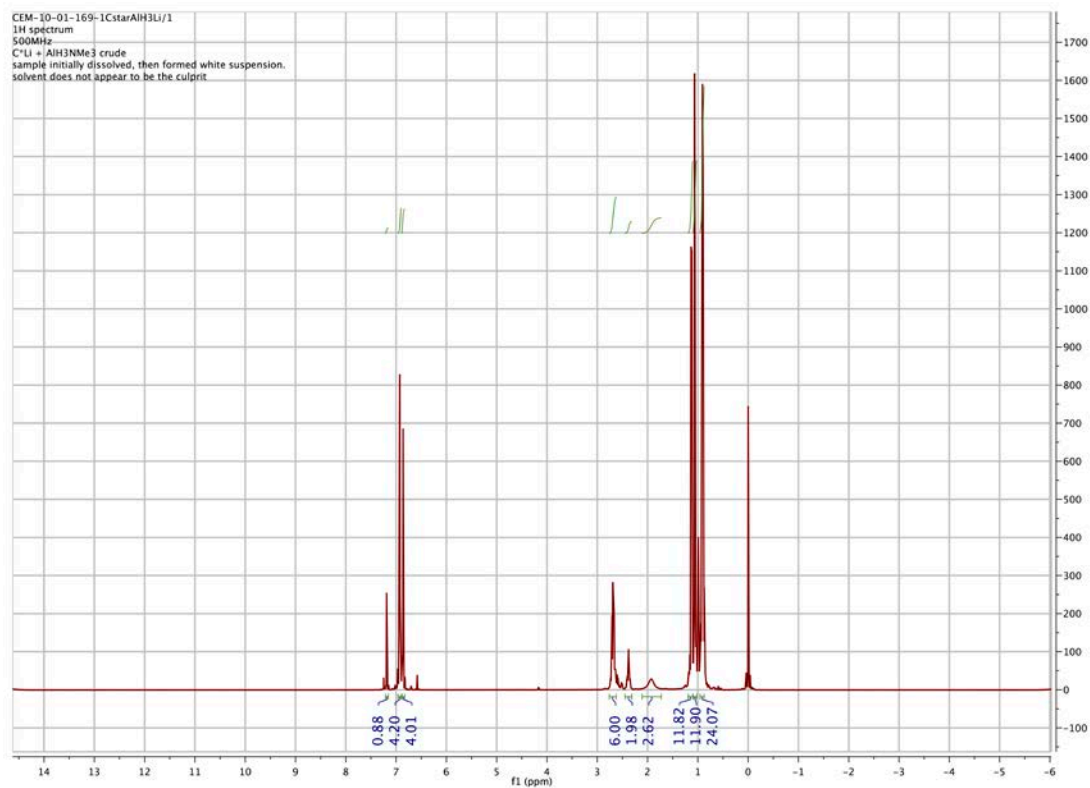
S12: ^1H NMR spectrum of $(\text{Ar}^{\text{Pr}^i}_4\text{AlH}_2)_2$ in C_6D_6 at 25°C .



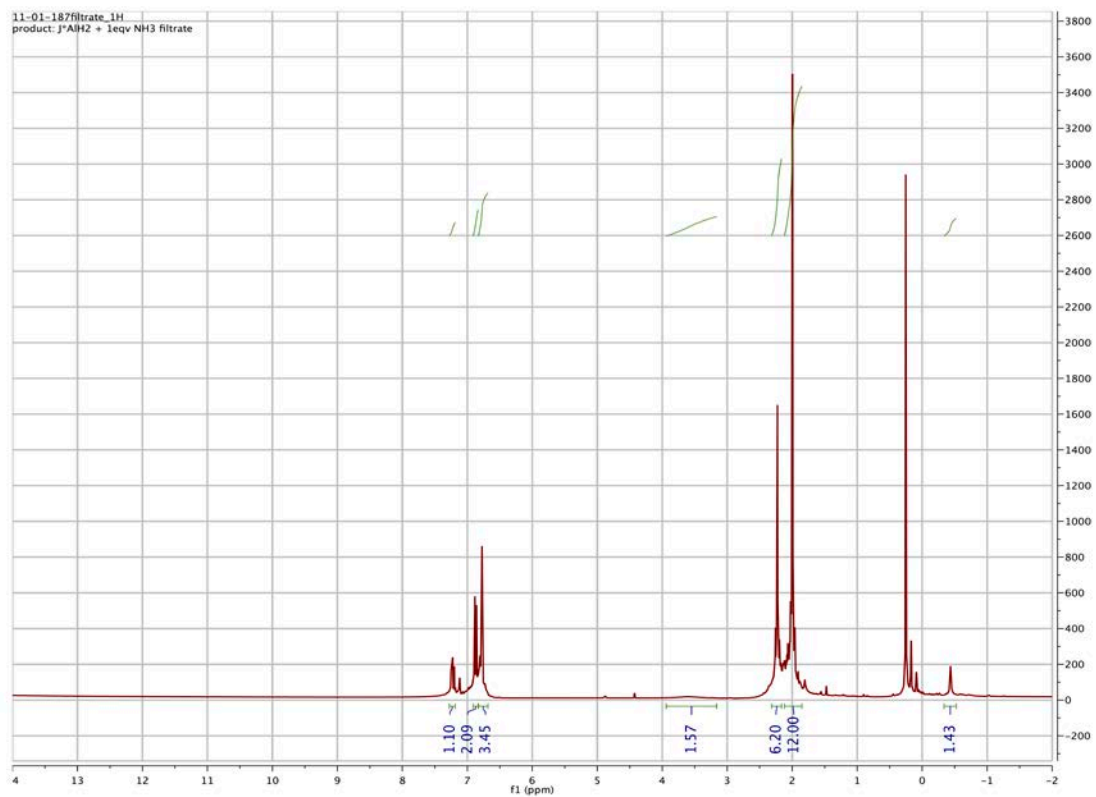
S13: ^1H NMR spectrum of $(\text{Ar}^{\text{Pr}^8}\text{AlH}_2)_2$ in C_6D_6 at 25 °C.



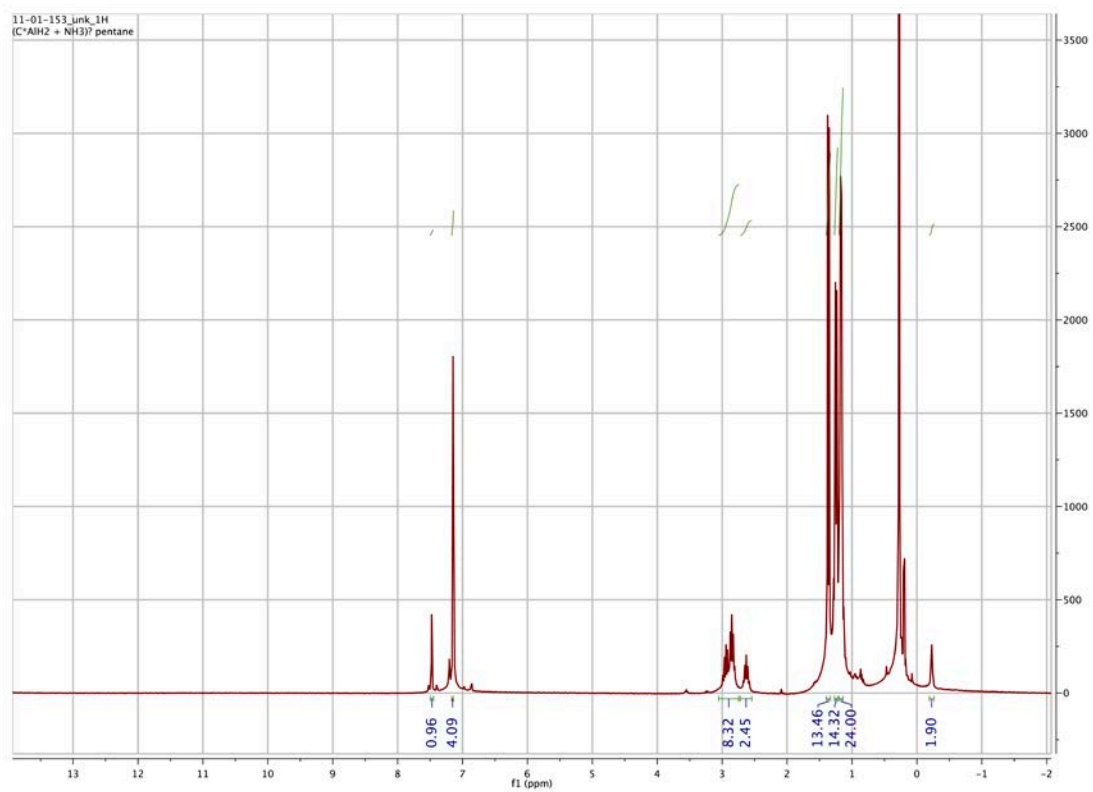
S14: ^1H NMR spectrum $\text{LiAlH}_3\text{Ar}^{\text{Pr}^4}$ in C_6D_6 at $25\text{ }^\circ\text{C}$.



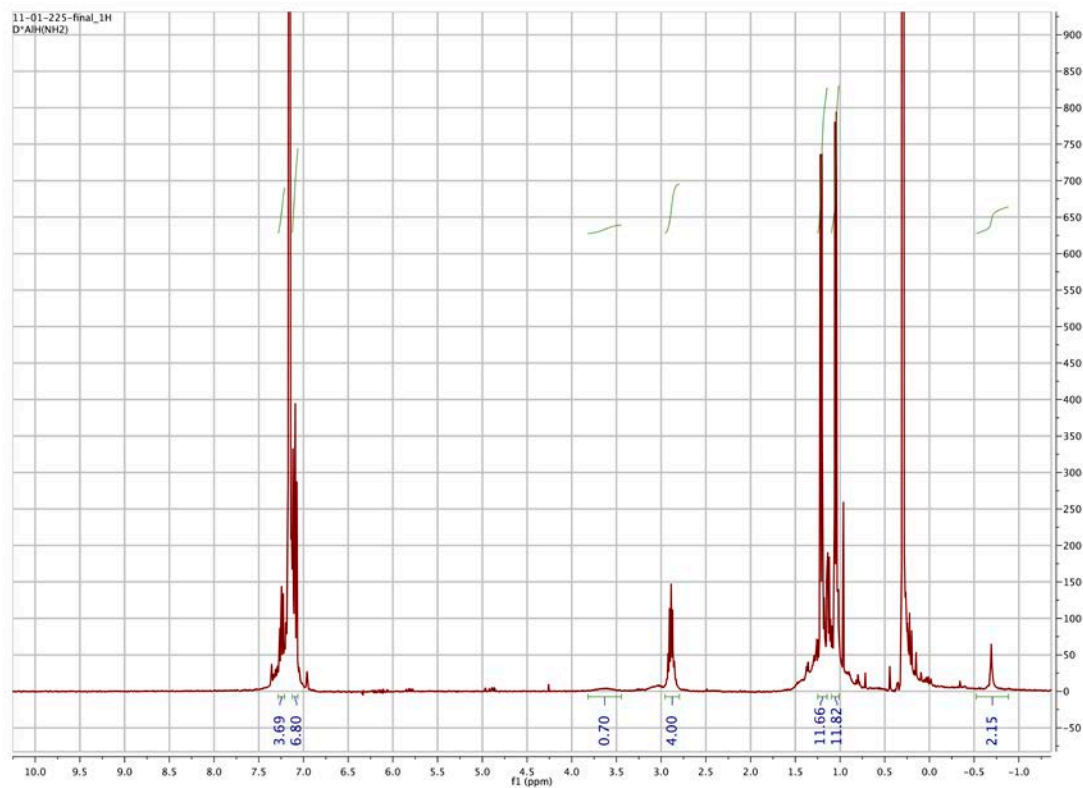
S15: ^1H NMR spectrum of $\text{LiAlH}_3\text{Ar}^{\text{Pr}^i}_8$ in C_6D_6 at 25 °C.



S16: ^1H NMR spectrum of **1** in C_6D_6 at 25°C .



S17: ¹H NMR spectrum of **3** in C₆D₆ at 25 °C.



S18: ^1H NMR spectrum of **2** in C_6D_6 at 25 $^\circ\text{C}$.