Supporting Information
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Structurally Stimulated Deprotonation/Alumination of the TMP Anion**

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General Methods

Hexane was distilled over sodium benzophenone. TMEDA was distilled and stored over 4Å molecular sieves. nBuLi and iBu₂AlCl were purchased from Aldrich and used as received. TMP(H) was purchased from Merck and dried over 4Å molecular sieves prior to use. All synthetic work was carried out under an inert Argon atmosphere using standard Schlenk techniques. Data for the X-ray crystal structure determination of 2 was obtained with a Nonius Kappa CCD Diffractometer using graphite monochromated Cu-Kα radiation (λ = 1.5418Å). ¹H and ¹³C NMR spectra were obtained on a Bruker DPX400 spectrometer operating at a frequency of 400.13 and 100.62 MHz respectively. All chemical shifts are quoted relative to TMS standard at 0.00 parts per million. ¹H-¹H and ¹H-¹³C correlations were identified using COSY and HSQC NMR techniques respectively. Due to the air sensitive nature of product 2, elemental analysis could not be satisfactorily obtained.

Spectrum 1 ¹H NMR spectrum of 2 in deuterated cyclohexane solution
Figure 1 Molecular structure of the dimeric form of 2 [the CH(Me)$_2$ of $^i$Bu groups have been removed for clarity].