Supporting Information

Computational prediction and analysis of the 27Al solid-state NMR spectrum of methylaluminoxane (MAO) at variable temperatures and field strengths

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FIG. S1. SSNMR spectra of MAO at 398 K absent free TMA, simulated at different field strengths (the corresponding proton resonance frequencies are also provided). The Boltzmann averaged contributions to the spectra from h-h-h, s-h-h and s-s-h Al sites are given separately. Species with 3-coordinate, 4-coordinate, or h-h-o aluminum sites are not predicted to be stable at this temperature.
FIG. S2. SSNMR spectra of MAO at 498 K absent free TMA, simulated at different field strengths (the corresponding proton resonance frequencies are also provided). The Boltzmann averaged contributions to the spectra from Al sites are given separately. Species with 3-coordinate, 4-coordinate, or h-h-o aluminum sites are not predicted to be stable at this temperature.
FIG. S3. SSNMR spectra of MAO at 77 K absent free TMA, simulated at different field strengths (the corresponding proton resonance frequencies are also provided) with various Gaussian broadening frequencies applied. The Boltzmann averaged contributions to the spectra from Al sites are given separately.
FIG. S4. SSNMR spectra of MAO at 298 K absent free TMA, simulated at different field strengths (the corresponding proton resonance frequencies are also provided) with various Gaussian broadening frequencies applied. The Boltzmann averaged contributions to the spectra from Al sites are given separately.
FIG. S5. SSNMR spectra of MAO at 398 K absent free TMA, simulated at different field strengths (the corresponding proton resonance frequencies are also provided) with various Gaussian broadening frequencies applied. The Boltzmann averaged contributions to the spectra from Al sites are given separately.
FIG. S6. SSNMR spectra of MAO at 498 K absent free TMA, simulated at different field strengths (the corresponding proton resonance frequencies are also provided) with various Gaussian broadening frequencies applied. The Boltzmann averaged contributions to the spectra from Al sites are given separately.
FIG. S7. SSNMR spectra of \((\text{AlMe}_3)_2\) and \((\text{AlMe}_3)_3\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S8. Calculated EFG parameters and NMR chemical shifts of TMA monomer and dimer.
FIG. S9. SSNMR spectra of \((\text{AlOMe})_{6,c}\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S10. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{6,c}\).
FIG. S11. SSNMR spectra of (AlOMe)$_6$·(AlMe$_3$), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).
FIG. S12. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_6$·(AlMe)$_3$.

- $\mathbf{C_Q} = 21.2$ MHz, $\eta = 0.78$, $\delta = 115$ ppm
- $\mathbf{C_Q} = 17.6$ MHz, $\eta = 0.09$, $\delta = 100$ ppm
- $\mathbf{C_Q} = 20.9$ MHz, $\eta = 0.53$, $\delta = 140$ ppm
- $\mathbf{C_Q} = 20.3$ MHz, $\eta = 0.79$, $\delta = 113$ ppm
- $\mathbf{C_Q} = 42.7$ MHz, $\eta = 0.60$, $\delta = 224$ ppm
- $\mathbf{C_Q} = 17.9$ MHz, $\eta = 0.09$, $\delta = 100$ ppm
- $\mathbf{C_Q} = 21.0$ MHz, $\eta = 0.78$, $\delta = 115$ ppm
FIG. S13. SSNMR spectra of \((\text{AlOMe})_{6,c} \cdot (\text{AlMe}_3)_2\) simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S14. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{6,c} \cdot (\text{AlMe}_3)_2\).
FIG. S15. SSNMR spectra of (AlOMe)$_{6,c}$·(AlMe$_3$)$_3$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

![SSNMR Spectra Diagram]

FIG. S16. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{6,c}$·(AlMe$_3$)$_3$. 

![Calculated EFG Parameters Diagram]
FIG. S17. SSNMR spectra of \((\text{AlOMe})_{6,c} \cdot \text{(AlMe}_3\text{)}_4\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S18. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{6,c} \cdot \text{(AlMe}_3\text{)}_4\).

\[
\begin{align*}
\text{CQ} &= 16.0 \text{ MHz} \\
\eta &= 0.22 \\
\delta &= 84 \text{ ppm} \\
\text{CQ} &= 17.4 \text{ MHz} \\
\eta &= 0.64 \\
\delta &= 115 \text{ ppm} \\
\end{align*}
\]

\[
\begin{align*}
\text{CQ} &= 15.9 \text{ MHz} \\
\eta &= 0.20 \\
\delta &= 84 \text{ ppm} \\
\text{CQ} &= 25.2 \text{ MHz} \\
\eta &= 0.90 \\
\delta &= 161 \text{ ppm} \\
\end{align*}
\]

\[
\begin{align*}
\text{CQ} &= 17.4 \text{ MHz} \\
\eta &= 0.56 \\
\delta &= 115 \text{ ppm} \\
\text{CQ} &= 25.6 \text{ MHz} \\
\eta &= 0.90 \\
\delta &= 161 \text{ ppm} \\
\end{align*}
\]

\[
\begin{align*}
\text{CQ} &= 17.4 \text{ MHz} \\
\eta &= 0.56 \\
\delta &= 115 \text{ ppm} \\
\text{CQ} &= 25.6 \text{ MHz} \\
\eta &= 0.90 \\
\delta &= 161 \text{ ppm} \\
\end{align*}
\]
FIG. S19. SSNMR spectra of (AlOMe)$_8$,$_c$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S20. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_8$,$_c$.
FIG. S21. SSNMR spectra of (AlOMe)$_{8,c}$·(AlMe$_3$), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S22. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{8,c}$·(AlMe$_3$).
FIG. S23. ssnmr spectra of \((\text{alome})_{8,c} \cdot (\text{alme})_{2}\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S24. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{8,c} \cdot (\text{AlMe}_3)_2\).
FIG. S25. SSNMR spectra of \((\text{AlOMe})_{8,c} \cdot (\text{AlMe}_3)_3\) simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

\[
\begin{array}{c}
600 \text{ MHz (14.1 T)} & 700 \text{ MHz (16.4 T)} \\
(\text{AlOMe})_{8,c} \cdot (\text{AlMe}_3)_3 \\
3 \text{ Coordinate} & 4 \text{ Coordinate} \\
h-h-h & s-h-h \\
s-s-h &
\end{array}
\]

\[
\begin{array}{c}
800 \text{ MHz (18.8 T)} & 830 \text{ MHz (19.6 T)} \\
(\text{AlOMe})_{8,c} \cdot (\text{AlMe}_3)_3 \\
3 \text{ Coordinate} & 4 \text{ Coordinate} \\
h-h-h & s-h-h \\
s-s-h &
\end{array}
\]

\[
\begin{array}{c}
900 \text{ MHz (21.1 T)} & 1000 \text{ MHz (23.5 T)} \\
(\text{AlOMe})_{8,c} \cdot (\text{AlMe}_3)_3 \\
3 \text{ Coordinate} & 4 \text{ Coordinate} \\
h-h-h & s-h-h \\
s-s-h &
\end{array}
\]

FIG. S26. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{8,c} \cdot (\text{AlMe}_3)_3\).
FIG. S27. SSNMR spectra of (AlOMe)$_{8,c}$·(AlMe$_3$)$_4$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S28. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{8,c}$·(AlMe$_3$)$_4$. 

\begin{align*}
C_Q &= 17.5 \text{ MHz} \\
\eta &= 0.59 \\
\delta &= 116 \text{ ppm} \\

C_Q &= 14.7 \text{ MHz} \\
\eta &= 0.27 \\
\delta &= 87 \text{ ppm} \\

C_Q &= 14.6 \text{ MHz} \\
\eta &= 0.24 \\
\delta &= 87 \text{ ppm} \\

C_Q &= 17.4 \text{ MHz} \\
\eta &= 0.59 \\
\delta &= 115 \text{ ppm} \\

C_Q &= 25.9 \text{ MHz} \\
\eta &= 0.99 \\
\delta &= 159 \text{ ppm} \\

C_Q &= 26.2 \text{ MHz} \\
\eta &= 0.97 \\
\delta &= 159 \text{ ppm} \\

C_Q &= 26.0 \text{ MHz} \\
\eta &= 0.99 \\
\delta &= 159 \text{ ppm} \\
\end{align*}
FIG. S29. SSNMR spectra of \((\text{AlOMe})_{10,c}\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S30. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{10,c}\).
FIG. S31. SSNMR spectra of $(\text{AlOMe})_{10,c} \cdot (\text{AlMe}_3)$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S32. Calculated EFG parameters and NMR chemical shifts of $(\text{AlOMe})_{10,c} \cdot (\text{AlMe}_3)$. 
FIG. S33. SSNMR spectra of (AlOMe)$_{10,c}$·(AlMe$_3$)$_2$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S34. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{10,c}$·(AlMe$_3$)$_2$. 
FIG. S35. SSNMR spectra of $(\text{AlOMe})_{10,c}$·$(\text{AlMe}_3)_3$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S36. Calculated EFG parameters and NMR chemical shifts of $(\text{AlOMe})_{10,c}$·$(\text{AlMe}_3)_3$. 

- $C_Q = 9.1$ MHz, $\eta = 0.85$, $\delta = 92$ ppm
- $C_Q = 8.4$ MHz, $\eta = 0.79$, $\delta = 92$ ppm
- $C_Q = 7.9$ MHz, $\eta = 0.89$, $\delta = 92$ ppm
- $C_Q = 35.5$ MHz, $\eta = 0.67$, $\delta = 168$ ppm
- $C_Q = 18.1$ MHz, $\eta = 0.35$, $\delta = 109$ ppm
- $C_Q = 17.9$ MHz, $\eta = 0.35$, $\delta = 109$ ppm
- $C_Q = 18.2$ MHz, $\eta = 0.33$, $\delta = 109$ ppm
- $C_Q = 18.4$ MHz, $\eta = 0.93$, $\delta = 149$ ppm
- $C_Q = 18.6$ MHz, $\eta = 0.93$, $\delta = 149$ ppm
- $C_Q = 18.8$ MHz, $\eta = 0.91$, $\delta = 149$ ppm
- $C_Q = 34.5$ MHz, $\eta = 0.71$, $\delta = 168$ ppm
- $C_Q = 33.7$ MHz, $\eta = 0.74$, $\delta = 168$ ppm
FIG. S37. SSNMR spectra of (AlOMe)$_{12,c}$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S38. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{12,c}$. 

\[ C_Q = 18.0 \text{ MHz} \]
\[ \eta = 0.70 \]
\[ \delta = 98 \text{ ppm} \]
FIG. S39. SSNMR spectra of \((\text{AlOMe})_{14,c}\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S40. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{14,c}\).
FIG. S41. SSNMR spectra of (AlOMe)$_{14,c}$·(AlMe$_3$), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S42. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{14,c}$·(AlMe$_3$).
FIG. S43. SSNMR spectra of (AlOMe)$_{16,c}$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S44. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{16,c}$. 

\[
\begin{align*}
\text{C}_Q &= 18.1 \text{ MHz} \quad \eta = 0.45 \quad \delta = 99 \text{ ppm} \\
\text{C}_Q &= 15.6 \text{ MHz} \quad \eta = 0.00 \quad \delta = 87 \text{ ppm}
\end{align*}
\]
FIG. S45. SSNMR spectra of \((\text{AlOMe})_{18,c}\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S46. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{18,c}\).
FIG. S47. SSNMR spectra of \((\text{AlOMe})_{20,c}\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S48. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{20,c}\).
FIG. S49. SSNMR spectra of (AlOMe)$_{30,c}$·(AlMe$_3$), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).
FIG. S50. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{20}$·(AlMe$_3$).
FIG. S51. SSNMR spectra of \((\text{AlOMe})_{20,c} \cdot (\text{AlMe}_3)_2\) simulated at different field strengths (the corresponding proton resonance frequencies are also provided).
FIG. S52. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_2$•••(AlMe)$_3$. 

$C_Q = 18.1$ MHz  
$\eta = 0.99$  
$\delta = 147$ ppm

$C_Q = 34.8$ MHz  
$\eta = 0.72$  
$\delta = 180$ ppm

$C_Q = 8.9$ MHz  
$\eta = 0.85$  
$\delta = 77$ ppm

$C_Q = 12.8$ MHz  
$\eta = 0.35$  
$\delta = 83$ ppm

$C_Q = 13.8$ MHz  
$\eta = 0.26$  
$\delta = 79$ ppm

$C_Q = 17.4$ MHz  
$\eta = 0.29$  
$\delta = 91$ ppm

$C_Q = 13.8$ MHz  
$\eta = 0.25$  
$\delta = 83$ ppm

$C_Q = 14.1$ MHz  
$\eta = 0.10$  
$\delta = 91$ ppm

$C_Q = 18.0$ MHz  
$\eta = 0.79$  
$\delta = 76$ ppm

$C_Q = 12.9$ MHz  
$\eta = 0.22$  
$\delta = 83$ ppm

$C_Q = 17.2$ MHz  
$\eta = 0.50$  
$\delta = 91$ ppm

$C_Q = 13.8$ MHz  
$\eta = 0.74$  
$\delta = 147$ ppm

$C_Q = 18.0$ MHz  
$\eta = 0.90$  
$\delta = 100$ ppm

$C_Q = 12.9$ MHz  
$\eta = 0.11$  
$\delta = 88$ ppm

$C_Q = 17.2$ MHz  
$\eta = 0.26$  
$\delta = 91$ ppm

$C_Q = 13.9$ MHz  
$\eta = 0.25$  
$\delta = 89$ ppm

$C_Q = 17.8$ MHz  
$\eta = 0.72$  
$\delta = 91$ ppm

$C_Q = 18.0$ MHz  
$\eta = 0.60$  
$\delta = 105$ ppm

$C_Q = 12.9$ MHz  
$\eta = 0.47$  
$\delta = 87$ ppm

$C_Q = 17.8$ MHz  
$\eta = 0.34$  
$\delta = 83$ ppm

$C_Q = 12.8$ MHz  
$\eta = 0.20$  
$\delta = 83$ ppm

$C_Q = 34.2$ MHz  
$\eta = 0.73$  
$\delta = 187$ ppm

$C_Q = 18.0$ MHz  
$\eta = 0.59$  
$\delta = 100$ ppm

$C_Q = 18.0$ MHz  
$\eta = 0.59$  
$\delta = 100$ ppm

$C_Q = 17.8$ MHz  
$\eta = 0.51$  
$\delta = 104$ ppm

$C_Q = 17.8$ MHz  
$\eta = 0.51$  
$\delta = 104$ ppm
FIG. S53. SSNMR spectra of \((\text{AlOMe})_{10,\text{t}}\) simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S54. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{10,\text{t}}\).
FIG. S55. SSNMR spectra of \((\text{AlOMe})_{10,t} \cdot (\text{AlMe}_3)\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S56. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{10,t} \cdot (\text{AlMe}_3)\).
FIG. S57. SSNMR spectra of $(\text{AlOMe})_{10,t} \cdot (\text{AlMe}_3)_2$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S58. Calculated EFG parameters and NMR chemical shifts of $(\text{AlOMe})_{10,t} \cdot (\text{AlMe}_3)_2$. 
FIG. S59. SSNMR spectra of \((\text{AlOMe})_{10,t} \cdot (\text{AlMe}_3)_3\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S60. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{10,t} \cdot (\text{AlMe}_3)_3\).
FIG. S61. SSNMR spectra of \((\text{AlOMe})_{10}, \cdot (\text{AlMe}_3)_4\)·, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S62. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{10}, \cdot (\text{AlMe}_3)_4\)·.
FIG. S63. SSNMR spectra of (AlOMe)$_{12,t}$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S64. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{12,t}$. 

<table>
<thead>
<tr>
<th>Field Strength</th>
<th>$C_Q$ (MHz)</th>
<th>$\eta$</th>
<th>$\delta$ (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>600 MHz (14.1 T)</td>
<td>21.4 MHz</td>
<td>0.66</td>
<td>117 ppm</td>
</tr>
<tr>
<td>700 MHz (16.4 T)</td>
<td>18.4 MHz</td>
<td>0.64</td>
<td>106 ppm</td>
</tr>
<tr>
<td>800 MHz (18.8 T)</td>
<td>14.4 MHz</td>
<td>0.39</td>
<td>92 ppm</td>
</tr>
<tr>
<td>830 MHz (19.6 T)</td>
<td>21.4 MHz</td>
<td>0.66</td>
<td>117 ppm</td>
</tr>
<tr>
<td>900 MHz (21.1 T)</td>
<td>18.4 MHz</td>
<td>0.64</td>
<td>106 ppm</td>
</tr>
<tr>
<td>1000 MHz (23.5 T)</td>
<td>14.4 MHz</td>
<td>0.39</td>
<td>92 ppm</td>
</tr>
</tbody>
</table>
FIG. S65. SSNMR spectra of \((\text{AlOMe})_{12,t} \cdot (\text{AlMe}_3)\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S66. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{12,t} \cdot (\text{AlMe}_3)\).
FIG. S67. SSNMR spectra of (AlOMe)$_{12}$,·(AlMe)$_3$$_2$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S68. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{12}$,·(AlMe)$_3$$_2$. 

CQ  = 9.6 MHz 
η = 0.73 
δ = 79 ppm

CQ  = 14.8 MHz 
η = 0.27 
δ = 91 ppm

CQ  = 18.2 MHz 
η = 0.72 
δ = 106 ppm

CQ  = 14.6 MHz 
η = 0.44 
δ = 89 ppm

CQ  = 20.0 MHz 
η = 0.80 
δ = 144 ppm

CQ  = 15.4 MHz 
η = 0.27 
δ = 91 ppm

CQ  = 19.1 MHz 
η = 0.28 
δ = 106 ppm

CQ  = 33.5 MHz 
η = 0.82 
δ = 175 ppm

CQ  = 8.5 MHz 
η = 0.76 
δ = 79 ppm

CQ  = 14.1 MHz 
η = 0.56 
δ = 89 ppm

CQ  = 22.1 MHz 
η = 0.86 
δ = 140 ppm

CQ  = 19.1 MHz 
η = 0.62 
δ = 104 ppm

CQ  = 33.2 MHz 
η = 0.83 
δ = 175 ppm
FIG. S69. SSNMR spectra of (AlOMe)$_{12,t}$ ⋅ (AlMe$_3$)$_3$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S70. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{12,t}$ ⋅ (AlMe$_3$)$_3$. 
FIG. S71. SSNMR spectra of \((\text{AlOMe})_{12}, (\text{AlMe}_3)_4\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

\[
\begin{align*}
\delta (\text{ppm}) & \quad \delta (\text{ppm}) & \quad \delta (\text{ppm}) \\
600 \text{ MHz} (14.1 \text{ T}) & \quad 700 \text{ MHz} (16.4 \text{ T}) & \quad 800 \text{ MHz} (18.8 \text{ T}) \\
3 \text{ Coordinate} & \quad 4 \text{ Coordinate} & \quad h-h-o \\
\end{align*}
\]

FIG. S72. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{12}, (\text{AlMe}_3)_4\).
FIG. S73. SSNMR spectra of $\text{(AlOMe)}_{14,t}$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

<table>
<thead>
<tr>
<th>Field Strength</th>
<th>Frequency (MHz)</th>
<th>Field (T)</th>
</tr>
</thead>
<tbody>
<tr>
<td>600 MHz</td>
<td>14.1</td>
<td></td>
</tr>
<tr>
<td>700 MHz</td>
<td>16.4</td>
<td></td>
</tr>
<tr>
<td>800 MHz</td>
<td>18.8</td>
<td></td>
</tr>
<tr>
<td>830 MHz</td>
<td>19.6</td>
<td></td>
</tr>
<tr>
<td>900 MHz</td>
<td>21.1</td>
<td></td>
</tr>
<tr>
<td>1000 MHz</td>
<td>23.5</td>
<td></td>
</tr>
</tbody>
</table>

![SSNMR Spectra](image)

FIG. S74. Calculated EFG parameters and NMR chemical shifts of $\text{(AlOMe)}_{14,t}$.

- $C_Q = 21.4$ MHz, $\eta = 0.66$, $\delta = 117$ ppm
- $C_Q = 18.5$ MHz, $\eta = 0.66$, $\delta = 105$ ppm
- $C_Q = 13.5$ MHz, $\eta = 0.20$, $\delta = 91$ ppm
- $C_Q = 15.2$ MHz, $\eta = 0.37$, $\delta = 91$ ppm
FIG. S75. SSNMR spectra of (AlOMe)$_{14,t}$・(AlMe$_3$), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S76. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{14,t}$・(AlMe$_3$).
FIG. S77. SSNMR spectra of (AlOMe)$_{14,t}$·(AlMe$_3$)$_2$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S78. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{14,t}$·(AlMe$_3$)$_2$. 
FIG. S79. SSNMR spectra of $(\text{AlOMe})_{14,t} \cdot \text{(AlMe}_3)_3$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S80. Calculated EFG parameters and NMR chemical shifts of $(\text{AlOMe})_{14,t} \cdot \text{(AlMe}_3)_3$. 

- $C_Q = 19.8 \text{ MHz}$, $\eta = 0.82$, $\delta = 144 \text{ ppm}$
- $C_Q = 8.9 \text{ MHz}$, $\eta = 0.82$, $\delta = 80 \text{ ppm}$
- $C_Q = 18.9 \text{ MHz}$, $\eta = 0.28$, $\delta = 105 \text{ ppm}$
- $C_Q = 18.5 \text{ MHz}$, $\eta = 0.71$, $\delta = 89 \text{ ppm}$
- $C_Q = 33.3 \text{ MHz}$, $\eta = 0.83$, $\delta = 175 \text{ ppm}$
- $C_Q = 15.7 \text{ MHz}$, $\eta = 0.46$, $\delta = 89 \text{ ppm}$
- $C_Q = 13.4 \text{ MHz}$, $\eta = 0.26$, $\delta = 86 \text{ ppm}$
- $C_Q = 14.3 \text{ MHz}$, $\eta = 0.22$, $\delta = 87 \text{ ppm}$
- $C_Q = 16.9 \text{ MHz}$, $\eta = 0.74$, $\delta = 115 \text{ ppm}$
FIG. S81. SSNMR spectra of (AlOMe)₄₄ · (AlMe₃)₄, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S82. Calculated EFG parameters and NMR chemical shifts of (AlOMe)₄₄ · (AlMe₃)₄.
FIG. S83. SSNMR spectra of (AlOMe)$_{16,t}$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S84. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{16,t}$. 

\[
\begin{align*}
\text{C}_Q &= 21.2 \, \text{MHz} \\
\eta &= 0.68 \\
\delta &= 118 \, \text{ppm} \\
\eta &= 0.68 \\
\delta &= 106 \, \text{ppm} \\
\eta &= 0.20 \\
\delta &= 90 \, \text{ppm} \\
\eta &= 0.36 \\
\delta &= 91 \, \text{ppm}
\end{align*}
\]
FIG. S85. SSNMR spectra of \((\text{AlOMe})_{16,n} \cdot \text{(AlMe}_3\)\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S86. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{16,t}\)\).
FIG. S87. SSNMR spectra of (AlOMe)\textsubscript{16,t} \cdot (AlMe\textsubscript{3})\textsubscript{2}, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S88. Calculated EFG parameters and NMR chemical shifts of (AlOMe)\textsubscript{16,t} \cdot (AlMe\textsubscript{3})\textsubscript{2}.
FIG. S89. SSNMR spectra of \((\text{AlOMe})_{16,n} \cdot (\text{AlMe}_3)_3\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S90. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{16,t} \cdot (\text{AlMe}_3)_3\).
FIG. S91. SSNMR spectra of (AlOMe)$_{16,n} \cdot$ (AlMe$_3$)$_4$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S92. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{16,n} \cdot$ (AlMe$_3$)$_4$. 
FIG. S93. SSNMR spectra of (AlOMe)$_{18,t}$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S94. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{18,t}$. 
FIG. S95. SSNMR spectra of (AlOMe)\textsubscript{18,t} \cdot (AlMe\textsubscript{3}), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S96. Calculated EFG parameters and NMR chemical shifts of (AlOMe)\textsubscript{18,t} \cdot (AlMe\textsubscript{3}).
FIG. S97. SSNMR spectra of \((\text{AlOMe})_{18,t} \cdot (\text{AlMe}_3)_2\) simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S98. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{18,t} \cdot (\text{AlMe}_3)_2\).
FIG. S99. SSNMR spectra of (AlOMe)$_{18,t}$·(AlMe)$_3^3$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S100. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{18,t}$·(AlMe)$_3^3$. 

<table>
<thead>
<tr>
<th>Field Strength (MHz, T)</th>
<th>CQ (MHz)</th>
<th>η</th>
<th>δ (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>600 MHz (14.1 T)</td>
<td>17.1 MHz</td>
<td>0.74</td>
<td>88 ppm</td>
</tr>
<tr>
<td>700 MHz (16.4 T)</td>
<td>16.3 MHz</td>
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<td>92 ppm</td>
</tr>
<tr>
<td>800 MHz (18.8 T)</td>
<td>15.0 MHz</td>
<td>0.32</td>
<td>92 ppm</td>
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<tr>
<td>830 MHz (19.6 T)</td>
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<tr>
<td>900 MHz (21.1 T)</td>
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<td>83 ppm</td>
</tr>
<tr>
<td>1000 MHz (23.5 T)</td>
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<td>83 ppm</td>
</tr>
<tr>
<td></td>
<td>14.5 MHz</td>
<td>0.27</td>
<td>83 ppm</td>
</tr>
<tr>
<td></td>
<td>14.9 MHz</td>
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</tr>
<tr>
<td></td>
<td>15.0 MHz</td>
<td>0.23</td>
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<tr>
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<td>15.5 MHz</td>
<td>0.18</td>
<td>88 ppm</td>
</tr>
<tr>
<td></td>
<td>16.5 MHz</td>
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<tr>
<td></td>
<td>16.8 MHz</td>
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<td></td>
<td>17.1 MHz</td>
<td>0.32</td>
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<td>18.9 MHz</td>
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<td>20.4 MHz</td>
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<td></td>
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<td>92 ppm</td>
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<tr>
<td></td>
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<td>31.2 MHz</td>
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<td>31.5 MHz</td>
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<td></td>
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<td>0.28</td>
<td>92 ppm</td>
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<tr>
<td></td>
<td>32.1 MHz</td>
<td>0.32</td>
<td>92 ppm</td>
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<td>32.4 MHz</td>
<td>0.28</td>
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<td></td>
<td>32.7 MHz</td>
<td>0.32</td>
<td>92 ppm</td>
</tr>
<tr>
<td></td>
<td>33.0 MHz</td>
<td>0.28</td>
<td>92 ppm</td>
</tr>
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</table>

CQ = 17.1 MHz
η = 0.74
δ = 116 ppm
CQ = 16.3 MHz
η = 0.23
δ = 88 ppm
CQ = 15.0 MHz
η = 0.32
δ = 88 ppm
CQ = 14.8 MHz
η = 0.22
δ = 89 ppm
CQ = 14.5 MHz
η = 0.17
δ = 83 ppm
CQ = 14.2 MHz
η = 0.27
δ = 83 ppm
CQ = 14.0 MHz
η = 0.18
δ = 83 ppm
CQ = 13.6 MHz
η = 0.12
δ = 83 ppm
CQ = 13.2 MHz
η = 0.10
δ = 83 ppm
CQ = 12.8 MHz
η = 0.08
δ = 83 ppm
CQ = 12.4 MHz
η = 0.06
δ = 83 ppm
CQ = 12.0 MHz
η = 0.04
δ = 83 ppm
CQ = 11.6 MHz
η = 0.02
δ = 83 ppm
CQ = 11.2 MHz
η = 0.00
δ = 83 ppm
CQ = 10.8 MHz
η = 0.00
δ = 83 ppm
CQ = 10.4 MHz
η = 0.00
δ = 83 ppm
CQ = 10.0 MHz
η = 0.00
δ = 83 ppm
CQ = 9.6 MHz
η = 0.00
δ = 83 ppm
CQ = 9.2 MHz
η = 0.00
δ = 83 ppm
CQ = 8.8 MHz
η = 0.00
δ = 83 ppm
CQ = 8.4 MHz
η = 0.00
δ = 83 ppm
CQ = 8.0 MHz
η = 0.00
δ = 83 ppm
CQ = 7.6 MHz
η = 0.00
δ = 83 ppm
CQ = 7.2 MHz
η = 0.00
δ = 83 ppm
CQ = 6.8 MHz
η = 0.00
δ = 83 ppm
CQ = 6.4 MHz
η = 0.00
δ = 83 ppm
CQ = 6.0 MHz
η = 0.00
δ = 83 ppm
CQ = 5.6 MHz
η = 0.00
δ = 83 ppm
CQ = 5.2 MHz
η = 0.00
δ = 83 ppm
CQ = 4.8 MHz
η = 0.00
δ = 83 ppm
CQ = 4.4 MHz
η = 0.00
δ = 83 ppm
CQ = 4.0 MHz
η = 0.00
δ = 83 ppm
CQ = 3.6 MHz
η = 0.00
δ = 83 ppm
CQ = 3.2 MHz
η = 0.00
δ = 83 ppm
CQ = 2.8 MHz
η = 0.00
δ = 83 ppm
CQ = 2.4 MHz
η = 0.00
δ = 83 ppm
CQ = 2.0 MHz
η = 0.00
δ = 83 ppm
CQ = 1.6 MHz
η = 0.00
δ = 83 ppm
CQ = 1.2 MHz
η = 0.00
δ = 83 ppm
CQ = 0.8 MHz
η = 0.00
δ = 83 ppm
CQ = 0.4 MHz
η = 0.00
δ = 83 ppm
CQ = 0.0 MHz
η = 0.00
δ = 83 ppm
FIG. S101. SSNMR spectra of \((\text{AlOMe})_{18,n} \cdot (\text{AlMe}_3)_4\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

<table>
<thead>
<tr>
<th>Field Strength</th>
<th>Proton Resonance Frequencies</th>
</tr>
</thead>
<tbody>
<tr>
<td>600 MHz (14.1 T)</td>
<td>3 Coordinate: 700 MHz (16.4 T)</td>
</tr>
<tr>
<td>700 MHz (16.4 T)</td>
<td>4 Coordinate: 800 MHz (18.8 T)</td>
</tr>
<tr>
<td>800 MHz (18.8 T)</td>
<td>3 Coordinate: 830 MHz (19.6 T)</td>
</tr>
<tr>
<td>900 MHz (21.1 T)</td>
<td>4 Coordinate: 1000 MHz (23.5 T)</td>
</tr>
</tbody>
</table>

FIG. S102. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{18,n} \cdot (\text{AlMe}_3)_4\).
FIG. S103. SSNMR spectra of (AlOMe)$_{20,\text{t}}$ simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S104. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{20,\text{t}}$. 
FIG. S105. SSNMR spectra of $\text{(AlOMe)}_2\cdot\text{(AlMe}_3\text{)}$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S106. Calculated EFG parameters and NMR chemical shifts of $\text{(AlOMe)}_2\cdot\text{(AlMe}_3\text{)}$. 

```
C Q = 19.7 MHz
n = 0.84
δ = 145 ppm

C Q = 8.7 MHz
n = 0.85
δ = 78 ppm

C Q = 32.7 MHz
n = 0.84
δ = 173 ppm
```

```
C Q = 13.2 MHz
n = 0.19
δ = 84 ppm

C Q = 18.5 MHz
n = 0.74
δ = 106 ppm

C Q = 15.1 MHz
n = 0.21
δ = 78 ppm
```

```
C Q = 14.1 MHz
n = 0.18
δ = 86 ppm

C Q = 14.9 MHz
n = 0.25
δ = 106 ppm

C Q = 15.3 MHz
n = 0.30
δ = 82 ppm
```

```
C Q = 15.4 MHz
n = 0.33
δ = 91 ppm

C Q = 16.0 MHz
n = 0.41
δ = 173 ppm

C Q = 15.4 MHz
n = 0.33
δ = 90 ppm
```

```
C Q = 16.5 MHz
n = 0.74
δ = 106 ppm

C Q = 16.0 MHz
n = 0.25
δ = 78 ppm

C Q = 15.4 MHz
n = 0.39
δ = 84 ppm
```

```
C Q = 19.1 MHz
n = 0.67
δ = 107 ppm

C Q = 21.2 MHz
n = 0.71
δ = 118 ppm
```

```
C Q = 21.2 MHz
n = 0.71
δ = 118 ppm

C Q = 15.6 MHz
n = 0.24
δ = 92 ppm

C Q = 15.1 MHz
n = 0.30
δ = 86 ppm
```

```
C Q = 15.4 MHz
n = 0.33
δ = 90 ppm

C Q = 14.2 MHz
n = 0.33
δ = 92 ppm

C Q = 15.4 MHz
n = 0.33
δ = 90 ppm
```

```
C Q = 19.1 MHz
n = 0.67
δ = 107 ppm

C Q = 21.2 MHz
n = 0.71
δ = 118 ppm

C Q = 15.6 MHz
n = 0.24
δ = 92 ppm
```

```
C Q = 15.3 MHz
n = 0.30
δ = 86 ppm

C Q = 15.4 MHz
n = 0.39
δ = 84 ppm

C Q = 15.4 MHz
n = 0.39
δ = 84 ppm
```

```
C Q = 15.4 MHz
n = 0.33
δ = 90 ppm

C Q = 15.4 MHz
n = 0.33
δ = 90 ppm

C Q = 15.4 MHz
n = 0.33
δ = 90 ppm
```

```
C Q = 19.1 MHz
n = 0.67
δ = 107 ppm

C Q = 21.2 MHz
n = 0.71
δ = 118 ppm

C Q = 15.6 MHz
n = 0.24
δ = 92 ppm
```
FIG. S107. SSNMR spectra of (AlOMe)$_{20,n}$・(AlMe$_3$)$_2$, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S108. Calculated EFG parameters and NMR chemical shifts of (AlOMe)$_{20,t}$・(AlMe$_3$)$_2$. 
FIG. S109. SSNMR spectra of \((\text{AlOMe})_{20,t} \cdot (\text{AlMe}_3)_3\), simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

![SSNMR spectra](image)

FIG. S110. Calculated EFG parameters and NMR chemical shifts of \((\text{AlOMe})_{20,t} \cdot (\text{AlMe}_3)_3\).
FIG. S111. SSNMR spectra of (AlOMe)₂₀,ₙ・(AlMe₃)₄, simulated at different field strengths (the corresponding proton resonance frequencies are also provided).

FIG. S112. Calculated EFG parameters and NMR chemical shifts of (AlOMe)₂₀,ₙ・(AlMe₃)₄.