

Supporting information for

**A Facile Approach Using  $\text{MgCl}_2$  to Formulating  
High Performance  $\text{Mg}^{2+}$  Electrolytes for Rechargeable Mg Batteries**

*Tianbiao Liu\**, *Yuyan Shao*, *Guosheng Li\**, *Meng Gu*, *Jianzhi Hu*,  
*Suochang Xu*, *Zimin Nie*, *Xilin Chen*, *Chongmin Wang*, *Jun Liu\**

Energy Process and Materials Division

Pacific Northwest National Laboratory (PNNL), *P.O. Box 999, Richland, WA 99352 (USA)*

*E-mail: [Tianbiao.Liu@pnnl.gov](mailto:Tianbiao.Liu@pnnl.gov); [Guosheng.Li@pnnl.gov](mailto:Guosheng.Li@pnnl.gov); [Jun.Liu@pnnl.gov](mailto:Jun.Liu@pnnl.gov)*

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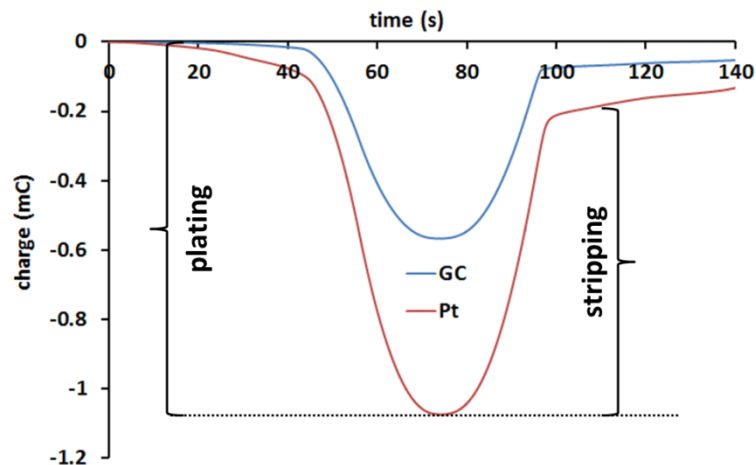


Figure S1. Plots of charge over time of the Mg plating and subsequent stripping processes of the  $\text{MgCl}_2\text{-AlCl}_3$  electrolyte on GC (blue trace, 89% coulombic efficiency) and Pt (red trace, 90% coulombic efficiency) working electrodes. Conditions: scan rate, 50 mV/s; reference electrode, a Mg strip; counter electrode, glassy carbon; 22 °C; under 1.0 atm Ar.

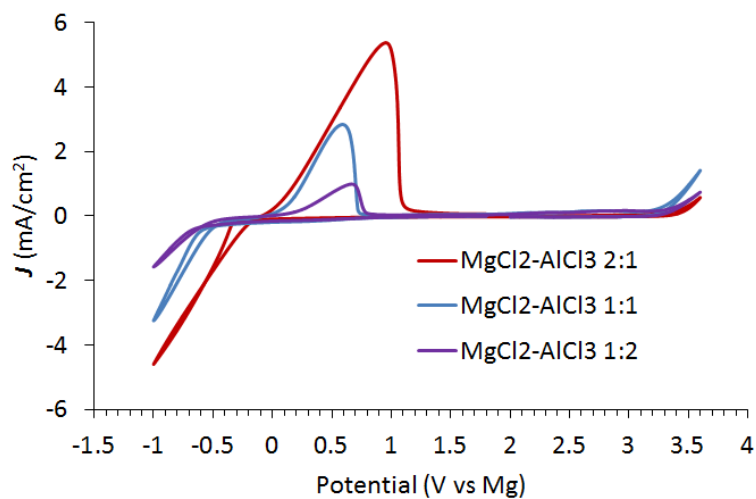


Figure S2. Comparison of CVs of the  $\text{MgCl}_2\text{-AlCl}_3$  electrolyte at different ratios, 2:1 (red trace), 1:1 (blue trace) and 1:2 (purple trace) recorded on a GC electrode. Conditions: scan rate, 50 mV/s; reference electrode, a Mg strip; counter electrode, glassy carbon; 22 °C; under 1.0 atm Ar.

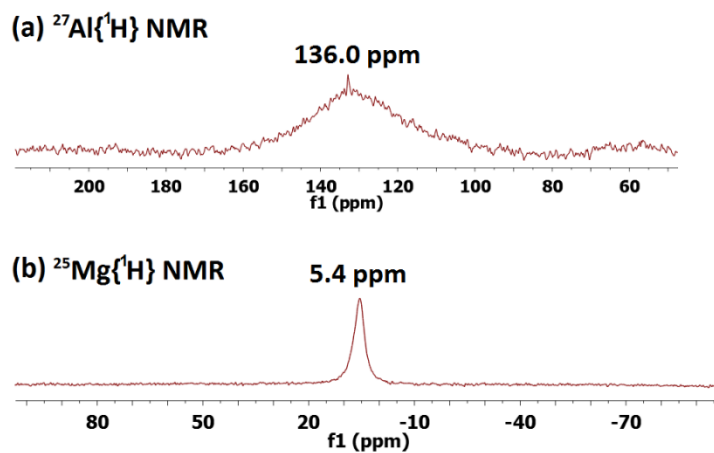


Figure S3.  $^{27}\text{Al}\{^1\text{H}\}$  NMR and  $^{25}\text{Mg}\{^1\text{H}\}$  NMR spectra of the  $\text{MgCl}_2\text{-AlPh}_3$  electrolyte recorded in THF at 22 °C.

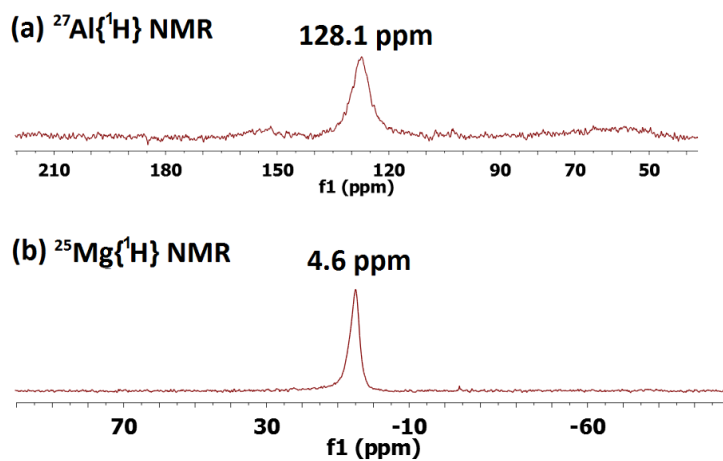


Figure S4.  $^{27}\text{Al}\{^1\text{H}\}$  NMR and  $^{25}\text{Mg}\{^1\text{H}\}$  NMR spectra of the  $\text{MgCl}_2\text{-AlEtCl}_2$  electrolyte recorded in THF at 22 °C.

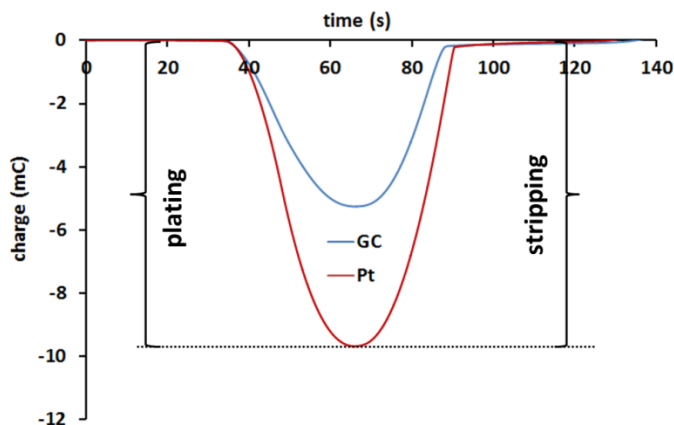


Figure S5. Plots of charge over time of the Mg plating and subsequent stripping processes of the  $\text{MgCl}_2\text{-AlPh}_3$  electrolyte on GC (blue trace, ca. 100% coulombic efficiency) and Pt (red trace, ca. 100% coulombic efficiency) working electrodes. Conditions: scan rate, 50 mV/s; reference electrode, a Mg strip; counter electrode, glassy carbon; 22 °C; under 1.0 atm Ar.

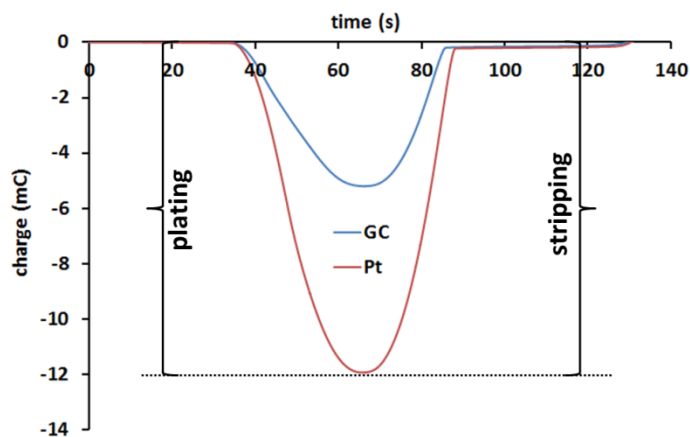


Figure S6. Plots of charge over time of the Mg plating and subsequent stripping processes of the  $\text{MgCl}_2\text{-AlEtCl}_2$  electrolyte on GC (blue trace, ca. 100% coulombic efficiency) and Pt (red trace, ca. 100% coulombic efficiency) working electrodes. Conditions: scan rate, 50 mV/s; reference electrode, a Mg strip; counter electrode, glassy carbon; 22 °C; under 1.0 atm Ar.

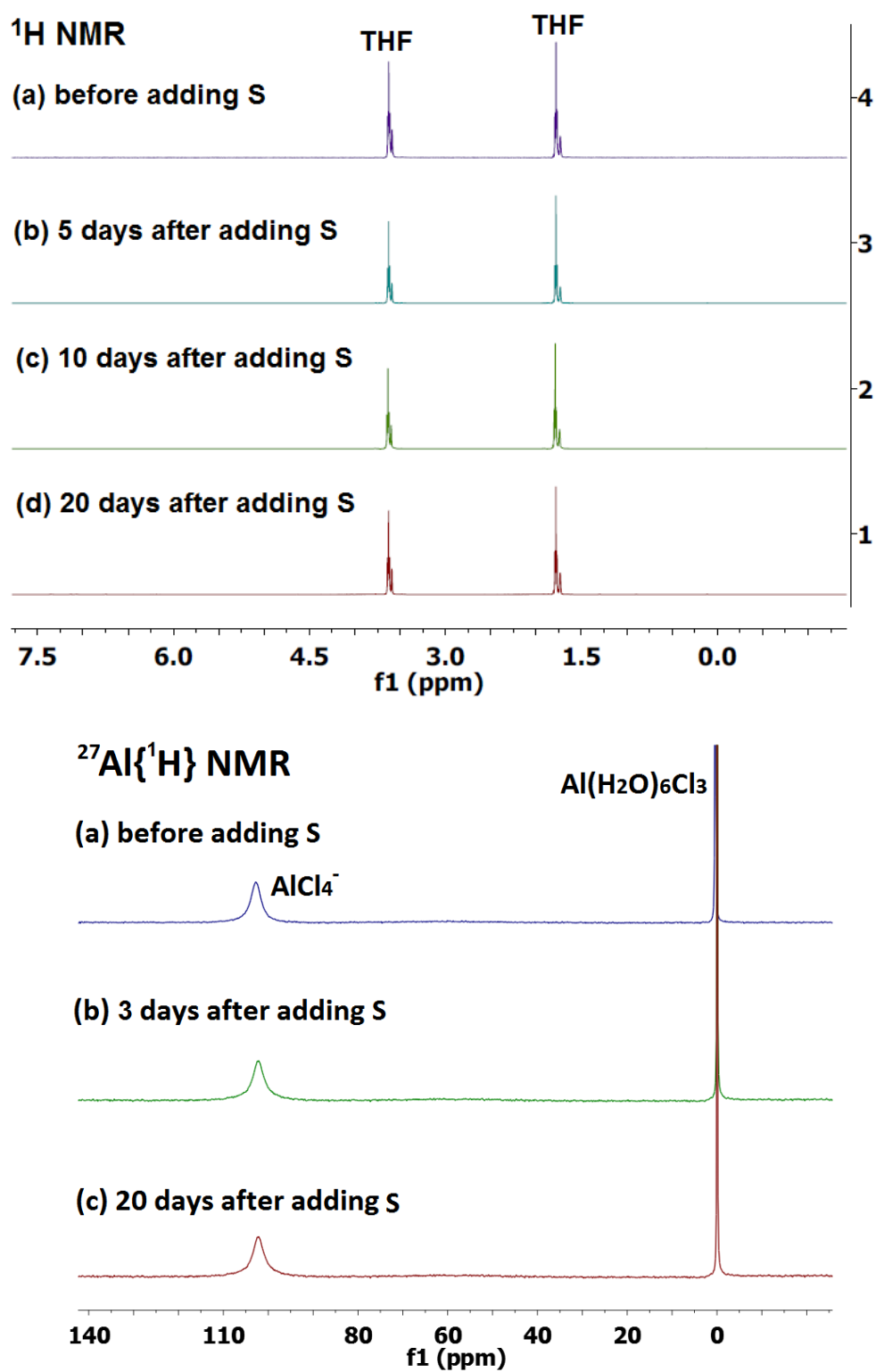


Figure S7. Sulfur compatibility of the  $\text{MgCl}_2\text{-AlCl}_3$  electrolyte followed by  $^1\text{H}$  NMR (top) and  $^{27}\text{Al}\{^1\text{H}\}$  NMR (bottom) spectroscopy in THF at 22 °C. For  $^{27}\text{Al}\{^1\text{H}\}$  NMR, a sealed

capillary containing 40 mM  $\text{Al}(\text{H}_2\text{O})_6\text{Cl}_3$  (0 ppm) as the internal reference for chemical shift and concentration was placed in the J-Young NMR tube.

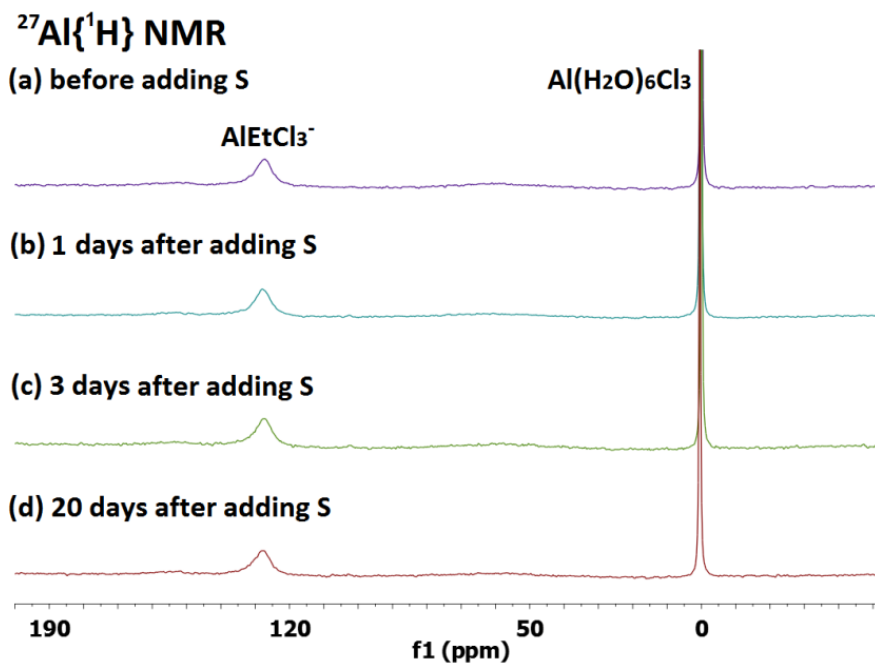
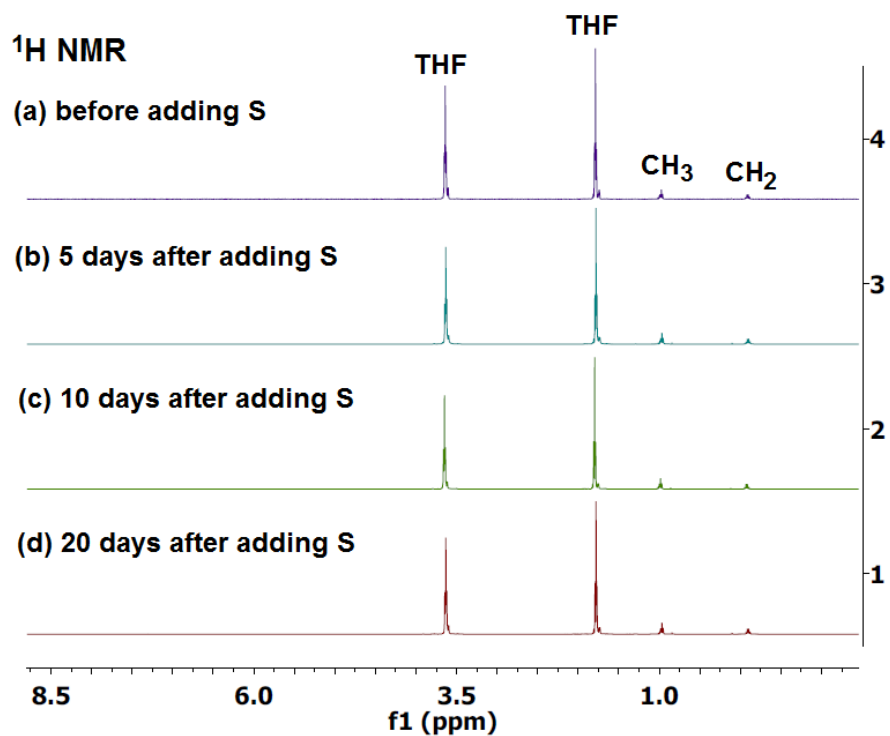
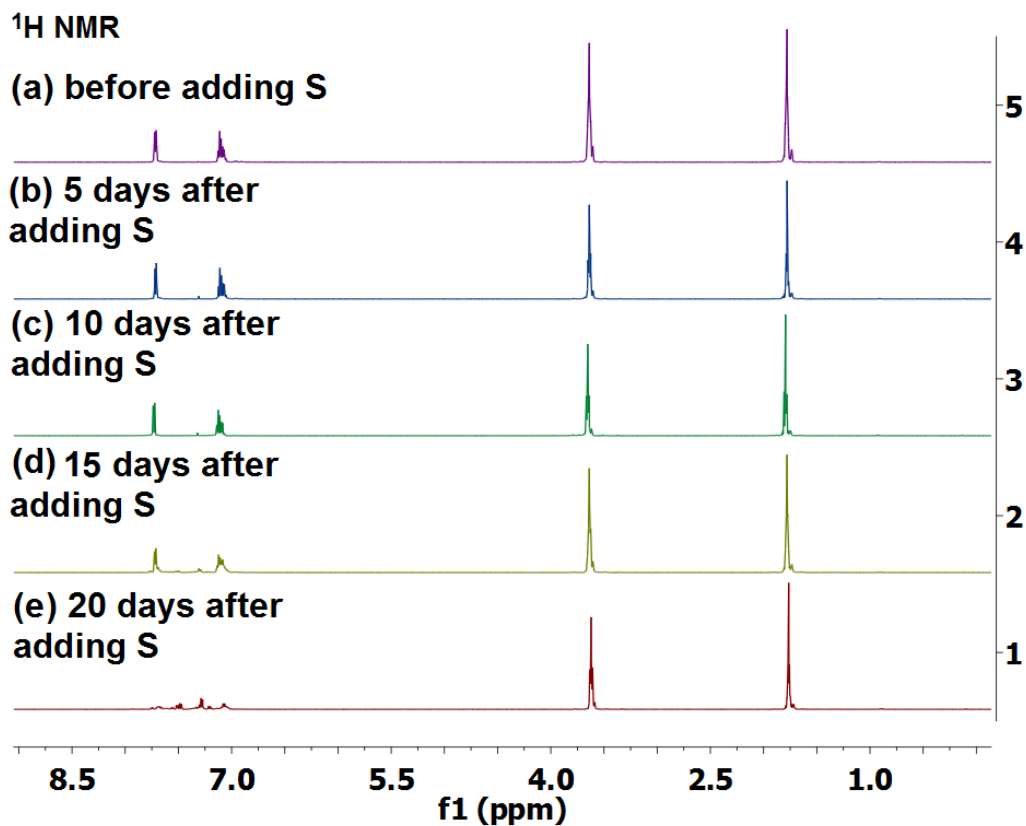
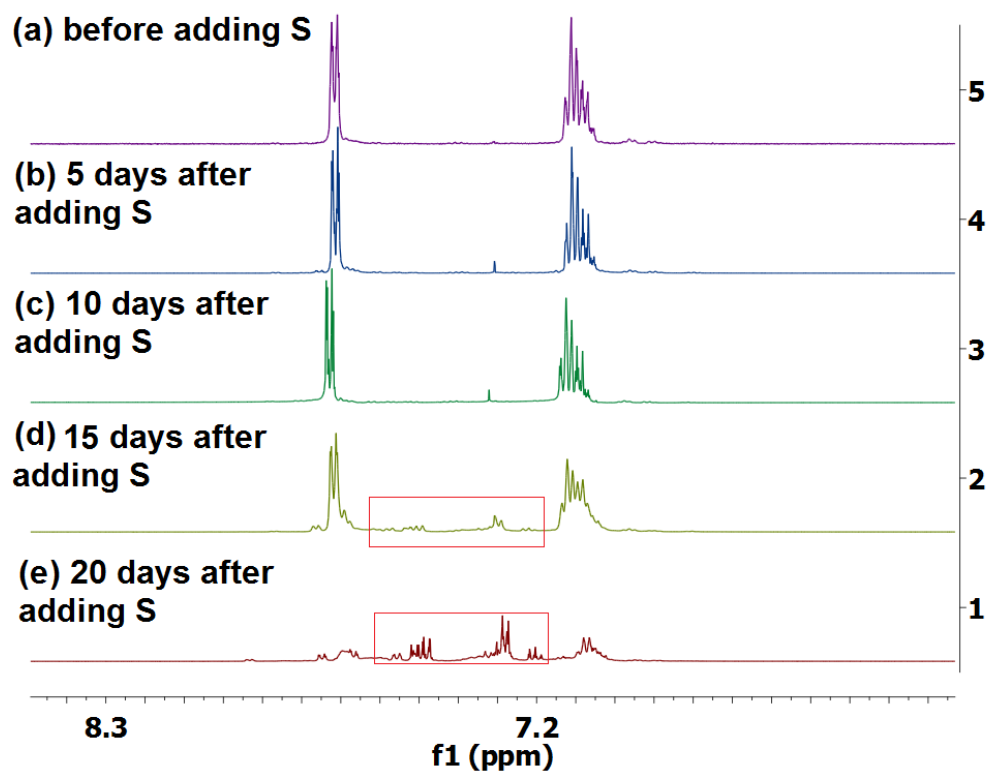


Figure S8. Sulfur compatibility of the  $\text{MgCl}_2\text{-AlEtCl}_2$  electrolyte followed by  $^1\text{H}$  NMR (top) and  $^{27}\text{Al}\{^1\text{H}\}$  NMR (bottom) spectroscopy in THF at 22 °C. For  $^{27}\text{Al}\{^1\text{H}\}$  NMR, a sealed capillary containing 40 mM  $\text{Al}(\text{H}_2\text{O})_6\text{Cl}_3$  (0 ppm) as the internal reference for chemical shift and concentration was placed in the J-Young NMR tube.



### Proton resonances of phenyl region





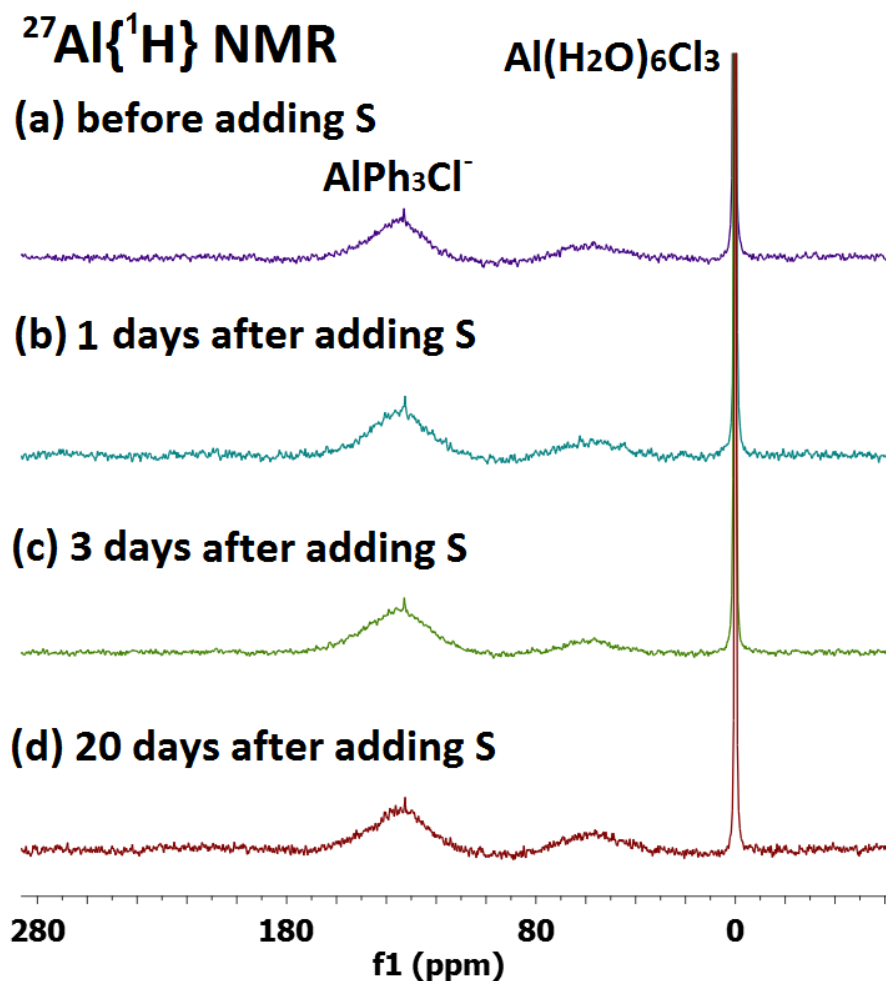


Figure S9. Sulfur compatibility of the  $\text{MgCl}_2\text{-AlPh}_3$  electrolyte followed by  $^1\text{H}$  NMR (top and middle) and  $^{27}\text{Al}\{^1\text{H}\}$  NMR (bottom) spectroscopy in THF at 22 °C. In  $^1\text{H}$  NMR spectra, the new peaks in red rectangles appear in the phenyl region and highlighted indicate degradation. For  $^{27}\text{Al}\{^1\text{H}\}$  NMR, a sealed capillary containing 40 mM  $\text{Al}(\text{H}_2\text{O})_6\text{Cl}_3$  (0 ppm) as the internal reference for chemical shift and concentration was placed in the J-Young NMR tube.

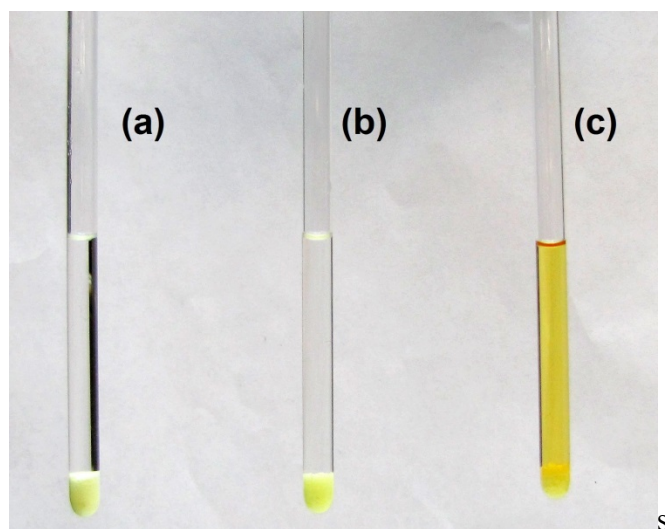


Figure S10. The images of the NMR tubes containing the  $\text{MgCl}_2\text{-AlCl}_3$  electrolyte (a), the  $\text{MgCl}_2\text{-AlEtCl}_2$  electrolyte (b), the  $\text{MgCl}_2\text{-AlPh}_3$  electrolyte (c) after sulfur treatment for 20 days.

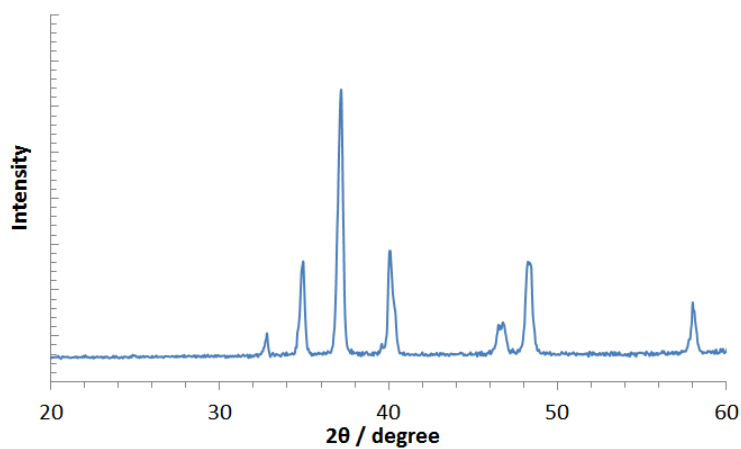


Figure S11. XRD pattern of deposited Mg on a Pt plate using the  $\text{MgCl}_2\text{-AlEtCl}_2$  electrolyte.