Supporting Information

Electrolyte Additive Enabled Low Temperature Lithium Metal Batteries

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Figure S1. The CE of DMSO/DOL electrolyte at RT with a) 2 vol%; b) 10 vol%; c) 20 vol%; and d) 40 vol% of DMSO at 0.5 mA cm⁻² and 1 mA h cm⁻².



Figure S2. Step-wise cycling tests in Li/Li symmetric cells with DOL electrolyte under varying temperatures from 0 °C to -80 °C at 0.25 mA cm⁻² and 0.5 mA h cm⁻². The cell failed at -60 °C.



Figure S3. The morphology of deposited Li metal on Cu substrate at 0.5 mA cm⁻² for 8 hours at RT with a) DOL electrolyte and b) DMSO/DOL electrolyte.



Figure S4. a) EIS plots of DOL and DMSO/DOL electrolytes in SS/SS cells, respectively, at different temperatures at 0 °C, -40 °C, and -80 °C (inset). b) The digital photos of DOL and DMSO/DOL electrolytes after stored at RT and -80 °C overnight. Li salt precipitated in DOL electrolyte at -80 °C.



Figure S5. Raman spectroscopy of pure DMSO, $DOL/LiClO_4$ electrolyte, and $DOL/LiClO_4$ electrolyte with 5 vol% DMSO. The blue dotted lines represent two characteristic peaks of DMSO, while the red dotted lines labelled the shifted peaks in the DMSO added $DOL/LiClO_4$ electrolyte.



Figure S6. The first 20 hours of cycling in Li/Li symmetric cells with DOL electrolyte and DMSO/DOL electrolyte at a) RT; b) -20 °C, at 0.5 mA cm⁻², 1mA h cm⁻². The fluctuation labelled by the blue box represents the stabilization process in DMSO added electrolytes.



Figure S7. EDX results of the SEI formed on Li metal anode after 5 cycles of Li plating and stripping in Li/Li cells at 0.5 mA cm⁻² and 1 mA h cm⁻². a) The percentage of the Cl in the SEI formed in DOL electrolyte from RT to -40 °C and in DMSO/DOL electrolyte from RT to -80 °C. b) The percentage of S in SEI formed in DMSO/DOL electrolyte from RT to -80 °C.



Figure S8. High resolution Li 1s and Cl 2p XPS spectra for the SEI formed on Li metal anode in Li/Li symmetric cells after 5 cycles at 0.5 mA cm⁻² and 1 mA h cm⁻² at RT with DOL electrolyte and DMSO/DOL electrolyte, respectively.



Figure S9. Proposed schemes of the solvation behavior of Li ion and the structure of the SEI in a) pure DOL electrolyte and b) DMSO/DOL electrolyte, respectively.