Effect of catholyte additives on the performance of lithium polysulfide cells

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Background:

- Lithium-sulfur batteries (LSB) are an attractive next generation for lithium-ion batteries thanks to higher energy density, good safety and lower cost.
- LSBs are hampered by poor coulombic efficiency and capacity retention due to long chain lithium polysulfide molecules transporting to the anode and irreversibly forming an insulating layer of Li_2S, in a process known as polysulfide shuttle.
- We are developing a flow battery configuration for Li-S chemistry which minimises this shuttle effect and prevents the formation of Li_2S by limiting the lower voltage of discharge. Sulfur is stored in solution (catholyte) as lithium polysulfides.

Aims:

- The aim of this work is to optimise the catholyte formulation, by using different solvents and electrolyte additives. We are studying the passivation and cycling of the lithium anode and characterising the catholyte with in situ techniques.

Application:

- StorTera are developing the SLIQ single liquid flow battery system which offers a modular and scalable design allowing long duration energy storage up to 12 hours.
- StorTera are currently building a 200 kW/1.6 MWh (8 hour duration) battery system which will be installed in Edinburgh in 2024.
- Benefits:
  - Low cost of storage (<1 p/kWh target)
  - 20-year lifetime
  - Sustainable (>70% reusability)
  - High efficiency over lifetime (>90%)

Figure 1: *StorTera large scale lithium polysulphide test flow cell*

Methodology:

- Catholyte is prepared in an inert argon atmosphere on a Schlenk line

$$2Li_{(s)} + \frac{3}{4}S_{8\,(s)} \xrightarrow[Ar,\,\Delta]{solvent} Li_2S_{6\,(l)}$$

- Batteries are constructed in an inert atmosphere before being cycled at 1 mA in a voltage window of 2.8-2.0 V
- Different concentrations of $LiNO_3$ is added to form a solid electrolyte interphase (SEI) layer

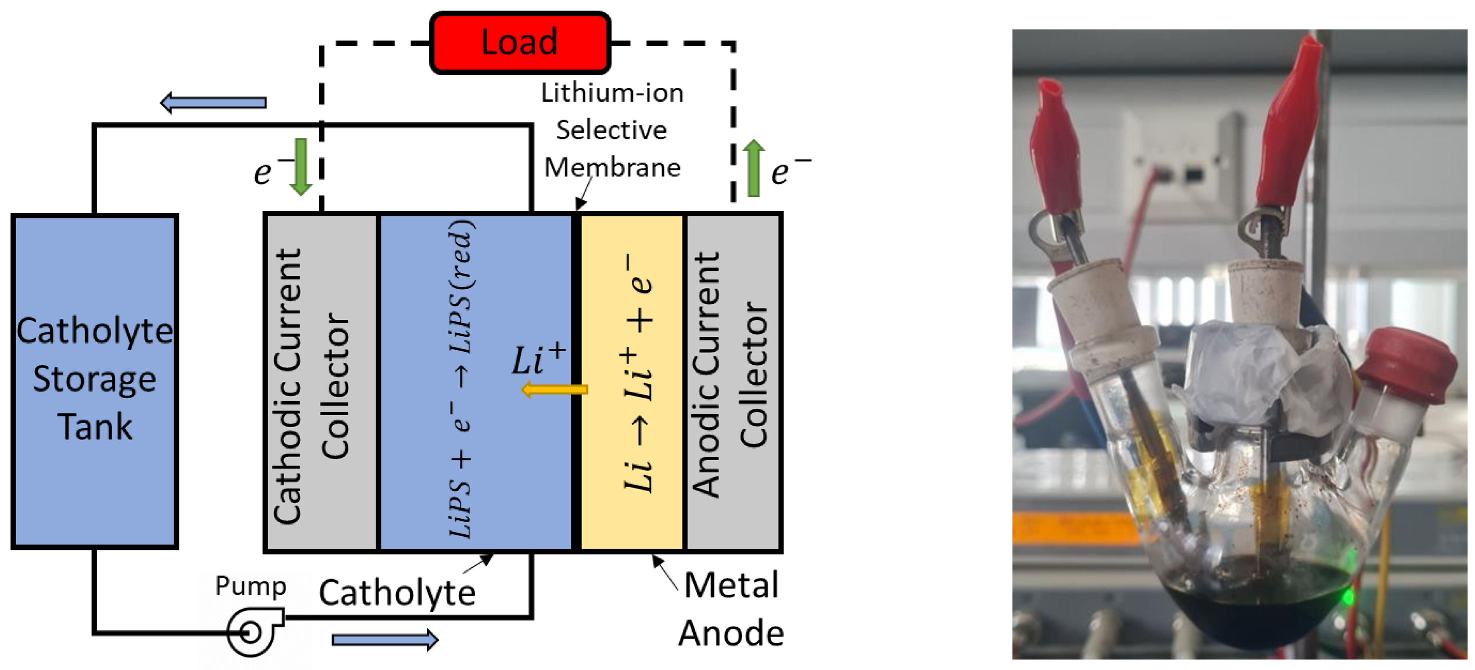

Figure 2: *Schematic of the flow cell (left) and an image of the test cell (right)*

Anode Pre-treatment:

- Pre-treatment of anode with LiTFSI to protect the lithium metal from corrosion
- Benefits of the protection layer formed are increased capacity but only when used sparingly
- Lower capacity of cycles 2-5 suggest that SEI layer has been removed and the anode can be corroded

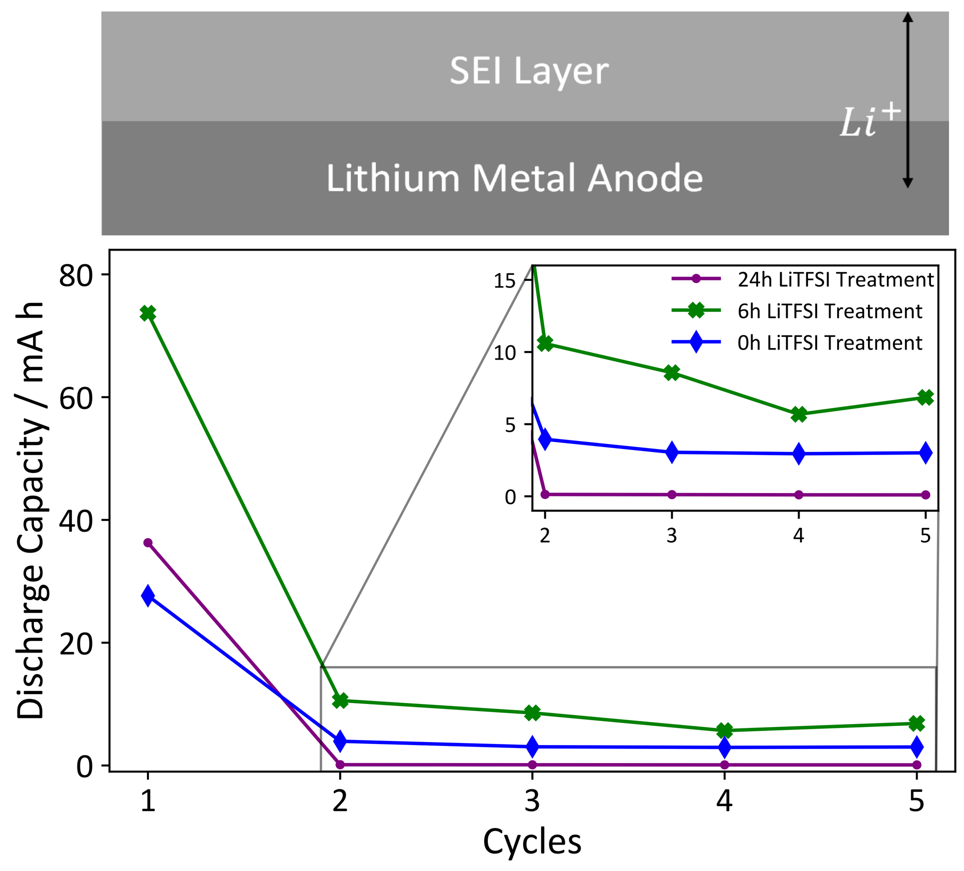

Figure 3: *Discharge capacity of pre-treated cell (bottom) and schematic of SEI layer formed (top)*

Cycling Data:

- DOL-DME cells have higher capacity but addition of DMSO in moderation increases capacity retention
- Higher DMSO content increases capacity in 1st cycle only

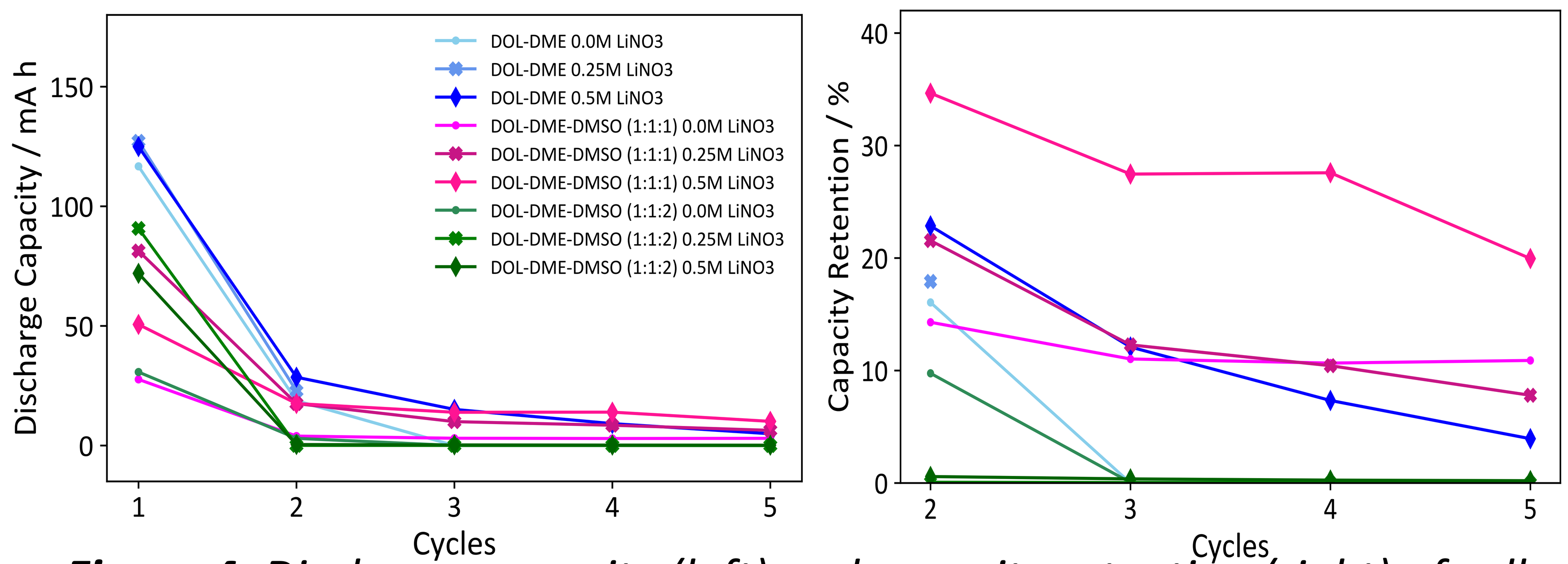

Figure 4: *Discharge capacity (left) and capacity retention (right) of cell*

Conclusions & Further Work:

- DMSO decreases capacity but improves cell stability
- Pre-treatment of anode in moderation increases capacity
- Development of stable cell geometry for *in situ* studies
- SEI layer will be studied through *in situ* electrochemical quartz crystal microbalance (QCM) and infrared (IR) spectroscopy of the lithium anode and catholyte using a modified flow cell

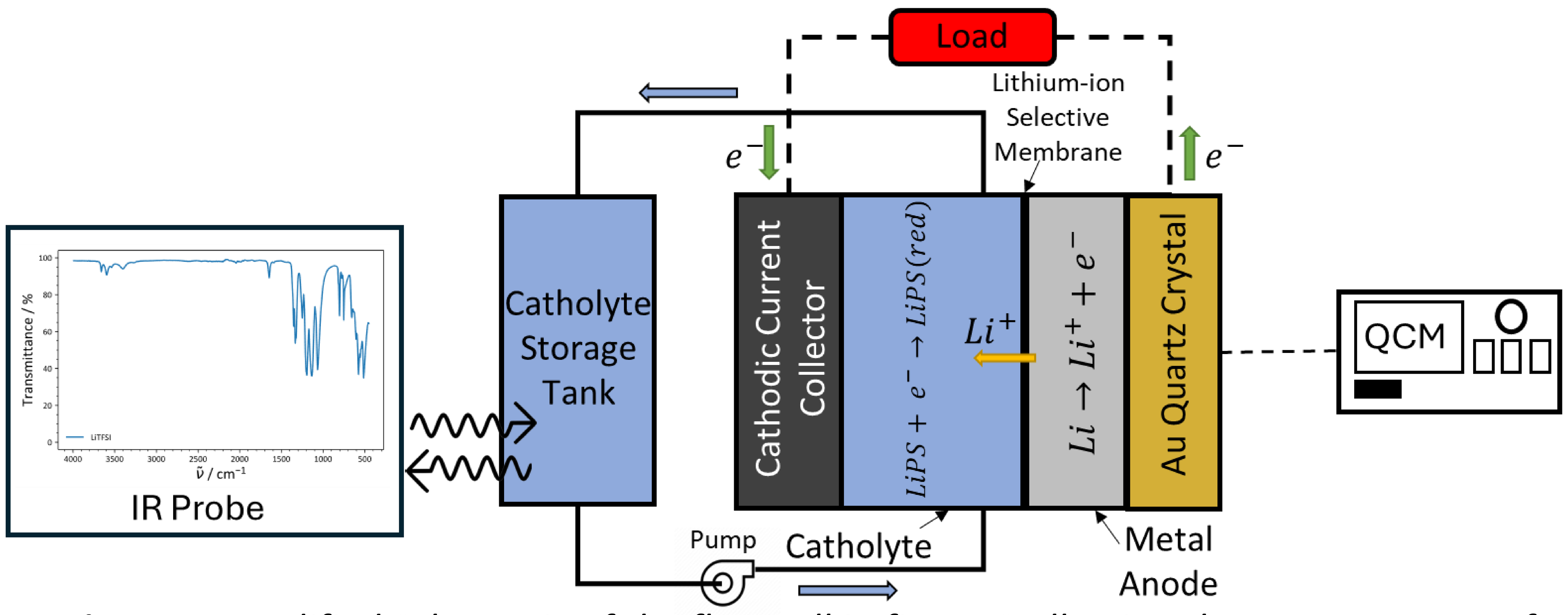

Figure 5: *Modified Schematic of the flow cell in figure 2 allowing the measurement of IR spectra and mass deposition*

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