Supporting information for

In Situ MRI of operating solid-state lithium metal cells based on ionic plastic crystal electrolytes

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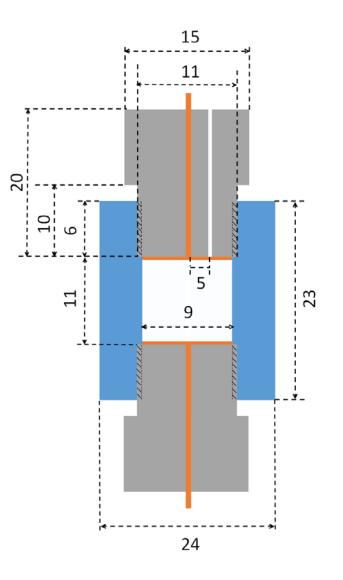
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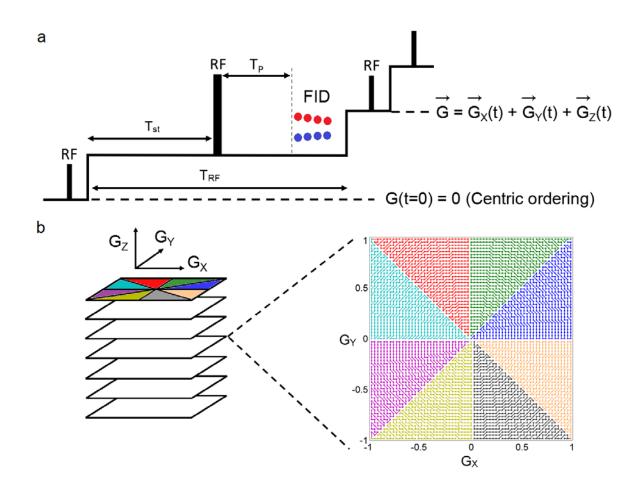
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## Disk-cell design



**Figure S1** The diagram of the air-tight symmetric disk-cell. The cell material is polyetheretherketone (PEEK). The dimensions are given in mm. Coaxial holes are 0.5 mm. An air relieve channel is provided in the top end plug to prevent air pressure build up during cell assembly. The air relieve channel was sealed with epoxy resin following the completion of cell assembly.



**Figure S2** 3D Centric-Scan SPRITE experiment. (a) Three orthogonal gradients  $G_X$ ,  $G_Y$  and  $G_Z$  are incremented with a period  $T_{RF}$  starting from the k-space origin (G = 0). The gradient stabilization period ( $T_{st} = 1 \text{ ms}$ ) is followed by a low flip angle broadband RF pulse ( $\alpha \approx \pi/40$ ; 2 µs), phase encoding period ( $T_P \ge 0.05 \text{ ms}$ ) and complex FID sampling with a dwell time of 4 µs. (b) An example of k-space sampling employing eight sectoral interleaves in the X-Y plane (right) and equal increments along Z || B<sub>0</sub> (left). The gradient values ( $G_X$ ,  $G_Y$ ,  $G_Z$ ) are selected on the Cartesian grid and cover a matrix of  $64 \times 64 \times 64$  (64 planes, 8 sectors per plane). A magnetization recovery delay (at least  $5T_1$ ) is set between the interleaves to avoid the artifacts due to magnetization saturation effects. The nominal image resolution is determined by the maximum gradient strength ( $G_{max}$ ) and  $T_P: 2\pi/(\gamma T_P G_{max})$ .

## Electrochemical performance of the disk cell

Electrochemical performance of the cell can be characterized by the potential across the device measured during galvanostatic cycling (at a constant current density). The total cell resistance is contributed by electrode/electrolyte interfaces and the bulk electrolyte.

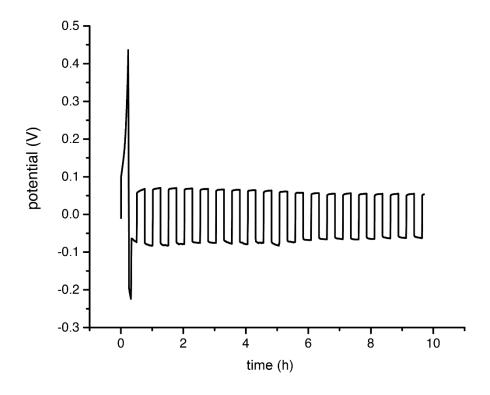
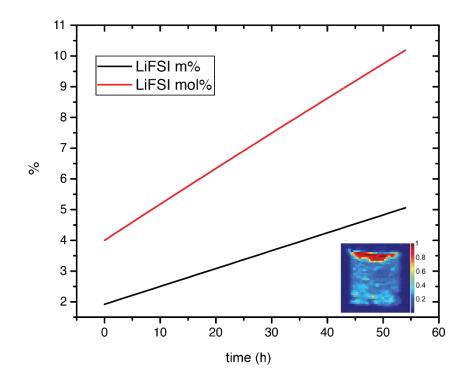


Figure S3 A preconditioning effect observed in the model disc-cell. The cell was cycled at a current amplitude of  $10 \,\mu$ A with a 15 min period at 21 °C.



**Figure S4** Estimated LiFSI concentration (weight and molar percent) near the working electrode as a function of time. Estimations are based on the Faraday's constant and the current magnitude.

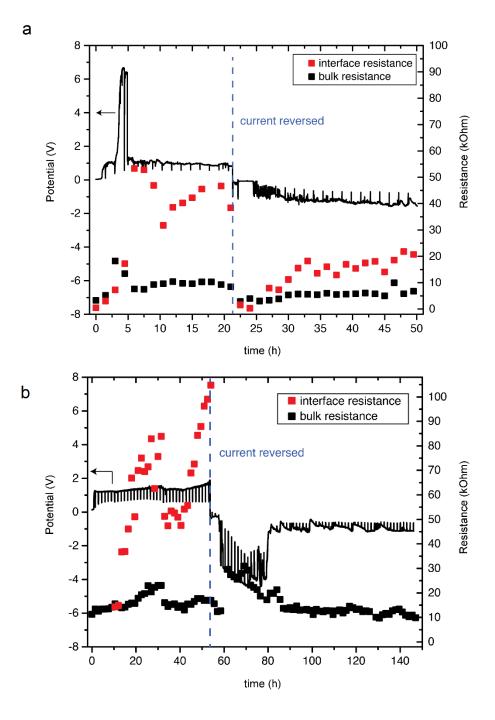


Figure S5 The potential profiles of the model Li-symmetric cells: (a) Wire-cell and (b) Diskcell. The cell resistances were monitored by EIS measurements every  $\approx 1.5$  hours during the charging and discharging processes. Note, the regular vertical lines on the profiles indicate interruptions of the galvanostatic process required for the EIS measurements. Bulk and interface resistances were extracted from the EIS Nyquist plots. The dash lines indicates the time when the current was switched from 10  $\mu$ A to -10  $\mu$ A.