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In-operando Electron Paramagnetic Resonance Spectroscopy of Lithium Anodes During Electrochemical Cycling

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The formation of mossy lithium and lithium dendrites so far prevents the use of lithium metal anodes in lithium ion batteries. *In-operando* measurement techniques to monitor mossy lithium and dendrite formation during electrochemical cycling can help to develop solutions for this problem [1].

In-operando conduction electron paramagnetic resonance (EPR) spectroscopy [2] is presented as a modality for time-resolved monitoring of lithium plating/dissolution mechanisms in a lithium-metal/$LiFePO_4$ (LFP) cell. The experiments are made possible by a novel concentric battery cell design that is compatible with resonators used in standard EPR spectrometers operating in the X band frequency range. It is shown that the time-resolved *in-operando* EPR data are consistent with *post mortem* scanning electron microscopy (SEM) analysis.

To demonstrate the viability of the *in-operando* EPR method, two cells using different electrolytes were studied. When using an electrolyte containing fluoroethylene carbonate (FEC) additive [3], a higher reversibility of the lithium anode and a reduced formation of mossy lithium were observed.

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References

[1] R. Bhattacharyya, B. Key, H. Chen, A. S. Best, A. F. Hollenkamp, and C. P. Grey, "In situ NMR observation of the formation of metallic lithium microstructures in lithium batteries.," *Nat. Mater.*, **9**, 504–510 (2010).

[2] G. Feher, A. F. Kip, "Electron Spin Resonance Absorption in Metals. I. Experimental," *Phys. Rev.*, **98**, 337–348 (1955).

[3] R. McMillan, H. Slegr, Z.X. Shu, and W.D. Wang, "Fluoroethylene carbonate electrolyte and its use in lithium ion batteries with graphite anodes," *J. Power Sources*, **81–82**, 20–26 (1999).

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