# Thermodynamic reduced order models to assess degradation in lithium ion cells

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Accurate, quantitative and rapid diagnosis of structural changes during aging of Li-ion cells is necessary for the next generation of batteries for energy storage and electric vehicles. Identifying and quantifying the mechanisms driving degradation in Li-ion cells can improve safety and estimates of state of health (SoH) and remaining useful battery life.

Entropy profiling (EP) [1] has recently been applied to observe changes from aging. This method yields complementary information to open circuit voltage (OCV) and incremental capacity (also known as $dQ/dV$) measurements. In EP, the response of the cell voltage to the temperature is monitored, yielding the system entropy change $\Delta S$, which is sensitive to configurational changes in the Li/vacancy structure. Our previous model related the $\Delta S$ and $dQ/dV$ signals to the fraction of point defects in the cathode [2], showing pinned Li sites near the defects to be responsible for the peak amplitude reductions with higher defect fraction.

Here, we combine parameterised mean-field models [3] with entropy and OCV profiles of coin cells with spinel lithium manganese oxide (LMO) cathodes of variable composition. We assess a) the mininium number of parameters necessary and b) the overall validity of the fitted parameters. The effects of point defect formation are distinct from from those arising from loss of lithium inventory and loss of positive active material [3]. $dQ/dV$ and entropy responses exhibit systematic decreases in peak amplitude dependent on the defect fraction in the cathode. The models allow high throughput validation of experimental profiles dependent on the simulation parameters.

The method has the potential to provide rapid diagnosis of the structure changes during cell aging, while explaining how and why the peaks shift in voltage and change in amplitude during cell aging and at variable temperature.

## REFERENCES

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