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The overvoltages generated in NMC cells during charging (i_{CH}) and discharging (i_{DCH}) and the OCV hysteresis (V_{HYS}) are obtained from the difference between the cell voltages and OCVs (Fig. 1), as described in the Methods section. The overvoltages are calculated at low rates ($C/50$ and $C/25$) for fresh and aged cells (Fig. 2). In general, we find that the generated overvoltage increases during cycling, except for particular SoC values, such as those near 60%, when discharging at $C/25$ (Fig. 2d).

Overvoltages generated in NMC cells at $C/50$ during (a) charging and (b) discharging and at $C/25$ during (c) charging and (d) discharging. The increases in overvoltage after a cycle-ageing test are shown in the bottom panels.

We find that the overvoltage depends on the charge and discharge rates, as expected from the literature²⁰. However, we note that the overvoltage rate dependence is also SoC dependent. For instance, this dependence can be clearly observed in the discharge overvoltage of an aged cell if we compare the results for an SoC of 64% at $C/50$ (Fig. 2b) and $C/25$ (Fig. 2d), in which a large variation is observed compared with neighbouring SoCs.

We find that the OCV hysteresis is larger for the aged cells (Fig. 3), except for SoCs, of approximately 25% and 13%, for which there is no variation with ageing. For both ageing states, various peaks are observed in the curves, which are related to a larger hysteresis at some particular SoCs. We find that these peaks tend to shift to higher SoCs as the cell ages.

If we compare the hysteresis increase during the cycling test (DV_{HYS}) with the difference between the increases in charge and discharge overvoltages ($Di_{CH}-DCH$), we find a correspondence between the two parameters (Fig. 5), as observed for the NMC cells (Fig. 4). Although the increase in discharge overvoltage is negative, the larger increase in charge overvoltage follows the same tendency as the hysteresis increase, and both terms have similar values. Larger differences can be observed at SoC values above 95%.

Incremental capacity of the (a,b) graphite and (c,d) NMC electrodes configured as half-cells during charging at $C/25$ and OCV hysteresis at room temperature. (a,c) represent fresh electrodes, and (b,d) represent aged electrodes. The yellow areas represent single-phase regions, and the grey areas delineate the operating SoC of the full cell.

Lfp battery cycle life

We analysed two types of commercial Li-ion cells with $\text{LiCoO}_2\text{-Li}(\text{NiMnCo})\text{O}_2$ (LCO-NMC or simply NMC) and LiFePO_4 (LFP) as the positive electrode and graphite as the negative electrode. The NMC cells had a capacity of 2.8 Ah and were supplied by LG_Chem in the 18650 format. The expected cycle life reported by the manufacturer at which the cell maintains a capacity equal to or higher than 78% of the nominal capacity is 300 cycles when charging and discharging at C/2 at a temperature of $23 \pm 2^\circ\text{C}$.

LFP cells were provided by AA Portable Power Corp in the 14430 format with a nominal capacity of 0.4 Ah. The expected cycle life reported by the manufacturer at which the cell maintains a capacity equal to or higher than 70% of the nominal capacity is 1000 cycles when charging and discharging at a symmetric rate of C/5 at a temperature of $20 \pm 5^\circ\text{C}$.

To evaluate the cells at various ageing levels, the cells were subjected to a cycle-ageing process. The cells were charged at a constant current and voltage at the recommended C-rate and discharged at the maximum discharge C-rate allowed by the manufacturer.

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