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Looking non-destructively at the aging mechanisms in commercial LiFePO₄//C 18650 cells with neutron diffraction and electrochemistry

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Aging in identical commercial 18650 LFP//C Li-ion cells, differing only in the choice of the carbon anode, was studied in a non-destructive way with neutron diffraction and electrochemistry. In all cells, the irreversible capacity losses due to active lithium losses upon battery formation and cycling were quantified by neutron diffraction. These values were validated by electrochemical techniques. The LFP//MCMB cell, having mesocarbon microbeads as carbon anode material, shows an excellent performance with only a 8% of relative capacity loss at 1C after 4780 cycles. The capacity drop was found to be related to the loss of active lithium and can be detected by neutron diffraction. The LFP//NC cell, having needle coke as carbon anode, suffers a higher irreversible capacity loss, probably due to formation of a thicker SEI layer. It also shows a poorer cycling performance compared to LFP//MCMB, with 23% relative capacity loss at 1C. Only 8% of this loss is due to decrease of active lithium amount and can be detected by neutron diffraction. No appreciable structural degradation of electrode materials can be seen in any cell with neutron diffraction. Based on the neutron diffraction results we can rule out a capacity decrease due to partial loss of carbon or LiFePO₄ active material from electrode delamination or particle isolation. For the mesocarbon microbead powder, XRD measurements reveal a larger c-lattice parameter and a smaller crystalline size, as compared to the needle coke powder. The comparative poor cycling performance of the LFP//NC cell is probably due to the higher initial irreversible capacity loss due to SEI formation (which grows more rapidly on aging), different charging kinetics and structural differences in the carbon powders at the anode.

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