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Aging studies and influence of anode in LiFePO4-based cells with neutron diffraction

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Aging in identically prepared 18650-type LFP/C cells, differing only in the choice of the carbon anode, was investigated by neutron diffraction. In all cells, not only the irreversible capacity losses due to formation but also the active lithium losses due to cycling could be quantified by neutron diffraction and were validated by electrochemical measurements. The LFP/MCMB cell, having mesocarbon microbeads as carbon anode, showed an excellent performance, suffering only a 8% relative capacity loss on 1C charge-discharge for 4780 cycles, and no loss due to calendric aging. The LFP/NC cell, having needle coke as carbon anode, suffers a higher irreversible capacity loss, probably due to formation of a more unstable SEI layer. It also shows calendric aging as well as a poorer cycling performance compared to LFP/MCMB, with a 23% relative capacity loss under similar cycling conditions. This loss is mainly due to active lithium loss and can be detected by neutron diffraction. With neutron diffraction we can rule out capacity loss due to structural degradation, partial loss of carbon or LiFePO4 active material from electrode delamination or particle isolation. In LFP/NC cells, we were able to recover part of the lost capacity at 1C by a slower charging. However, in LFP//MCMB cells, we were not able to recover any of the lost capacity by this procedure. Thus, kinetics plays a more crucial role in LFP/NC than in LFP/MCMB cells. The comparative poor cycling performance of the LFP/NC cell is probably due to the higher tortuosity of the NC anode.
N. Paul, J. Wandt, S. Seidlmayer, S. Schebesta, M. J. Mühlbauer, O. Dolotko, H. A. Gasteiger, R. Gilles, Journal of Power Sources 345 (2017) 85-96.

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