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Effects of lithiation on the fracture toughness and mechanical properties of LiMn2O4 cathode battery materials

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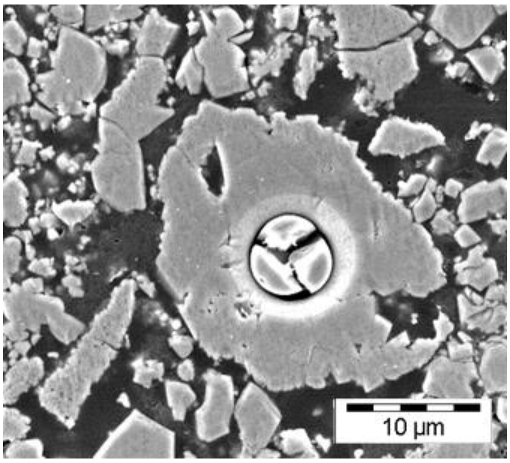
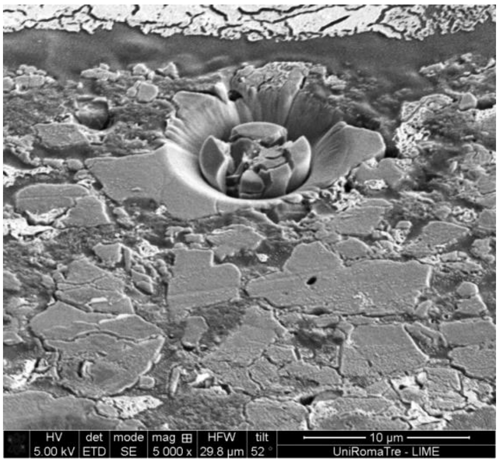
The micro-pillar splitting method has been used to assess the influence of lithiation on the fracture toughness of $Li_xMn_2O_4$ micro-particles used as cathode materials in lithium ion battery composites. The materials under investigation consisted of hard LiMn2O4 particles embedded in a soft and compliant epoxy matrix to form the composite electrode. Five different samples were extracted from commercial battery cells at different states of charge (SoC% = 0-20-50-75-100%). These correspond to different lithium concentrations in the particles, as measured by inductively coupled plasma optical emission spectrometry (ICP-OES). Experimental results from the pillar splitting experiments show a significant effect of the SoC%, and therefore the lithiation level, on the fracture toughness and failure mechanisms of the $Li_xMn_2O_4$ particles. Specifically, the toughness of the fully charged electrodes (de-lithiated material) is much lower that the fully discharged electrodes. SEM observation of split pillars (see figure) confirms a significant change in toughness of the materials as a function of the lithium concentration in the particles. The results compare well with recent investigations where a loss in ductility of electrode materials has been observed after de-lithiation. This suggests that a knowledge of the changes in toughness of the materials may be extremely important for prediction of in-service damage of the electrodes due to diffusion-induced stress during charge/discharge cycles. An analysis of pillar splitting for a hard film on a compliant substrate material shows that the critical load for splitting is relatively insensitive to the substrate compliance for a large range of material properties. This ensures a correct estimation of the critical splitting load in the case of the composite materials studied in this investigation.



Fully charged electrode

(very brittle failure – low Kc)



Fully discharged electrode

(damage tolerant behavior – at least 30% higher Kc)