In-situ Study on Working SnO Anode Particles of Li-ion Battery by Transmission X-ray Microscopy (TXM)

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A tomographic transmission X-ray microscopy (TXM) was constructed at beam line 01B1 of the National Synchrotron Radiation Research Center (NSRRC) in Taiwan, R.O.C. The advantages of hard X-ray imaging are high penetration for nondestructive and *in situ* imaging, short wavelength for high resolution, rich image contrast mechanism, element specific and no charging effect on image resolution, which is a powerful tool to *in situ* monitor the bulk material change during discharging and charging process.

Sn-based Li storage materials have once been considered as the most promising replacement of carbonaceous anodes in lithium ion batteries. Nevertheless, the stress-induced pulverization of the electrode caused by the large specific volume change (Li^+ insertion) in discharging and charging (Li^+ extraction) reactions is observed. The potential curves of SnO for first two cycles are shown in Fig. 1. The denotations of a~m respectively represent the corresponding statuses related to Fig. 2. Then, Fig. 2 shows the TXM images measured *in situ* during discharging and charging processes. Before cycling, the SnO particles are in an integrated form (Fig. 2(a)). During first constant current discharging stage (Fig. 2(b)~2(e)), the perimeter of SnO particles become vague. It might be due to the formation of Li_2O which means the SnO is reduced to Sn. Upon further discharging at lower potential window (Fig. 2(f)~2(g)), the reaction of Li with Sn is involved. It's worth mentioning that the volume variation is dramatic during the formation of Li_2O process. Moreover, this phenomenon does not go back upon charging, which may indicate that the Li was trapped, thus the first coulombic efficiency is around 65%. It is obvious that the end of first cycle, the shape of the SnO is retained. The structural stability of SnO is quite reversible. During 2nd cycle (Fig. 2(k)~2(m)), a reversible structural variation was observed. The volume variations at second cycle are less than the variations observed at first cycle which may suggest the reversibility is obtained after first cycle. There is an idle period about 3h between first and second cycle. Not like pure Sn electrode which has dramatic structural change in idle period, the SnO electrode did not show any change on structure. To our knowledge, this is the first time that the images are directly being captured during Li insertion and extraction processes. TXM strongly provides the evidence, showing the changes of particle size and morphology during Li alloying and dealloying processes.

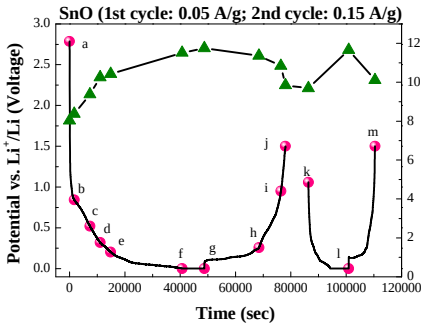

Fig. 1: the potential curves of SnO and corresponding dimension changes at specific status.

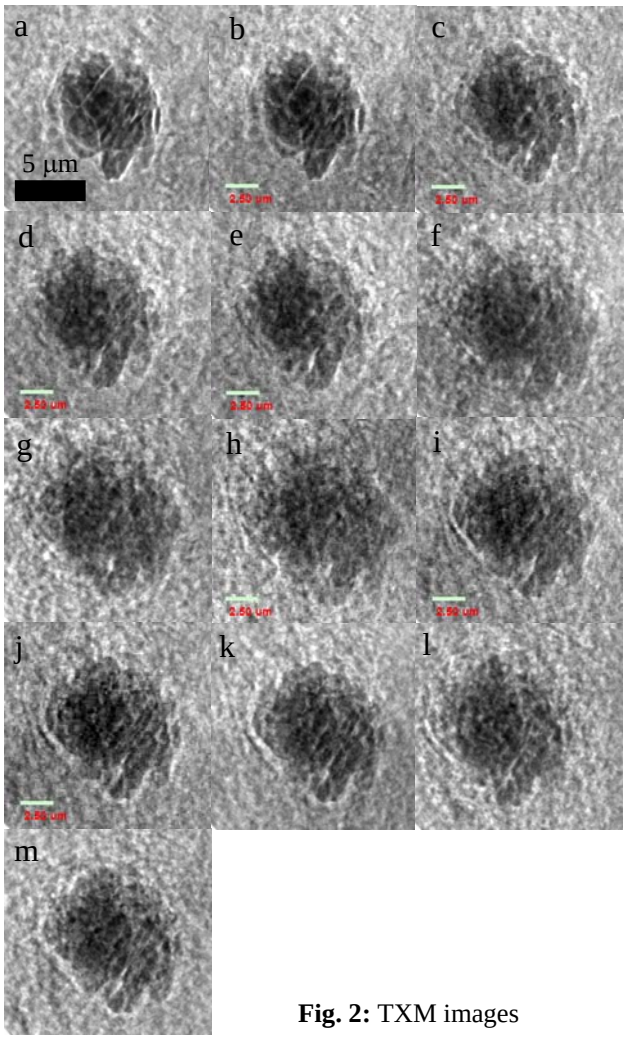

Fig. 2: TXM images