



Communication

Rate mechanism of vanadium oxide coated tin dioxide nanowire electrode for lithium ion battery

Lifen Wang^{a,b}, Jian Yan^c, Zhi Xu^a, Wenlong Wang^a, Jianguo Wen^{b,*}, Xuedong Bai^{a,*}^a Beijing National Laboratory for Condensed Matter Physics and Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China^b Center for Nanoscale Materials, Nanoscience and Technology Division, Argonne National Laboratory, Argonne, IL 60439, USA^c School of Materials Science and Engineering Hefei University of Technology, Hefei, Anhui 230009, China

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ABSTRACT

Correlating composition and structures with battery performance is key aspect of electrode material design and improvement. Here utilizing *in situ* open cell transmission electron microscopy, we studied the *in situ* cycling rate performance of vanadium oxide coated tin dioxide nanowire electrode by tuning the lithiation/delithiation current. *In situ* results show that the good rate performance of such high capacity compositional material lies in the layered vanadium oxide coating strategy. For cycling at high rate, the layered vanadium oxide also serves as fast ions and electrons transportation route while tin nanoparticles aggregate to the surface with sizes controlled by the coating layer, cycle induced volume change is released to the surface and excellent mechanical tolerance of tin nanoparticle and inner nanowire ensure improved cyclability of the electrode.

1. Introduction

The increasing needs for high energy density and high power batteries have generated significant research interest in lithium ion batteries [1,2]. Tin-dioxide is one of the most promising potential materials for lithium ion battery with a high theoretical capacity of 781 mAh/g [3–5]. However, the lithium oxide matrix produced in the first cycle inhibits the conductivity of lithium ions and causes severe capacity fade in subsequent cycles. Tin aggregation on the lithium matrix, which may penetrate the separating membrane and lead a short circuit is another concern [6]. To mitigate these adverse effects, a variety of nanoscale structures have been designed [7–14]. For example, a yolk-shell structure in which the active material is encapsulated inside elastic carbon spheres has demonstrated long cycle life [13]. Similarly, for V₂O₅ coated SnO₂ nanowires, cyclability is much improved at high charge and discharge rate with a high capacity. However it is not clear how this composition structure play such a strong role on the performance.

Here, utilizing *in situ* open cell TEM, we identify a key mechanism in which V₂O₅ coated SnO₂ nanowire can achieve excellent high rate performance based on the lithium storage mechanisms of SnO₂ nanowire and V₂O₅ coating layer. The *in situ* results indicate that tin nanoparticles formed on the conducting V₂O₅ surface at the first high rated lithiation process exhibits both high capacity and cyclability.

2. Materials and methods

Nanosized half-cell lithium ion batteries were fabricated in the double tilt holder using the customer designed piezo-driven nanomanipulator [14]. Individual SnO₂ or V₂O₅ coated SnO₂ nanowire serving as working electrode was attached to a gold tip with carbon epoxy loading on nanomanipulator. Lithium metal scratched by a tungsten tip served as the counter electrode. Lithium oxide naturally formed on lithium metal surface served as solid electrolyte [15,16]. Once all materials were loaded, the double tilt holder was transferred from an argon gas filed glove box into a TEM vacuum column. Under the observation of 200 kV electron beam, nanowire was driven to touch the electrolyte. Constant current of ± 100 pA or ± 1 nA was applied on the half-cell to cycle corresponding to different rate.

3. Results and discussion

3.1. Composition structure effect on the cyclability

Fig. 1 shows a comparison between lithiation behavior of a single SnO₂ nanowire and one coated with V₂O₅. The data reveals influence of V₂O₅ coating on SnO₂ nanowire during the first lithiation process. The pristine SnO₂ nanowire was single crystalline with a straight and uniform morphology. When a constant current of -100 pA was applied to the nanowire with respect to lithium metal, lithiation started from the

* Corresponding authors.

E-mail addresses: jwen@anl.gov (J. Wen), xdbai@iphy.ac.cn (X. Bai).

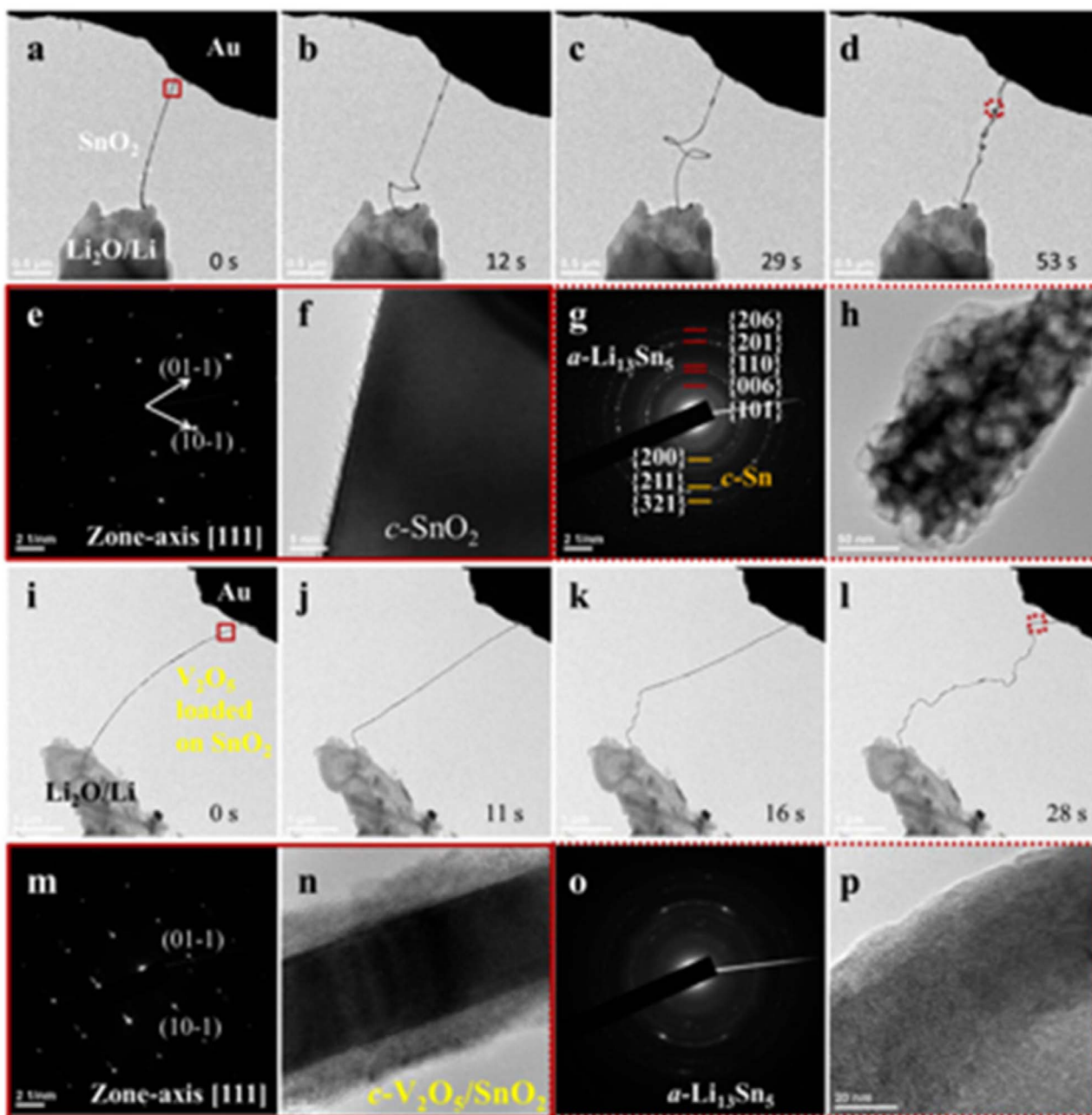


Fig. 1. Comparison of nanowires during the first lithiation process with and without V_2O_5 coated on the surface. a, Pristine SnO_2 nanowire in an open cell before lithiation started. Lithium foil adhered to a tungsten wire serving as counter electrode, the thin oxide layer covered on the foil formed when transferring from glove box to TEM column serves as electrolyte, and SnO_2 nanowire attached to a gold wire serves as the working electrode. e, f Selected area diffraction pattern (SADP) and corresponding high resolution TEM (HRTEM) image of the pristine nanowire marked by red tangle in a showing single crystalline feature. b, c, Time sequenced TEM images of SnO_2 nanowire during the first lithiation process when a constant current of -100 PA applied to the nanowire with respect to lithium. Lithiation front started from the near lithium end with enormous morphology changes of swelling and tangle introduced by lithiation. d, Thermal runaway cause breakdown of the first lithiation process. g, h SADP and corresponding HRTEM image of the area marked by dashed circle in d show lithiated phase of $Li_{13}Sn_5$ embedded in Li_2O matrix. i Pristine V_2O_5 loaded SnO_2 nanowire in an open cell. m, n SADP and corresponding HRTEM image of the area marked by red tangle in i show crystalline V_2O_5 covering single crystal SnO_2 nanowire on the surface. j-l Time sequenced TEM image of the first lithiation process show elongation of the nanowire confined in the longitude direction. o, p SADP and corresponding HRTEM image of the nanowire marked by dashed red circle in l after the first lithiation process.

lithium metal end. Tremendous volume expansion occurred in both radial and longitudinal directions. Strain induced bending and curling of the nanowire accompanied the propagation of the reaction front. The single crystalline SnO_2 nanowire was converted to a mixture of crystalline Sn and $Li_{13}Sn_5$ in amorphous Li_2O matrix. The nanowire was robust mechanically despite the huge volume change, but thermal runaway was observed when the lithiation reaction front finally reached the end. At that point, the lithiated nanowire melted and separated into two parts without any further cyclability.

In this case, the reason for the thermal runaway could be the long diffusion distance of lithium ions across the entire nanowire that Joule heat was accumulated. On the other hand, the lithium oxide matrix produced from the lithiation is poor in lithium ion conduction forming local hot spot during reaction process [17]. Such thermal runaway phenomena have also been reported in other tin-oxide based lithium ion batteries [18]. As comparison, *in situ* records of the first lithiation process of V_2O_5 coated SnO_2 nanowire under the same condition is shown in Figs. 1i-j. Pristine nanowire was crystalline V_2O_5 layer loaded

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