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Electrochemical Characteristics of Si/Mo Multilayer Anode for Lithium-Ion Batteries

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Si can react with lithium to form alloys with a high Li/Si ratio [1]. Electrochemical Li-Si reaction occurs between 0 and 0.3 V against Li/Li^+, so that the battery with high-energy density can be realized. In the case of $Li_{22}Si_5$, the theoretical capacity corresponds to 4200 mAh/g. However, Si undergoes larger volume change than any other alloy material during lithium insertion and extraction. The large volume difference (up to four times larger for $Li_{4.4}Si$ compared to Si) between the lithiated and lithium-free host causes cracking and crumbling problems of the Si particles, thereby inducing the morphological changes and the losses of electrical contact between host material and current collector. Thus, its capacity on cycling fades remarkably rapid [2].

The research groups of Huggins [3,4] and Besenhard [5] have proposed that the volume expansion due to the insertion of lithium can be reduced in micro- and submicro-structured matrix alloys. As the composite materials made of active nano-grains which reacts with lithium embedded in an inactive matrix incapable of alloy formation with lithium, silicides such as Cr-Si [6], Ni-Si [7], TiN-Si [8] and Mg_2Si [9,10,11] have been researched. The their improved cycleability suggested from an action by inactive matrix, which buffers the volumetric change of the Si phase and provides a good connection for electronic or ionic conduction during lithium insertion and extraction processes. These research efforts of mechanical alloying by high-energy ball mill have focused on reducing the particle size of the host material, using multi-phase materials or intermetallic compounds. Although these solutions are desirable, they could not overcome the kernel of problems, which related to large irreversible capacity due to electrolyte decomposition affecting the formation of solid electrolyte interface (SEI) films and the pulverization of active material.

Therefore, it is necessary to develop a material system that can certainly reduce the mechanical stress and the volume change related to the alloying process between Li and Si. As a good example, Yoshio *et al*. reported a good result that carbon-coated Si prepared by a thermal vapor decomposition (TVD) achieved a realistic alternative showing the stable capacity of 980 mAh/g until 20 cycles [12]. Also, Fujitani and co-workers suggested Si thin film having deposition thikness over 6 μm by thin film fabrication process [13,14]. Their research showed an outstanding result achieving large discharge capacity over 3500 mAh/g and high initial efficiency.

We investigated sequentially deposited Si/Mo multilayer. In thin film deposition techniques, the multilayer structure can be prepared more properly than the composite material since it offers at least a certain control of the grain size in the growth direction. Its characterization is closely related to the microstructural parameters of the multilayer structure such as the each layer thickness, the number of multilayer period, the ratio of the active/inactive elements, the degree of the interface perfection, and the selection of materials. Concerning the application of active/inactive multilayer as an anode, in which the active layer is sandwiched between two inactive layers, it can be achieved by magnetron sputtering and electron beam evaporation [15,16]. The factors such as the strong affinity between inactive material and Si, the formation of a mixed layer or an intemetallic compound via diffusion of an element constituent and high electrical conductivity of inactive material due to low electrical conductivity of Si must be considered for the selection of promising inactive material. Inactive metal incapable of alloy formation with lithium include Ca, Cu, Cr, Mg, Mn, Mo, Nb, Ni, Ta, Ti, V, Fe, Co, and the like. Among these, Mo is particularly preferred in this experiment considering factors mentioned before.

In this study, it is firstly discussed about the structural properties of multilayer and then is mentioned the electrochemical characteristics of thin film as an anode in lithium-ion batteries.

The results will be presented in the meeting.

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