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Evaluations of Si based ternary anode materials by using RF/DC magnetron sputtering for lithium ion batteries

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Generally, the high energy lithium ion batteries depend intimately on the high capacity of electrode materials. For anode materials, the capacity of commercial graphite is unlike to increase much further due to its lower theoretical capacity of 372 mAhg-1. To improve upon graphite-based negative electrode materials for Li-ion rechargeable batteries, alternative anode materials with higher capacity are needed. Therefore, some metal anodes with high theoretic capacity, such as Si, Sn, Ge, Al, and Sb have been studied extensively.

This work focuses on ternary Si-M1-M2 composite system, where M1 is Ge that alloys with Li, which has good cyclability and high specific capacity and M2 is Mo that does not alloy with Li.

The Si shows the highest gravimetric capacity (up to 4000mAhg-1 for Li21Si5). Although Si is the most promising of the next generation anodes, it undergoes a large volume change during lithium insertion and extraction. It results in pulverization of the Si and loss of electrical contact between the Si and the current collector during the lithiation and delithiation. Thus, its capacity fades rapidly during cycling. Si thin film is more resistant to fracture than bulk Si because the film is firmly attached to the substrate. Thus, Si film could achieve good cycleability as well as high capacity.

To improve the cycle performance of Si, Suzuki et al. prepared two components active (Si)-active(Sn, like Ge) elements film by vacuum deposition, where Sn particles dispersed homogeneously in the Si matrix. This film showed excellent rate capability than pure Si thin film. In this work, second element, Ge shows also high capacity (about 2500mAhg-1 for Li21Ge5) and has good cyclability although it undergoes a large volume change likewise Si. But only Ge does not use the anode due to its costs. Therefore, the electrode should be consisted of moderately Ge contents.

Third element, Mo is an element that does not alloys with Li such as Co, Cr, Fe, Mn, Ni, V, Zr. In our previous research work, we have fabricated Si-Mo (active-inactive elements) composite negative electrodes by using RF/DC magnetron sputtering method. The electrodes showed excellent cycle

characteristics. The Mo-silicide (inert matrix) dispersed homogeneously in the Si matrix and prevents the active material from aggregating. However, the thicker film than 3 μm with high Mo contents showed poor cycling performance, which was attributed to the internal stress related to thickness.

In order to deal with the large volume expansion of Si anode, great efforts were paid on material design. One of the effective ways is to find suitably three-elements (Si-Ge-Mo) contents. In this study, the Si based composites of 45～65 Si at.% and 23～43 Ge at.%, and 12～32 Mo at.% are evaluated the electrochemical characteristics and cycle performances as an anode. Results from six different compositions of Si-Ge-Mo are presented compared to only the Si and Ge negative electrodes.