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Application of plasma sputtering nanoparticle films to negative electrode of Li ion battery

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Li ion batteries are developed as a promising power source for portable electronics, hybrid electrical vehicles and electric vehicles. Increasing the capacity of negative electrode of Li ion battery is attractive route to lower battery weight and volume. Li ion batteries with nano-structured electrodes significantly improve the charge/discharge capability. There has been increasing in using nano-materials for advanced negative electrode of Li ion battery, particularly for increasing the energy density by using high specific capacity materials. An attractive alternative material for the graphite electrode is Si, large due to its wide abundance and order of magnitude higher charge storage capacity (theoretical values of 4,200 mAh/g for Si vs 372 mAh/g for graphite). However, the insertion of Li into Si to form the fully lithiated silicide $Li_{4.4}Si$ is associated with a large volume change of > 400%, which can cause the material to pulverize and lose contact with the current collector, resulting in a decrease in charge storage capacity over time. Ge is also one of most promising materials for negative electrode, due to its higher charge storage capacity of 1,600 mAh/g, where the insertion of Li into Ge to form the fully lithiated $Li_{4.4}Ge$ is associated with a volume change of > 200%. Intense research activities are currently undertaken in the field of Si and Ge materials to realize the high capacity electrode related above-mentioned problems [1].

In this study, we present performance of Li ion batteries with Si and Ge nanoparticle films, which were fabricated using a RF magnetron sputtering process under a high pressure conditions > 1 Torr. At such high pressure, the mean free pass of Si and Ge atoms released from a target is as short as the order of micrometer, and gas-phase nanoparticle formation is possible. The Si and Ge nanoparticles fabricated in discharge plasma are transported to a substrate located in the downstream region by neutral gas flow. The RF magnetron sputtering discharge plasma was generated by applying 13.56 MHz voltage to the powered electrode. The discharge power was about 50 W. The sputtering target was a poly-crystalline Si and Ge disk (1 inch) with a purity of 99.99 %. Figure 1 is an atomic force microscope image of Ge nanoparticle films fabricated in the high-pressure sputtering process. Grain size is roughly estimated to be 50 nm. In the plasma process, nanoparticle films were successfully fabricated.

We evaluated performance of Li ion batteries with Si and Ge nanoparticle films as a negative electrode. The electrolyte was 1M $LiPF_6$. For the measurement of negative-electrode property, a Li metal sheet of 0.25 mm thickness was used as a positive electrode. Li intercalation capacity was measured with applying a constant current in the potential range from 0 to 2.0 V with the computer-controlled DC source. The charge/discharge capacity of Li ion battery with Si and Ge negative electrodes shows 1,621 mAh/g for 35th cycles and 577 mAh/g for the 50th cycles, respectively, which are much larger than 372 mAh/g of conventional graphite negative electrode. This clearly shows that the developed high-pressure sputtering process is quite effective for the electrode fabrication of high performance Li ion battery.

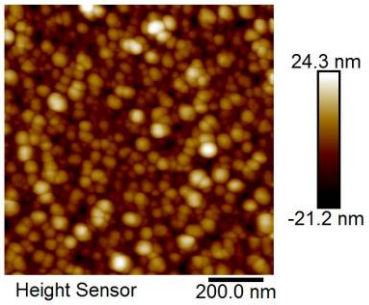

Figure 1. Atomic force microscope image of Ge nanoparticle film fabricated in a high-pressure sputtering process.

Reference

[1] M. Shiratani, K. Kamataki, G. Uchida, K. Koga, H. Seo, N. Itagaki, T.Ishihara, MRS Proceedings, 1678, mrss14-1678-n08-58 (2014).