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High-pressure He sputtering for porous-film fabrication for Li-ion-battery anode

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The automobile industry is currently shifting towards hybrid and electric vehicles powered by electrochemical energy storage systems, or batteries. However, these batteries are less fuel efficient than conventional gasoline systems, and it is therefore important to develop high-performance batteries that have a high energy density, high electromotive force, and a long charge/discharge cycle life. Recently, because of the limited capacity of carbon (graphite) anodes in Li-ion batteries, the development of alternative anode materials that are reactive with Li has been actively promoted.1-3 Among these, Si, Ge, and Sn are the most interesting materials because they have high theoretical capacities of 4,200, 1,600, and 993 mAh/g, respectively, which are much higher than the value of 372 mAh/g for conventional carbon active material. However, high-capacity Si, Ge, and Sn materials undergo large volume changes during Li alloying/de-alloying reactions of 400, 370, and 360%, respectively, and prolonged cycling leads to electrode pulverization from the current collector, resulting in capacity fading. In this study, we focus on Ge as a Li-ion battery anode material, for which the room temperature diffusivity of Li is 400 and 40 times higher than that for Si and Sn, respectively, and the intrinsic electrical conductivity σ is the order of 10 S/m which is four orders of magnitude higher than that of Si at room temperature of 10^{-3} S/m. The significant difference in σ is mainly due to the variation of carrier density, which is related to the electron density in the conduction band. Ge has a smaller band gap E_g of 0.67 eV and a higher electron density in the conduction band than Si with E_g of 1.1 eV. We fabricated nanostructured Ge and GeSn films using He radio-frequency magnetron plasma sputtering deposition.[1-6] As shown in fig. 1, monodisperse amorphous Ge and GeSn nanoparticles of 30–40 nm size were arranged without aggregation by off-axis sputtering deposition in the high He-gas-pressure range of 0.1 Torr. The Ge film porosity was over 30%. We tested the charge/discharge cycle performance of Li-ion batteries with nanostructured Ge and GeSn anodes. As shown in fig. 2, the Ge anode with an arrangement of nanoparticles showed a Li-storage capacity of 565 mAh/g after the 60th cycle. The capacity retention was markedly improved by the addition of 3at% Sn in Ge anode. The GeSn anode (3at% Sn) achieved a higher capacity of 1,128 mAh/g after 60 cycles with 92% capacity retention. Precise control of the nano-morphology and electrical characteristics by a single step procedure using low temperature plasma is effective for stable cycling of high-capacity Ge anodes.[2]

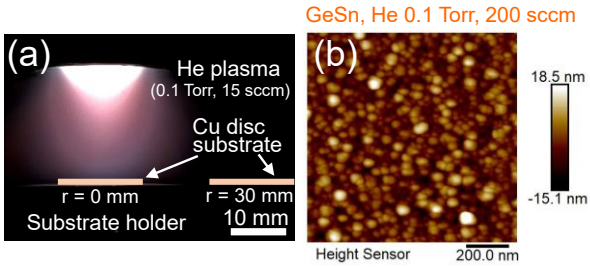

Figure 1: (a) image of plasma emission in front of Ge sputtering target. (b) surface AFM image of Ge nanoparticle anode film deposited in He high pressure condition of 01 Torr.[2]

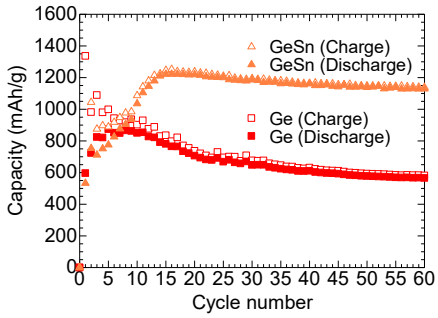

Figure 2: Cycle performance of Li-ion batteries with Ge and GeSn (3at% Sn) anodes deposited at a substrate position of 30 mm under a He gas pressure and gas flow rate of 0.1 Torr and 200 sccm, respectively.[2]

References

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