A Facile Synthesis of Silicon Oxide-Graphene Composite for Rechargeable Lithium Battery Anode

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Carbon composite materials of silicon and silicon oxide (e.g., SiO) have been considered as alternatives to graphite for next generation rechargeable lithium batteries due to high theoretical capacity of 3579 mAh/g for silicon at room temperature, which is about ten times than that of commercial graphite anode, and safe operation voltage above lithium.[1] Earlier reports showed that silicon oxide with low fraction oxygen less than 1.3 mole improves capacity retention despite the reduction in capacity.[2] Silicon oxide is known to form irreversibly inactive phases of lithium oxide and lithium silicates during initial charging (lithiation),[3] lowering of initial coulombic efficiency, but stabilizing the cycling ability by accommodating the volume change.[4,5] For the synthesis of silicon oxide, earth-abundant silica should be the most economic source of silicon.[6] In this work, we report a facile synthetic method to prepare amorphous silicon oxide at low temperature using silica, and then silicon oxide-graphene composite material, and the evaluation of cycling ability of composite anode.

Crystal structure of as-synthesized silicon oxide powders was examined using high-resolution transmission electron microscopy and X-ray diffraction, and particle morphology using field-emission scanning electron microscopy. The 2016 coin lithium cell, which consists of silicon oxide-graphene composite electrode as a working electrode and lithium metal as counter electrode, and 1M $LiPF_6$-containng carbonate based electrolyte, was assembled in the Ar-filled glove box for the evaluation of electrochemical charge-discharge cycling ability between 0.05 and 1.5 V at 300 mA/g (~0.2 C).

Fig. 1 shows the voltage profiles of the lithium cell with silicon oxide-graphene composite electrode at different cycle number. The plateau at 0.32 V during the first charging is due to the lithiation of silicon oxide forming lithium oxide, lithium silicates and lithiated silicon. Overall, smooth profile shape by lithiation and delithiation of the composite is due to amorphous silicon oxide, and the formation of amorphous silicon and lithiated silicon.[1,5] The first charge and discharge capacities are 1499 and 1205 mAh/g, respectively, calculated on the basis of whole weight of composite, with initial coulombic efficiency of 80 %. The composite electrode exhibits a stable cycling ability over 30 cycles, delivering discharge capacities of 1205 –1078 mAh/g. Discharge capacity is retained as 89 % of the initial capacity after the 30th cycle, with the well-maintained coulombic efficiency as 99 %. Further materials characterization, and interfacial stability to various electrolyte component and its relation to cycling performance would be discussed in the meeting.

Acknowledgements
This research was supported partly by the Korean Ministry of Education and National Research Foundation of Korea (NRF) through the Human Resource Training Project for Regional Innovation (2012026203) and partly by the Converging Research Center Program (2012K001255) through the Ministry of Science, ICT & Future Panning.

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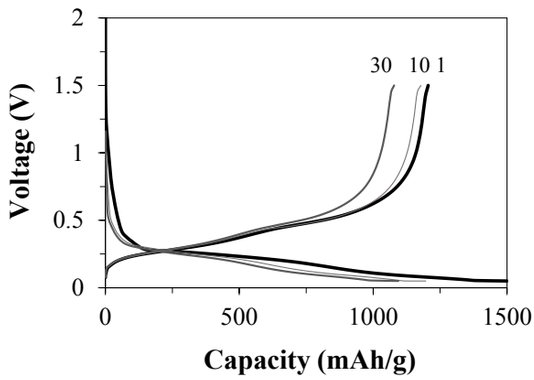

Fig. 1. Voltage profiles of the lithium cell with silicon oxide-graphene composite electrode at different cycle number.