Capacity Study of Lithium Intercalation into Carbon Nanotube Based Electrodes

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INTRODUCTION

Lithium battery research has been of great interest over the last decade. A great amount of research has been focused on the role of carbon as an electrode material for lithium-ion batteries because of its excellent intercalation properties and safety over lithium metal. Recently, a new form of carbon, the carbon nanotube, has been proposed as a potential substitute for the traditional graphite for lithium intercalation. Carbon nanotubes are essentially graphene sheets rolled into cylinders. It has been speculated that a higher lithium capacity may be obtained in carbon nanotubes if all the interstitial sites are accessible for lithium intercalation.

Recently, several groups have reported electrochemical lithium intercalation into single-walled nanotubes (SWNTs) [1-4] and multi walled nanotubes (MWNTs) [5-7]. Particularly purified SWNTs have produced large reversible capacities from 400-600mAh/g, which are higher than the capacities observed from graphite. The galvanostatic charge-discharge results from this study also show lithium capacities comparable to those reported earlier. To account for the excess lithium capacity, several mechanisms have been proposed including formation of lithium multi-layers on graphite sheets, Li_2 covalent molecules, formation of LI-C-H bonds, filling micro-cavities, and adsorption of lithium on both sides of isolated graphene layers. [8-13] Not only the large reversible capacities but also the large irreversible capacities have also been reported on SWNTs. The irreversible capacities are believed to be caused by (1) formation of solid electrolyte interface (SEI) and (2) capillary effects. On graphite, the SEI layer is a passivation layer produced from the decomposition of the electrolyte and the exfoliation of graphite surface when co-intercalation of solvated electrolyte occurs. Once formed, the SEI layer is stable and allows the reversible intercalation of lithium into graphite. The disadvantage of the SEI layer is that the formation of the SEI layer reduces the actual amount of the working electrode. SEM results from this study show the formation of an SEI layer at the surface of the SWNT electrodes. Because SWNTs are expensive, the exfoliation of SWNT during the SEI formation is a problem in terms of capacity and cost. The reversible capacities were further improved by ball-milling and chemical etching up to 1000mAh/g [1-4]. These increases in the reversible capacities coincided with reduction in the irreversible capacities of SWNTs. One explanation is that shortening or defect formation on SWNTs increases lithium diffusion into the interior or inner cores of the SWNTs and reduces the irreversible losses associated with capillary effects of lithium. [2-5] Despite these findings, the mechanism for lithium intercalation into SWNTs remains unclear. It appears that the manner in which the SWNTs are treated may affect the lithium intercalation capacities. In this study, results concerning the effects of electrode preparation on both reversible and irreversible capacities will be presented. The quality and degree of purification of the SWNTs, the procedures for fabricating the electrodes and SWNT surface modification will be discussed as they pertain to lithium capacity.

EXPERIMENTAL

As-prepared SWNTs were purchased from Carbolex and purified. Also purified SWNTs were purchased from Carbon Nanotechnologies Inc. A three-electrode system was used with two lithium foils as the reference electrode and the counter electrode and SWNTs based electrode as the working electrode. Electrochemical evaluations are performed by galvanostatic charge-discharge cycling tests and cyclic voltammetry using EG&G instrument Potentiostat/Galvanostat model 263A. The characterizations of SWNTs are performed by SEM, TEM, and Raman spectroscopy.

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