Introduction

High theoretical capacity anodes (such as Si and Sn) have attracted a lot of attention. However, they undergo enormous volume expansion in comparison to graphite, leading to rapid capacity loss and reduced cycle life, thereby inhibiting their potential for commercialization in lithium ion batteries (LIBs). Due to the disadvantages these anodes, various allotropes of carbon, such as carbon nanotubes (CNTs) and graphene nanosheets (GNSs), has been under intensive investigation. They are particularly interesting due to their ability to provide fast lithium intercalation/de-intercalation activity while maintaining structural stability during charge/discharge cycles. Furthermore, their low cost and scalability makes them an attractive candidate in the practical application of LIBs.

Experimental

A floating catalyst chemical vapor deposition (CVD) method has been developed to synthesize CNTs and nitrogen doped CNTs (N-CNTs). Three types of GNSs with varying size, edge sites, defects and layer number has been successfully synthesized. CVD technique was performed to synthesize nitrogen doped graphene nanosheets (N-GNSs) with the help of the presence of ammonia gas. More details can be found in our previous work.[1-5]

Results and Discussion

N-CNTs show approximately double reversible capacity of CNTs: 494 mAh g\(^{-1}\) vs. 260 mAh g\(^{-1}\), and present a much better rate capability than CNTs. Our work indicated that the performance improvement is attributed to high concentration nitrogen (up to 16.4 at.%) into CNTs facilitating the higher electrical conductivity and the more defect sites in anodes which provide more Li\(^+\) storage electrochemical active locations, demonstrating that one doping strategy is effective to increase anode performance in LIBs.

Three types of GNSs with varying size, edge sites, defects and layer number have been successfully achieved. It was demonstrated that controlling GNS morphology and microstructure has important effects on cyclic performance and rate capability in LIBs. Diminished GNS layer number, decreased size, increased edge sites and increased defects in the GNS anode can be highly beneficial to lithium storage and results in increased electrochemical performance. Interestingly, GNSs treated by hydrothermal approach delivered a high reversible discharge capacity of 1348 mAh g\(^{-1}\). Our results show that the controlled design of high performance GNS anodes is an important concept in LIB applications.

Similar to N-CNTs, N-GNS anode has a superior specific capacity as anodes of LIBs in comparison to GNS. Interestingly, the specific capacity of N-GNSs evidently increases with charge/discharge cycles, exhibiting superior electrochemical performance. N-GNS presented a specific capacity of 684 mAh g\(^{-1}\) in the 501st cycles while only 452 mAh g\(^{-1}\) in the 100th cycle, accounting for higher cycling stability and larger specific capacity. The significant improvement is due to the fact that the nitrogen introduction in the graphene plane leads to an increase in the number of defect sites and vacancies as lithium active sites on the surface of GNS. Our work opens a door for using novel carbon as anodes to increase battery performance.

References