Effects of Silicon and Carbon Composition on Carbon Nanotubes in Lithium-Ion Batteries
Sadie Roberts, Georgia Institute of Technology – Georgia Tech SURF 2011 Fellow
Faculty Mentor: Dr. Gleb Yushin Graduate Mentor: Kara Evanoff

Introduction
Lithium-ion (Li-ion) batteries are attractive for many applications due to their high energy density and low self-discharge rate. To further increase the energy and power densities of Li-ion technology, significant activity to increase the capacity of the anode has been conducted. Silicon anodes have the potential to increase the current state-of-art anode (graphite) capacity by nearly an order of magnitude. However, achieving high capacity and stability for Si-based anodes has been challenging due to the large volume changes that occur during electrochemical alloying and dealloying of Li. [1]

In this study, anodes consisting of vertically aligned carbon nanotubes (VACNTs) coated with nano-thick films of Si and C are considered. Prior work has previously shown the ability of this architecture to minimize degradation effects related to Li,Si, volume expansion and to enhance the anode’s thermal and electrical conductivity [2]. Here we consider the effect of varying Si and C composition (e.g. thickness) on the stability of the electrochemical performance of this anode architecture.

Procedure
VACNTs were grown via low-pressure chemical vapor deposition (CVD) [3]. Here iron(II) chloride powder is used as the growth catalyst. Several advantages of this method include fast growth rate and the lack of catalyst pre-deposition as required by other CNT synthesis techniques. After, VACNTs (~0.30 – 1.30 mm) were Si coated by low pressure CVD of SiH₄. Following, the Si-coated VACNTs (VACNT-Si) were C coated by atmospheric pressure CVD. Si and C depositions were carried out for different reaction times to vary the final Si and C composition (Figure 1).

VACNT-based electrodes were assembled into electrochemical half cells with Li foil as a reference electrode and LiPF₆ in carbonates as electrolyte. Charge/discharge cycling was conducted on a multi-channel Arbin Instruments system.

Results and Discussion
The dealloying capacity and Coulombic efficiency for batteries of different Si and C compositions are shown in Figure 2 and Figure 3, respectively. The results presented are not exhaustive as many of the electrodes synthesized have either not achieved a significant number of cycles or have yet to be tested electrochemically.

Cells with high Si content have high theoretical capacities yet it was observed that cells with lower Si content had higher dealloying capacities (Figure 2). There irregularities may be due to the poor mechanical durability of the electrode during cycling, weak electrical contact between the electrode and the cell, or to variances in the battery assembly process. Prior work has shown that the presence of an external C layer leads to higher efficiencies [2]. Interestingly, the Coulombic efficiencies achieved are similar despite changes in composition (Figure 3). This would suggest that differences in cell preparation are significant, beyond those generated by different electrode composition.

Conclusion
VACNT-based electrodes were fabricated and tested to compare the stability of the electrochemical performance as a function of Si and C composition. The results of this work are preliminary thus no conclusive statements correlating the Si composition and electrochemical stability can be determined at this time. Further consideration of more Si compositions with varying C layer thicknesses tested under additional electrochemical methods (cyclic voltammetry, impedance spectroscopy) will help reveal the relationship between Si composition (thickness) and stability.

References

Figure 2: Dealloying Capacity

Figure 3: Coulombic Efficiency

Figure 1: Si Composition (wt. %) of Electrodes

<table>
<thead>
<tr>
<th>CNT only</th>
<th>Si coated</th>
<th>Si/C coated</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>14</td>
<td>24</td>
</tr>
<tr>
<td>26</td>
<td>75</td>
<td>82</td>
</tr>
<tr>
<td>100</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* = C Coated