Ni-Composite Microencapsulated Graphite as the Negative Electrode in Lithium-Ion Batteries

I. Initial Irreversible Capacity Study

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A novel approach for suppressing the solvated lithium intercalation in graphite was developed by microencapsulating graphite with nanosized Ni-composite particles. The Ni-composite graphite showed great improvement in charge-discharge performance, coulomb efficiency, and cycling behavior when used as the negative electrode in a Li-ion cell with propylene carbonate (PC)-based electrolyte. For example, a 10 wt % Ni-composite coating increased the initial charge-discharge coulomb efficiency of SFG75 graphite (75 μm, Timcal America) from 59 to 84% and the reversible capacity by 30-40 mAh/g. The Ni-composite coating consisted of nanosized particles distributed over the surface of the graphite particle, which effectively blocked some of the edge surfaces exposed to the electrolyte. This minimized solvated lithium intercalation at these edge sites, which subsequently minimized the PC reduction within the graphite and the exfoliation of the graphene layers, and also gas evolution. Corresponding improvements in both the charge-discharge performance and safety of the negative electrode in a rechargeable Li-ion cell resulted.

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Since Sony introduced the first commercial cell in 1990 that used a carbon material in lieu of lithium metal as the negative electrode, rechargeable Li-ion batteries have become readily available worldwide. Of various carbon materials, graphite is favored because it exhibits a high specific capacity, a most desirable charge potential profile, and superior cycling behavior. However, the irreversible reactions that take place with lithium during the first cycle have been one of the very persistent problems associated with the use of graphite. In commercial Li-ion cells, the loss of lithium due to these irreversible reactions is normally compensated for by using excess cathode material, however, this leads to a decrease in the specific energy density and thus an increase in the cell cost.2,3 These irreversible reactions can also cause gas evolution, which may result in some very serious safety issues, such as cell can buckling, cell venting, electrolyte spillage, and even fire.2,4

It is widely known2-5 that the irreversible capacity can originate from electrolyte decomposition followed by the formation of a solid electrolyte interface (SEI) film on the external surface of the graphite. However, it can also arise from solvated lithium intercalation between the graphene layers and subsequent reduction of the solvent,6-7 because the formation of a solvated lithium/graphite compound like Li$_x$(sol)C$_6$ is more favorable than the formation of LiC$_6$ during insertion.8 The irreversible capacities in both cases are largely dependent on the external surface area of the electrode,5 because the exposed surface is where the irreversible reactions take place. In fact, it has been shown that the irreversible capacity associated with the edge surfaces of graphite is substantially larger than that associated with the basal plane surfaces due to solvent cointercalation into the layer spacing.7

The irreversibly capacity associated with the use of graphite also varies considerably with different electrolytes, and especially with different solvents. It has been shown7-9 that the irreversible reactions are worse in propylene carbonate (PC)-based electrolyte than in ethylene carbonate (EC)-based electrolyte, and PC alone can cause severe degradation of the graphite structure by a process called “exfoliation.”10-11 The results by Ogumi et al.12 indicated that PC begins to decompose at ca. 1 V vs. Li/Li$^+$ and continues to decompose during the remainder of the intercalation process. It has further been suggested that this decomposition occurs continuously without forming a stable passive SEI layer on the edge surfaces of the graphite particles.7-11 The net effect is that the PC solvent continuously cointercalates with Li-ions into the graphene layers and subsequently reduces, which gives rise to a large irreversible capacity during cycling.9,11 Nevertheless, PC is still attractive for use as an electrolyte in Li-ion batteries, especially for low-temperature operation, because of its high salt solubility and low melting point (~55°C).

Recently, some attempts to suppress the irreversible solvated lithium intercalation into graphite have been made by adding inorganic agents such as CO$_2$, NO$_2$, and SO$_2$ or organic agents such as crown ethers to the electrolyte.12,16 These additives contribute to the formation of a less permeable SEI film which effectively decreases the diffusion of solvated lithium ions through the surface of the graphite, but these additives still induce some irreversible capacity in the first cycle because they also react with lithium. Another approach that has been reported17 is to electrochemically reduce EC on the external surface of the graphite particles to cover them with an effective SEI film prior to use in the PC electrolyte. With these works in mind, this two-part series reports on the development of a novel Ni-composite-coated graphite material that minimizes the initial irreversible capacity in a Li-ion cell.

Three types of graphite materials were coated with Ni-composite particles using a microencapsulation technique. In Part I of this series, the effect of the Ni coating on reducing the initial irreversible capacity is revealed, based on using galvanostatic, cyclic voltammetry, scanning electron microscopy (SEM) and Brunauer-Emmett-Teller (BET) surface area results obtained with both bare and Ni-coated graphite materials. Part II of this series reports on electrochemical impedance and self-intercalation studies with the same materials.

Experimental

Three types of synthetic graphite, G1 (1 μm Aldrich), KS10 (10 μm Timcal America), and SFG75 (75 μm Timcal America), were used as received. The microencapsulation of these graphites with Ni composite was carried out at 85-90°C for 0.5 h in a plating bath containing sodium hypophosphite as a reducing agent. The procedure has been described in detail elsewhere.18 After deposition, the concentration of nickel in the plating solution was analyzed by ethylenediaminetetraacetic acid (EDTA) complexometric titration.19 In this work, 3, 5, 8, 10, and 25 wt % Ni composites were deposited on KS10 graphite, while 10 wt % Ni composites were deposited on G1 and SFG75 graphites. Following the deposition process, the graphites were washed with deionized water and dried at 90°C overnight in a vacuum oven.

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The electrochemical characterizations were carried out in Swagelok three-electrode T-cells. A typical graphite negative electrode was prepared by mixing graphite powder with 6 wt % poly(vinylidene fluoride) (PVDF, Aldrich) powder and 1-methyl-2-pyrrolidinone solvent. The resulting slurry wasroller pressed onto a stainless steel current collector and dried under vacuum at 150°C for 12 h. The resulting pellet was approximately 95 μm thick and weighed about 10 mg. The counter and reference electrodes were made of Li foil (99.9%, Aldrich), and a sheet of Whatman glass fiber membrane (Baxter Diagnostics Co.) was used as a separator. The electrolyte used consisted of 1 M LiPF$_6$ in a 1:1:3 mixture of PC/EC/dimethyl carbonate (DMC) with less than 15 ppm H$_2$O and 80 ppm HF (EM, Inc.). Also, an electrolyte consisting of 1 M LiPF$_6$ in a 1:1 mixture of EC/DMC with less than 15 ppm H$_2$O and 80 ppm HF (EM, Inc.) was used for comparison and also as received. The procedure for the fabrication of the electrodes and the Li-ion cells are described in detail elsewhere.\textsuperscript{20} The charge-discharge behavior of each T-cell was investigated using an Arbin Battery Test (BT-2043) system at a current of 0.5 mA (C/8 rate) with cutoff potentials of 0.005 and 2 V vs. Li/Li$^+$. Cyclic voltammograms (CVs) were obtained using a scan rate of 0.05 mV/s. The surface images of the bare KS10 graphite and Ni-composite KS10 prior to any charge-discharge cycles were taken using a Hitachi S-2500 Delta scanning electron microscope. A Micromeritics Pulse Chemisorb 2700 was used to obtain the specific surface areas based on the single-point, BET method. Each sample was dried in a flowing stream of argon at 200°C for 1 h prior to the BET measurement.

Results and Discussion

Initial Charge-Discharge Behavior of KS10 in 1 M LiPF$_6$/EC/DMC and 1 M LiPF$_6$/PC/EC/DMC electrolytes.—To identify the contribution of the PC solvent to the initial irreversible capacity, especially the irreversible capacity related to solvated lithium intercalation, bare KS10 was cycled in 1 M LiPF$_6$/EC/DMC and 1 M LiPF$_6$/PC/EC/DMC electrolytes separately. The first cycle charge-discharge curves are given in Fig. 1.

Although the deintercalation capacities for KS10 are about the same in both electrolytes (approximately 300 mAh/g), the first lithium intercalation capacity in the PC/EC/DMC electrolyte is much larger at 742 mAh/g compared to that in EC/DMC electrolyte at 399.7 mAh/g. Moreover, the capacities between 0.56 and 0.2 V vs. Li/Li$^+$, which correspond to solvated lithium intercalation,\textsuperscript{5} are 338.3 and 55.2 mAh/g in PC/EC/DMC and EC/DMC electrolytes, respectively. Although a quantitative comparison is not possible, because of the different amounts of EC/DMC used in each electrolyte, it is clear that the large irreversible capacity in the PC/EC/DMC electrolyte is caused by PC reduction, with a minor contribution from EC decomposition. Therefore, the remaining experiments were all carried out in 1 M LiPF$_6$/PC/EC/DMC electrolyte to investigate the effect of microencapsulating graphite particles with Ni-composite on the initial irreversible capacity arising from both PC and EC.

Initial charge-discharge behavior of Ni-composite KS10.—The galvanostatic first discharge-charge cycles for bare and Ni-composite KS10 were carried out using a current of 0.5 mA (rate of C/8) with the cutoff potential range set between 0.005 and 2 V vs. Li/Li$^+$. Note that the capacity was based on the total weight of the negative electrode. Figure 2a and b illustrate the first cycle charge-discharge curves for bare KS10 and KS10 containing different amounts of Ni composite. Three discontinuities, denoted by points A, B, and C in Fig. 2a, are observed in the intercalation profile for bare KS10 at 0.71, 0.56, and 0.2 V vs. Li/Li$^+$, respectively. The curves above and below these points exhibit different slopes, which indicates that different electrochemical reactions are taking place within these potential ranges. According to Winter’s classification,\textsuperscript{1} the region AB between 0.56 and 0.71 V vs. Li/Li$^+$ corresponds to electrolyte decomposition/SEI film formation, the region BC between 0.2 and 0.56 V vs. Li/Li$^+$ is mainly related to solvated lithium intercalation and reduction, and the region CD below 0.2 V vs. Li/Li$^+$ is associated with lithium-graphite intercalation compound (Li-GIC) formation. The capacities arising from electrolyte decomposition and solvated lithium intercalation are irreversible and cannot be recovered during subsequent cycles. For bare KS10, about 35 mAh/g of initial intercalation capacity is produced in region AB, while 338 mAh/g of capacity is produced in region BC, reflecting that 47 and 45.6% of the total intercalation capacity of 740.4 mAh/g are consumed by SEI film for-

![Figure 1. Initial charge-discharge profiles for bare KS10 graphite in PC/EC/DMC (ratio of 1:1:3) and EC/DMC (ratio of 1:1) electrolytes (C/8 charge-discharge rate).](image1)

![Figure 2. Galvanostatic first cycle charge-discharge curves for bare KS10 graphite and KS10 graphite coated with different amounts of Ni composite in 1 M LiPF$_6$/PC/EC/DMC electrolyte (C/8 charge-discharge rate).](image2)
mation and solvated lithium intercalation, respectively. Clearly, in this case, the main irreversible capacity arises from solvated lithium intercalation inside the graphene layers rather than electrolyte decomposition on the external surface. In fact, the huge solvated lithium intercalation capacity produced in KS10 may be due to the large edge surface area of KS10.\textsuperscript{5} When 3 or 5 wt % Ni-composite is deposited on KS10, the plateau observed between 0.2 and 0.56 V vs. Li/Li\textsuperscript{+} of the first intercalation curve is decreased dramatically, but can still be observed in Fig. 2a. However, Fig. 2b shows that after 8 wt % Ni-composite is deposited on KS10, the plateau totally disappears on the first intercalation curve, with essentially the same behavior exhibited by the 10 and 25 wt % Ni-composites. These results reveal that microencapsulation of Ni composite on graphite is favorable for reducing the solvated lithium intercalation inside graphite. This is shown very clearly in Fig. 3, which displays the initial intercalation capacity ($C_{\text{int}}$) and initial discharge-charge coulomb efficiencies, both as functions of the Ni-composite content.

With a 3 wt % Ni-composite coating on KS10, the initial intercalation capacity of KS10 substantially decreases from 740.4 to 494.6 mAh/g and the initial intercalation-charge coulomb efficiency increases from 38.8 to 61.3%. These marked improvements result from the decreased solvated lithium intercalation as shown in Fig. 2. When the Ni-composite coating increases from 3 to 10 wt %, more of the solvated lithium intercalation reaction is suppressed and hence the initial intercalation capacity continues decreasing while the initial discharge-charge efficiency continues increasing. However, after a 10 wt % Ni-composite coating is deposited, the initial charge-discharge efficiency curves level off, while the initial intercalation capacity continues to decrease. The leveling off of the coulomb efficiency indicates that a 10 wt % Ni-composite coating is sufficient to cover the graphite surface and suppress the solvated lithium intercalation. In contrast, the continuously decreasing initial intercalation capacity indicates a decrease in the amount of useful material for reversible Li intercalation in the Ni-composite KS10 graphite. Thus, the optimum coating is obtained with the 10 wt % Ni-composite coating on KS10 graphite, which exhibits a maximum discharge-charge coulomb efficiency of about 76% with a corresponding intercalation capacity of 410 mAh/g.

To further investigate the effect of Ni composite on the irreversible capacity of different graphites, two other types of graphite with different particle sizes and shapes were each coated with a 10 wt % Ni composite. The particle sizes of the G1 and SFG75 graphites are 1 and 75 μm, respectively. The G1 graphite particles are also round, whereas the SFG75 graphite particles are flat and flaky.\textsuperscript{6} The initial charge-discharge profiles of the 10 wt % Ni-composite G1 and SFG75 are depicted in Fig. 4a and b, respectively. Both the bare G1 and SFG75 exhibit plateaus between 0.22 and 0.62 V vs. Li/Li\textsuperscript{+}, which were shown previously to be associated with solvated lithium intercalation.

Since there are more edge surfaces in G1 than in SFG75,\textsuperscript{6} the irreversible capacity corresponding to the plateau region between 0.22 and 0.62 V vs. Li/Li\textsuperscript{+} in the G1 graphite is larger than that in the SFG75 graphite. However, these plateau regions are clearly depressed when G1 and SFG75 graphite are microencapsulated with Ni composite, and as expected, lowering the irreversible capacity associated with solvated lithium intercalation also gives rise to the improved discharge-charge coulomb efficiency. The initial charge-discharge coulomb efficiencies for bare G1 and SFG75 are 35 and 59%, respectively, while the efficiencies for Ni-composite G1 and SFG75 are increased to 59 and 84%, respectively. Therefore, even for three different types of graphite, a Ni-composite coating significantly reduces or suppresses the initial irreversible reactions, which greatly improves the initial discharge-charge performance compared to the bare graphite precursors.

Note that a small plateau appears at 1.5 V vs. Li/Li\textsuperscript{+} in Fig. 2, 3, and 4 for all Ni-composite coated graphites. This plateau is located in the potential region corresponding to water reduction. Since microencapsulation of the graphite with Ni-composite was carried out in an aqueous solution, the coated graphite possibly contains a trace amount of water that cannot be removed by moderate heating. The reduction of trace water during the first intercalation process has been confirmed by Christie and Vincent.\textsuperscript{21} Their results show that the reduction of trace water occurs at 1.44 V vs. Li/Li\textsuperscript{+}, which is close to the plateaus observed in Fig. 2a and Fig. 4. The water reduction was also observed by Dautzenberg et al.\textsuperscript{22} and Fletcher et al.\textsuperscript{23}

Cyclic voltammograms.—To further illustrate how the Ni-composite coating affects graphite performance, cyclic voltammetry was
performed on bare and Ni composite KS10 with the same samples as in Fig. 2. Figure 5a shows the CVs for the first cycle which were obtained using a scan rate of 0.05 mV/s with the cutoff potentials set between 0.005 and 1.5 V. In Fig. 5a, two reduction peaks for the bare KS10 are observed in the cathodic curve at 0.2-0.6 V and at 0.6-0.9 V vs. Li/Li⁺. These peaks do not appear in the anodic curve, indicating that irreversible reactions are taking place in these two potential ranges. They are also in the same potential ranges as the plateaus in Fig. 2, confirming that the irreversible reactions occur during the initial intercalation process. The intercalation capacities of bare KS10 corresponding to these two peaks are 309.8 and 36.9 mAh/g, respectively. These values are comparable to the values obtained in Fig. 2 for bare KS10. In other words, the intercalation capacity associated mainly with solvated lithium intercalation is 8.4 times larger than the intercalation capacity corresponding to the irreversible reactions occurring on the external surface. Moreover, bare KS10 clearly fails in inhibiting the solvated lithium intercalation. However, with a 3 wt % Ni-composite coating on KS10, the peak between 0.2 and 0.6 V vs. Li/Li⁺ (Fig. 5b) decreases dramatically. When increasing the Ni-composite content even further from 3 to 8 wt %, this peak gradually disappears and does not return even for Ni-composite contents up to 25 wt % (Fig. 5c). These results again suggest that around a 10 wt % Ni-composite coating is sufficient to suppress the solvated lithium intercalation reaction. The capacities corresponding to these potential ranges are summarized in Table I. Comparing the bare KS10 with the 3 wt% Ni-composite KS10 shows that the capacity associated with solvated lithium intercalation decreases from 309.8 to 144.2 mAh/g; this corresponds to recovering or saving 53.4% of the lost capacity. This trend of recovering more of the lost capacity continues with increasing Ni-composite content, up to recoveries slightly higher than 85%.

One point that must be mentioned is that there was no gas evolution in the T-cells using 10 wt% Ni-composite KS10 or 10 wt % Ni-composite SFG75, while there was always gas evolution in the T-cells using bare KS10 or bare SFG75. Since gas evolution is mainly caused by electrolyte reduction, especially solvent reduction, the absence of gas evolution is a strong indication that the irreversible reactions, especially solvent reduction, are suppressed by Ni-composite microencapsulation. Therefore, a Ni-composite coating on graphite has the potential to even improve the safety of a Li-ion cell. The question that remains to be answered is, what is the mechanism associated with the Ni-composite coating that reduces irreversible capacity and improves both performance and safety? To address this issue, the bare and Ni-composite KS10 were physically characterized using SEM imaging and BET surface area analysis to develop a cause and effect relationship.

**SEM images and BET surface areas.**—Figure 6 shows the SEM images of KS10 graphite before (Fig. 6a) and after encapsulation with Ni composite (Fig. 6b). The bright spots in Fig. 6b were identified as containing Ni composite using energy-dispersive X-ray analysis (EDAX). This image shows very clearly that the Ni composite is evenly distributed over the surface of KS10, and that the sizes of the Ni-composite particles are on the order of nanometers or tens of nanometers. It is clear that the nanometer-sized Ni-composite particles appear to decrease the exposed surface of the graphite particles by depositing within the surface depressions. The specific surface areas of the bare and 10 wt % Ni-composite KS10, G1, and SFG75 are given in Table II (note that they are reported in terms of the net mass of graphite in the material to examine how the Ni-composite coating decreases the surface area relative to the bare graphite). A 5 wt % Ni-composite coating on KS10 causes the surface area to decrease from 14.5 to 12.4 m²/g, and it continues to decrease slightly upon increasing the Ni-composite content. These results corroborate very well with the SEM findings. Coating G1 and SFG75

![Figure 5](image-url)  
**Figure 5.** Cyclic voltammograms for bare KS10 graphite and KS10 graphite coated with different amounts of Ni composite in 1 M LiPF₆/PC/EC/DMC electrolyte (0.05 mV/s scan rate).

**Table I. Capacities obtained from Fig. 5 in the potential range between 0.2 and 0.6 V vs. Li/Li⁺.**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Capacity (mAh/g)</th>
<th>Capacity saved (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bare KS10</td>
<td>309.8</td>
<td>0</td>
</tr>
<tr>
<td>3 wt % Ni-composite-coated KS10</td>
<td>144.2</td>
<td>53.4</td>
</tr>
<tr>
<td>5 wt % Ni-composite-coated KS10</td>
<td>108.9</td>
<td>64.8</td>
</tr>
<tr>
<td>8 wt % Ni-composite-coated KS10</td>
<td>54.3</td>
<td>82.5</td>
</tr>
<tr>
<td>10 wt % Ni-composite-coated KS10</td>
<td>54.5</td>
<td>82.4</td>
</tr>
<tr>
<td>25 wt % Ni-composite-coated KS10</td>
<td>42.8</td>
<td>86.1</td>
</tr>
</tbody>
</table>
with Ni composite also causes a decrease in the surface areas of the bare G1 and SFG75.

To identify how the Ni-composite coating improves the first discharge-charge behavior, the initial irreversible capacity was analyzed with respect to the surface area. The initial irreversible capacity $C_{irr}$ was calculated by subtracting the first intercalation capacity from the first deintercalation capacity. The relative values of the initial irreversible capacity $C_{irr}/C_{int}$ as a function of the surface area are presented in Table II. The KS10 values of $C_{irr}/C_{int}$ decreased from 1.6 to 0.3 with a decrease of the surface area from 14.5 to 11.9 m$^2$/g. The same trend was observed for G1 and SFG75.

In order to identify other factors which may contribute to the occurrence of the irreversible capacity, the irreversible capacity was normalized with surface area ($C_{irr}$/SA) and is presented in Table II. If the irreversible capacity depends on surface area only, the normalized value $C_{irr}$/SA should be a constant value for each type of graphite before and after encapsulation with Ni composite. As shown in Table II, the values of $C_{irr}$/SA change from 31.2 to 7.1 mAh/m$^2$ for KS10, from 44.4 to 23.6 mAh/m$^2$ for G1, and from 65.2 to 20.3 mAh/m$^2$ for SFG75. Clearly there is a large variation in $C_{irr}$/SA for each type of graphite. Also, the $C_{irr}$/SA values decreased by increasing the amount of Ni composite. The results indicate that other factors beside the surface area contribute to the decreased irreversible capacity. Since the Ni-composite coating consisted of 10-15 nm particles that are larger than the graphene layer space of 0.3354 nm, one cannot expect insertion of nickel composite in the layer to narrow down the layer space. In addition, the graphite particle size is essentially the same before and after encapsulation because of a small thickness of the Ni composite coating, approximately 3.4 nm (calculated). Therefore, there is no change in the graphite structure or particle size that may contribute to the variation of the irreversible capacity.

The results can be explained, however, by taking into account the fact that the solvated lithium intercalation takes place mainly on the edge surfaces of graphite rather than on the basal planes of graphite. Thus, the decreased irreversible capacity with increasing Ni-composite content suggests that the Ni-composite particles may be deposited on the edge surfaces of the graphite. This reduces the edge surfaces exposed to the electrolyte and may make the surface less permeable to the rather large solvated lithium ion. The net effect is to suppress solvated lithium intercalation at the edge sites and subsequent reduction of the solvent within the graphene layers, which substantially decreases the first cycle irreversible capacity as shown in Table II. Also increasing the amount of Ni composite covering the graphite surface narrows further the interstitial space between the nickel composite particles, which further decreases the solvated lithium intercalation and reduction in graphite.

**Cycling behavior.**—Bare and 10 wt % Ni-composite KS10 were galvanostatically cycled at C/8, C/4, C/2.5, and C/8, rates for 20, 10, and 20 cycles, respectively. The cycling performance is presented in Fig. 7 in terms of the deintercalation capacity. The deintercalation capacities of the Ni-composite KS10 and bare KS10 both decrease with increasing deintercalation rate. However, at the same rate the deintercalation capacities of the Ni-composite KS10 are generally 30-40 mAh/g higher than those of the bare KS10. Again, this improved cycling ability is most likely due to Ni-composite suppressing PC solvent intercalation into and subsequent solvent reduction within the graphite, which often leads to destruction of the graphene layers by exfoliation. The increased deintercalation capacity associated with the Ni-composite KS10 may also result from enhanced kinetics, since Ni-composite graphite exhibits an improved conductivity. Based on an impedance study, Part II of this research illustrates these features.

**Conclusions**

Three types of graphite were microencapsulated with Ni-composite nanoparticles by a novel encapsulation deposition technique. These Ni-composite graphites were investigated as the negative electrode in a Li-ion cell with PC-based electrolyte. They showed a marked improvement in discharge-charge performance, coulomb...
Table II. BET surface areas and the initial irreversible capacities of bare and Ni-composite graphites.*

<table>
<thead>
<tr>
<th>Type of graphite</th>
<th>BET surface area (m²/g)</th>
<th>C_{irr}/C_{int}</th>
<th>C_{irr}/SA (mAh/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bare KS10 (10 µm)</td>
<td>14.5</td>
<td>1.6</td>
<td>31.2</td>
</tr>
<tr>
<td>5 wt % Ni-composite-coated KS10</td>
<td>12.4</td>
<td>0.6</td>
<td>14.8</td>
</tr>
<tr>
<td>10 wt % Ni-composite-coated KS10</td>
<td>12.3</td>
<td>0.3</td>
<td>8.2</td>
</tr>
<tr>
<td>25 wt % Ni-composite-coated KS10</td>
<td>11.9</td>
<td>0.3</td>
<td>7.1</td>
</tr>
<tr>
<td>Bare G1 (1 µm)</td>
<td>11.2</td>
<td>1.8</td>
<td>44.4</td>
</tr>
<tr>
<td>10 wt % Ni-composite-coated G1</td>
<td>9.2</td>
<td>0.7</td>
<td>23.6</td>
</tr>
<tr>
<td>Bare SFG75 (75 µm)</td>
<td>3.3</td>
<td>0.7</td>
<td>65.3</td>
</tr>
<tr>
<td>10 wt % Ni-composite-coated SFG75</td>
<td>3.1</td>
<td>0.2</td>
<td>20.3</td>
</tr>
</tbody>
</table>

* The BET surface area is based on the mass of graphite only.

efficiency, and cycling behavior compared to the bare graphite precursors. For example, a 10 wt % Ni-composite coating on SFG75 graphite increased the initial discharge-charge coulomb efficiency from 59 to 84% and the reversible deintercalation charge capacity by 30-40 mAh/g. The Ni-composite coating consisted of nanosized particles distributed over the surface of the graphite particle, which effectively filled in some of the surface depressions and thus blocked some of the edge surfaces exposed to the electrolyte. This minimized solvated lithium intercalation at these edge sites, which subsequently minimized PC reduction within and exfoliation of the graphene layers, it also minimized gas evolution from the cell during cycling. In conclusion, a Ni-composite graphite negative electrode, made from a novel microencapsulation technique, has the potential to significantly improve both the performance and safety of the Li-ion cell relative to that of the bare graphite precursor.

Figure 7. Cycling behavior of bare KS10 graphite and 10 wt % Ni-composite KS10 graphite at various charge-discharge rates.

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