The Effects of Various Conductive Additive and Polymeric Binder Contents on the Performance of a Lithium-ion Composite Cathode

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1

Abstract

Fundamental electrochemical methods, cell performance tests, and physical characterization tests such as electron microscopy were used to study the effects of levels of the inert materials (acetylene black (AB), a nano-conductive additive, and polyvinylidene difluoride (PVDF), a polymer binder) on the power performance of lithium-ion composite cathodes. The electronic conductivity of the AB/PVDF composites at different compositions was measured with a four-point probe direct current method. The electronic conductivity was found to increase rapidly and plateau at a AB:PVDF ratio 0.2:1 (by weight), with 0.8:1 being the highest conductivity composition. AB:PVDF compositions along the plateau of 0.2:1, 0.4:1, 0.6:1 and 0.8:1 were investigated. Electrodes of each of those compositions were fabricated with different fractions of AB/PVDF to active material. It was found that at the 0.8:1 AB:PVDF, the rate performance improved with increases in the AB/PVDF loading, whereas at the 0.2:1 AB:PVDF, the rate performance improved with decreases in the AB/PVDF loading. The impedance of electrodes made with 0.6:1 AB:PVDF was low and relatively invariant.

Introduction

The shift from electric vehicle (EV) applications to hybrid electric vehicle (HEV) applications during the beginning of this decade has changed the focus of the Li-ion research away from a high energy application toward a high-power-pulse application.[1] The EV application leads research towards lithium polymer cells and high capacity alloy anodes, both of which face serious challenges.[2,3] The HEV application stresses the need for high transport *via* thin dimensions. Power performance can be significantly improved with modifications of the electrode design but has been done so at the expense of the inherently high energy density of Li-ion cell chemistry. Both applications require advanced electrode engineering in order to meet stringent targets. Now, with the advent of the plug-in hybrid application (PHEVs) and, to some extent, rechargeable power tools, there will be a call for higher energy at moderate power levels.[1] This new requirement of Li-ion cells will demand a comprehensive consideration of the design of electrodes. Beyond power and energy requirements, there is a 10 to15 year lifetime requirement and a 3000 to 5000 deep discharge cycle requirement that will add additional burdens to proper electrode design. In order to achieve the lifetime requirement, it is understood that impurities that lead to side reactions must be minimized; moreover, the cycle life challenge will require both robust materials and robust electrode configurations. It is envisioned that the additional cost of removing impurities and modifying materials can be offset by the savings gained over the extended 10 to15 battery life span.

Early papers centering on binder or conductive additive properties were typically in regard to performance in polymer or gel systems.[4,5] Very few systematic studies have been done on PVDF acting solely as a binder in commonly available organic electrolyte systems such as $LiPF_6$ in carbonates such as ethylene carbonate (EC) and diethyl carbonate (DEC). There is very limited reporting on power performance of lithium ion cells with regard to systematic variations of binder and conductive additives in a broader range.[6-8] Most of the early papers report the effects of electronic conductivity or capacity improvement with the changes in the level of inert materials of the ratio at a low to moderate current density (0.1C to 1C). In most cases the active materials are made in house with lithium metal as the counter electrode. Some of the surface coating techniques on cathode materials involve the use of water based systems. This is inherently problematic as most of the metal oxides react with water to form electro-inactive surface layers, significantly increasing interfacial impedance. Minimizing the active material exposure to moisture is a key to designing electrodes with high power performance. In our capacity, our electrode material mixing, coating, drying, and cell fabrication facilities are under the most stringently controlled conditions of argon (Ar) filled glove boxes. The active materials are purchased from high volume manufacturers and subject to tight industrial quality control processes. All of the electrodes discussed here can be discharged to the manufacturer's specifications.[6] The overall cell impedance in the cells we fabricate is comparable to that of industrially fabricated high-power 18650 cells.[6,9] Coin cells can be cycled through hundreds of deep discharge cycles at ambient conditions with minimal cell capacity fade. Our superb environmental controls and fabrication techniques allow us to study power performance issues with great confidence.

We feel that an in-depth study of the contribution of the acetylene black (AB) nano-conductive additive and polyvinylidene difluoride (PVDF) polymer binder in a standard lithium-ion cell system is important to fully understand the contributions of these components. We recently reported that the interfacial impedance <u>dominates</u> the lithium-ion cell impedance and the power performance is improved with certain inactive material concentrations. We demonstrated that increasing the PVDF content in a cathode electrode may lead to decreased cell impedance, and that the cell impedance could be decoupled from the electronic conductivity of the cathode electrode.[6,10,11] This current work is to demonstrate the cell performance changes as a function of the ratio of PVDF to AB, and to map out the entire performance spectra of AB/PVDF to active material concentration.

Although many different types of conductive additives have been developed for lithium-ion cells, including carbon fibers and large graphites, there is no report of the AB being replaced as conductive additive in commercial cells. Although cost may be the main reason why manufacturers have shied away from these other materials, there is an inherent advantage of AB nanoparticles which is different from other types of nanoparticles as AB forms extended network structures to provide both conductivity and superb mechanical properties at low loadings.[12,13] With regard to the polymer, there are several choices for binder materials, from fluorinated polymers to water soluble latex binders.[14-17] As far as we know, PVDF is still a premium choice for lithium-ion cathode in the commercial cells. The current study therefore focuses on the AB and PVDF system

to demonstrate the functions that AB and PVDF play in the vast majority lithium ion cells and the possible direction to take to optimize these electrodes for PHEV applications.

Experimental

Materials. Battery-grade AB with an average particle size of 40 nm, a specific surface area of 60.4 m^2/g, and a material density of 1.95 g/cm^3 was acquired from Denka Singapore Private Limited. PVDF KF1100 binder with a material density of 1.78 g/cm^3 was supplied by Kureha, Japan. Anhydrous *N*-methylpyrrolidone (NMP) with less than 50 ppm of water content was purchased from Aldrich Chemical Company. Active cathode material $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$, with a mean particle size of 10 μm, a specific surface area of 0.78 m^2/g, and lattice density of 4.73 g/cm^3, was a gift from Toda, Japan. The manufacturer-suggested specific capacity is 173 mAh/g when cycled between 3 and 4.1 V. The active material was shipped under dry conditions from the manufacturer and stored in an Ar filled glove box with oxygen content less than 0.1 ppm and dew point below -80 ºC. AB and PVDF powders were dried at 120 ºC under 10^{-2} Torr dynamic vacuum for 12-24 hours. The dried AB and PVDF powder were also stored in an Ar filled glove box. AB/PVDF mixtures were made by dissolving 5 g of PVDF in 95 g of anhydrous NMP. A given amount of AB was dispersed in the PVDF polymer solution to meet the desired AB:PVDF ratios. To ensure the thorough mixing of the AB nanoparticles into the polymer solution, sonification was used. A Branson 450 sonicator equipped with a solid horn was used. The sonication power was set at 70%. A continuous sequence of 10 s pulses followed by 30 s rests was used. The sonic dispersion process

took ~30 min. The slurry properties for all AB/PVDF in NMP were constant after 20 min of sonification. Slurries with active cathode material were made by adding the targeted amount of $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ active material to the freshly premixed AB/PVDF/NMP slurry. The cathode mixture was homogenized using a Polytron PT10-3S homogenizer at 3000 rpm for 15 min until a viscous slurry was acquired. All of the mixing processes were performed in an Ar filled glove boxes.

Film and electrode casting. In order to understand the conductive behavior of the AB/PVDF composite system, specific amounts of AB were dispersed in PVDF-NMP solutions and cast into thin films. The film compositions extend from 0.1:1 to 1:1 AB:PVDF by weight. AB/PVDF films for four-point probe dc conductivity measurements were cast onto glass surfaces. Among the AB:PVDF ratios, 0.2:1, 0.4:1, 0.6:1 and 0.8:1 AB:PVDF ratios were chosen to mix with $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ active material in the fabrication of electrodes. At 0.2:1 AB:PVDF, slurries made of 1.2, 2.4, 4.8, 9.6 and 24% AB+PVDF with the rest being active material were cast into electrodes. At 0.4:1 AB:PVDF, slurries of 2.8, 11.2, and 21% AB+PVDF were cast into electrodes. At 0.6:1 AB:PVDF, slurries of 3.2, 12.8, and 24% AB+PVDF were cast into electrodes. At 0.8:1 AB:PVDF, slurries of 3.6, 9, 18, and 27% AB+PVDF were cast into electrodes. All electrode laminates were cast on to 30 μm thick battery-grade Al sheet using a Mitutoyo doctor blade and a Yoshimitsu Seiki vacuum drawdown coater to roughly the same loading per unit area of active material. The films and laminates were first dried under infrared lamps for 1 hr until most of the NMP was evaporated and they appeared dried. The films and laminates were further dried at 120°C under 10^{-2} Torr dynamic vacuum for

24 hrs. The film and laminate thicknesses were measured with a Mitutoyo micrometer with an accuracy of ± 1 μm. The typical thickness of an AB/PVDF film is *ca.* 20 μm with an average density of 1.2 g/cm^3. The typical thickness of the AB/PVDF/ LiNi$_{0.8}$Co$_{0.15}$Al$_{0.05}$O$_2$ film is around 50 μm with an initial porosity of ~ 50% after drying. The electrodes were compressed to 35% porosity before coin cell assembly using a calender machine from International Rolling Mill equipped with a continuously adjustable gap.

Film imaging. The surface morphologies of 0.2:1, 0.5:1, and 0.8:1 AB:PVDF ratio films were imaged by scanning electron microscopy (SEM) with a JEOL FESEM set at 5 kV. The internal morphology was imaged by transmission electron microscopy (TEM) with a JEOL 200CX microscope operating at 200 kV. A RMC Boeckeler PR XL ultramicrotome was used to prepare 50 nm thin sections of the films. Samples were embedded in low-viscosity Spurr's epoxy for added stability when sectioning. Films were cryomicrotomed at -100°C using a cryogenic attachment and a glass knife. After the samples were sectioned they were then carbon coated to help dissipate charge.

Four-point probe dc testing. The conductivities of both AB/PVDF and AB/PVDF/Active Material films were measured using a four-point probe. The dc conductivity measurement was performed using a Jandel® equal-distance linear four-point probe apparatus with a Solartron 1286 Electrochemical Interface and a CorrWare® software package. A direct current was applied between the two outer probes; the voltage was registered between the two inner probes.

Coin cell fabrication and testing. Coin cell assembly was prepared in standard 2325 coin cell hardware with $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ films as the active material. The laminates were prepared with either 0.2:1, 0.4:1, 0.6:1, or 0.8:1 AB:PVDF ratios with different active material loadings. A 1.26 cm diameter disk was punched out from the cathode laminate for use in the coin cell assembly. Detailed information regarding the different laminates and the cell configuration are listed in Table 1. MCMB10-28 graphite-based active material was used in making the counter electrode. The MCMB10-28 negative electrodes have 10% excess capacity per unit area compared to the corresponding cathodes of interest, and were compressed to 35% porosity. The counter electrodes were cut to 1.47 cm diameter disks, which is physically larger than the cathode to prevent lithium deposition at the electrode edges at high charge rates. The cathode electrode was placed in the center of the outer shell of the coin cell assembly and two drops (at *ca.* 0.02 ml per drop) of 1 M $LiPF_6$ in EC:DEC (1:1 weight ratio) electrolyte purchased from Ferro Inc. were added to wet the electrode. A 2 cm diameter of Celgard 2400 porous polyethylene separator was placed on top of the cathode electrode. Three more drops of the electrolyte were added to the separator. The counter electrode was placed on the top of the separator. Special care was taken to align the counter electrode symmetrically above the cathode. A stainless steel spacer and a Belleville spring were placed on top of the counter electrode. A plastic grommet was placed on top of the outer edge of the electrode assembly and crimp closed with a custom-built crimping machine manufactured by National Research Council of Canada. The entire cell fabrication procedure was done in an Ar-atmosphere glove box. The coin cell performance was evaluated in a thermal chamber at 30°C with a

Maccor Series 4000 Battery Test System. The cycling voltage limits were set at 4.1 V at the top of charge and 3.0 V at the end of discharge. The graphite-based coin cells were first formed by going through two cycles at C/25 at 30°C before testing. The EIS tests were performed at 40% depth of discharge (DOD) at frequencies between 0.01 Hz and 10^5 Hz using a Solartron 1260 impedance/gain-phase analyzer and Z-plot software package. Hybrid pulse power characterization (HPPC) tests were performed following the established procedures.[18]

Results and Discussion

1. Conductivity log plot of AB/PVDF composites.

The interaction of the inactive materials AB and PVDF plays a critical role in providing functionality to an electrode. AB, although only present at a level of a few weight percent in the electrode composite, has significant surface area, and therefore plays a dominant role in the interaction with the polymer binder. A semi-log plot of the conductivity versus AB content given in Figure 1 points to two conductivity regions. The first region is the low AB content region between 0:1 and 0.2:1 AB:PVDF, where the conductivity is quite low. This region corresponds to the gradual formation of an AB 3-D network structure within the PVDF phase. The conductivity improves rapidly as this network is fully formed at *ca.* 0.2:1 AB:PVDF.[12] The second region is the conductivity plateau region where the conductivity changes more slowly with increase of AB content. This region corresponds to the fully formed AB network. The conductivity is controlled by the

conductivity of the junctions between the AB aggregates. Addition of AB plays a competing role to increase the conducting material but to weaken the conductive network. The AB/PVDF composite forms a conductivity plateau from 0.2:1 ratio, and peaks at 0.8:1, then drops with further increase of AB content due to insufficient binder to maintain the AB network structure.[19] Any formulation variations or process inhomogenaities will cause significant conductivity fluctuations in the electrode in the first conductivity region. The plateau region has a relatively high, stable conductivity in a range of compositions, and is the focus of this work. There are two limit compositions in this plateau region: 0.2:1 AB:PVDF composition is the beginning of the conductivity plateau. This combination also corresponds to the full formation of the AB network within PVDF and has been found for other carbon/polymer systems to correspond to the composition of highest mechanical strength.[20,21] Another unique composition is 0.8:1 AB:PVDF ratio, which gives the highest electronic conductivity in the AB/PVDF composites. Further increase in the concentration of the AB leads to a decline in the electronic conductivity as well as major reductions of the mechanical strength and is therefore not of interest. In this paper we also evaluated two intermediate compositions: 0.4:1 and 0.6:1 AB:PVDF.

The diamond points in Figure 1 correspond to the measured conductivity of the AB/PVDF films at different AB:PVDF ratios, and the solid line connecting the diamond points is for visual aid. In a working electrode, the active material is mixed into the conductivity AB/PVDF composite. Due to the semiconductive nature of the oxide active materials, the active material particles mostly take up space and create tortuosity but do

11

not significantly contribute to the electronic conductivity of the composite. Therefore, the electronic conductivity of the film will decline with the addition of active material. Assuming the AB/PVDF distribution is uniform, the dotted lines account for the conductivity proportionally to the (AB+PVDF) volume fraction raised to a factor that accounts for tortuosity. The electrolyte resistance dominates the cell high frequency cut-off impedance also known as bulk impedance when the separator between the positive and negative electrode is 25 µm thick.

2. SEM and TEM images of the AB/PVDF composites.

The morphology of the AB/PVDF composites at 0.2:1, 0.5:1 and 0.8:1 AB:PVDF ratios is studied in detail by SEM and TEM imaging techniques (Figures 2 and 3). Large domains of PVDF phases exist at the 0.2:1 AB:PVDF ratio as can be visualized by SEM and TEM images; although, as discussed, this concentration still allows for a fully formed electronic network throughout the film.[12,19,20] Increases in the AB concentration tends to reduce the PVDF islands to ever smaller domains. At the 0.8:1 ratio, the individual domains of PVDF polymer are very difficult to visualize. This is a result of the strong association between the AB surface and the PVDF polymer.[13] At 0.2:1 AB:PVDF, the distribution of AB is uniform in the micro-scale but non-uniformed at nano-scale. At this low AB loading, the AB forms a network structure that allows the film to reach the percolation threshold well before that would be expected of randomly distributed spherical particles. The PVDF provides the critical binding force for the composite to stay together. The AB network formation is critical for both high

12

conductivity and improved mechanical strength compared to pure PVDF. With an

increase in AB content, there is associated an increase in film conductivity. Films of

AB/PVDF of compositions between 1:5 and 4:5 show a transition to fewer and fewer

pockets of pure binder. At 0.8:1 AB:PVDF ratio, the polymer phase is closely associated

with the AB particles with little excess polymer visible and a maximum in conductivity.

Due to the strong interaction of the PVDF with AB, there is limited PVDF polymer

available to provide continuous adhesion through the film, thus increases in AB:PVDF

above 0.8:1 leads to oversaturation of the polymer, and hence loss of mechanical integrity

and a reduction in conductivity.

3. Cell Performance

Slurries of AB/PVDF at ratios of 0.2:1, 0.4:1, 0.6:1 and 0.8:1 were used in the fabrication

of electrodes with different $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ active material loadings to investigate

the contribution of changing the AB/PVDF loading on electrode performance. At each

AB:PVDF ratio, different fractions of active material were added to investigate the

integrated contribution of a AB/PVDF composite to the cell performance. The electrodes

were carefully designed to have similar capacities per unit area. (Table 1) Counter

electrodes were punched from the same laminate of MCMB10-28 graphite to minimize

the variation from the counter electrode. However, low loading of the (AB+PVDF)

content electrodes tend to have higher capacity, therefore a separate anode laminate is

used for all low loading of the (AB+PVDF) laminates. The 0.2:1 and 0.8:1 AB:PVDF

based films are the outer limits of the ratios we tested; the electrochemical performances

of the two compositions follow different trends as the active material content changes.

We first discuss the impact of the PVDF poor composition of 0.8:1 AB:PVDF on the cell

performance provided in Figure 4. 3.6% of the (AB+PVDF) is the minimum required to

provide enough adhesion between particles. The integrity of the electrode is adversely

affected with less (AB+PVDF). The EIS tests were done at 40% of DOD from 10^{-2} Hz to

10^5 Hz frequency range. (Figure 4-a) The squashed semi-cycle reflects the charge transfer

impedance on the active material surface. Most of the impedance of the cell comes from

the charge transfer impedance, while the bulk resistance is fairly constant with change of

the (AB+PVDF) loading. Since the anode remains the same for all compositions, the

change of the charge transfer impedance is due to the changing composition of the

cathode. The apex frequencies of the semi-circles are presented in Figure 4-a for both

semi-cycles at 27% (AB+PVDF) loading. When the charge transfer impedance grows,

the two semi-cycles combine into a single, depressed semi-circle, making it difficult to

distinguish the apex of the charge transfer impedance phenomenon of either electrode.

The positions of these two frequencies were also labeled on the semi-cycle of other

loadings of this series. The charge transfer impedance increases in the cathode with

decreases of the amount of (AB+PVDF) composite. The increased charge transfer

impedance directly correlates to the decreasing binding force and aggregation of the AB

particles in the electrode.[22,23] The binder is also adsorbed to the active material surface

leaving less binder available for adsorption to the AB conductive additive. The deficit of

binder in the electrode composite contributes to the uneven distribution of the AB

additive leading to the impedance rise at high active material loadings.[6,10,24] The full cell

HPPC results are in very good agreement with the EIS results, showing a high area

specific impedance (ASI) at low (AB+PVDF) loading and a low ASI at high loading.

We now discuss the impact of the binder rich composition at 0.2:1 AB:PVDF ratio on the

laminate properties and cell performance(Figure 5). In this composite, the carbon

agglomerates into strands of carbon surrounded by PVDF and there are regions of just

PVDF as shown in the TEM (Figure 3-a). Laminates of 1.2% (AB+PVDF) with the rest

of the electrode as $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ active material can be made with adequate

mechanical integrity. Impedance tests were performed at the similar conditions as those

performed of laminates of 0.8:1 AB:PVDF. This time, however, the impedance

systematically increases as the amount of (AB+PVDF) increases. The apex frequencies of

the semi-circles are presented in Figure 5-a. Only one depressed semi-cycle can be seen

in this series of laminates. The trend of the impedance change of 0.2:1 AB:PVDF is just

opposite to the 0.8:1 AB:PVDF series. The difference in the impedance trend is due to a

different mechanism as to where the impedance originates. The more (AB+PVDF) at

0.2:1 AB:PVDF, the heaver the coating of PVDF on each of the active material particles.

In other words, at high binder to AB ratios, the excess binder is attracted to the active

material, leading to encapsulation of individual particles. This results in an increase of

impedance with increase in (AB+PVDF) due to the Lithium-ion blocking effect of the

PVDF polymers. A decrease of the (AB+PVDF) composite content decreases the ion-

blocking effect and decreases the impedance. Since 0.2:1 AB:PVDF has excess PVDF to

provide adhesion, electrodes at very low (AB+PVDF) loadings (e.g. 1.2%) can be made

with good mechanical strength. AB loading as low as 0.2% and 98.8% active material composite in the electrode is feasible with acceptable rate performance as seen in full cell HPPC performance shown in Figure 5-b. This may open a new direction in the formulation of the power-based cells where energy density is equally important.

The 0.2:1 and 0.8:1 AB:PVDF ratios represent either too much binder or too little binder when the active material content varies. Both of them represent extreme conditions that the AB/PVDF functions in a lithium-ion cell environment. 0.4:1 and 0.6:1 AB:PVDF ratios are in between the two extremes that may yield cell behavior that is of interest for more broad based formulation requirements. The impedance trend seen in 0.4:1 AB:PVDF ratio lithium ion cell series is similar to the one in 0.2:1 series as shown in Figure 6. Based on the electron microscopy information, the PVDF is still dominant in the 0.5:1 AB:PVDF composite. An ion-blocking effect exerted by PVDF phase is still dominant with increasing of the amount of (AB+PVDF) in the composite. However, the impedance increase is less prominent with increase of (AB+PVDF) compared to the 0.2:1 AB:PVDF series. A further increase of the AB:PVDF ratio to 0.6:1 appears to further stabilized the interfacial impedance regardless of the (AB+PVDF) contend as shown in Figure 7. Very small changes of the interfacial impedance and full cell ASI come with the change of (AB+PVDF) content in the 0.6:1 AB:PVDF series.

Conclusions

16

A complete picture of AB/PVDF contribution to the lithium-ion cell power performance was mapped out. Since the bulk impedance is very similar for the cells due to controlled designs, all of the charge transfer impedances are plotted in Figure 8 to demonstrate the performance with changing composition. A few general conclusions can be drawn from this chart. At higher PVDF contents such as 0.2:1 AB:PVDF, the active material particles are encapsulated by the AB/PVDF composite leading to an ion-blocking effect that dominants the cell resistant. This effect can be reduced by decreasing the total amount of (AB+PVDF) in the electrode. At high AB contents such as 0.8:1 AB:PVDF ratio, AB aggregation is the cause of cell resistant growth when (AB+PVDF) loading decreases. An increase of the overall amount of (AB+PVDF) will significantly reduces the impedance. At 0.6:1 AB:PVDF ratio, the cell resistance is stabilized regardless of the (AB+PVDF) content. This means that neither ion-blocking effect by the PVDF observed at 0.2:1 AB:PVDF ratio nor the AB aggregation at 0.8:1 ratio is dominant in 0.6:1 AB:PVDF combinations.

There is a trade-off in the electrode design to improve energy and power performance for a given active material. High loading of (AB+PVDF) at high AB:PVDF ratios may lead to electrode with good power performance. Both AB and PVDF have low density compared to the metal oxide active materials. Therefore increase (AB+PVDF) content leads to decreased volumetric energy density. An alternative choice for power performance is to exploit the low AB:PVDF ratio composition, where impedance decreases at decrease of (AB+PVDF) content. This direction of optimization may yield electrode that has high loading of active material with low impedance. This may satisfy

applications that both energy and power are important such as in the plug-in hybrid electric vehicles.

Acknowledgements

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Table and Figure Captions

Table 1. The compositions and configurations of the cathode electrodes used to fabricate the lithium-ion coin cells.

Figure 1. DC Conductivities of the AB and PVDF composites films. (100% legend represents the bulk film; 10%, 3% and 1% represent the uniformed reduction of conductivity to these values.

Figure 2. SEM surface images of the AB/PVDF composites. A. AB:PVDF = 0.2:1; B. AB:PVDF = 0.5:1; C. AB:PVDF = 0.8:1. (Scale bars: 300 nm)

Figure 3. TEM images of the AB/PVDF composites. A. AB:PVDF = 0.2:1; B. AB:PVDF = 0.5:1; C. AB:PVDF = 0.8:1. (Scale bars: 200 nm)

Figure 4. Impedance and ASI of $LiNi_{0.80}Co_{0.15}Al_{0.05}O_2$/Graphite Cells at AB:PVDF = 0.8:1. A. EIS test results; B. HPPC test results.

Figure 5. ASI Impedances of $LiNi_{0.80}Co_{0.15}Al_{0.05}O_2$/Graphite Cells at AB:PVDF = 0.2:1. A. EIS test results; B. HPPC test results.

Figure 6. Impedance and ASI of $LiNi_{0.80}Co_{0.15}Al_{0.05}O_2$/Graphite Cells at AB:PVDF = 0.4:1. A. EIS test results; B. HPPC test results.

Figure 7. Impedance and ASI of $LiNi_{0.80}Co_{0.15}Al_{0.05}O_2$/Graphite Cells at AB:PVDF = 0.6:1. A. EIS test results; B. HPPC test results.

Figure 8. ASI of the charge transfer impedance based on EIS data vs. inactive materials weight (AB+PVDF) for lithium-ion cells.