

**Use of recycled linear low-density polyethylene carbon in Li-ion anodes**

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## Abstract

Lithium-ion batteries are commonly used in many small electronics around the world. Efforts to make components of Li-ion batteries more sustainable have ranged from use of a brown algae extract in Li-ion anodes to efforts to recycle lithium. Linear-low density polyethylene (LLDPE) has been shown to exhibit the strong conductive properties required of a conductive agent in a Li-ion anode and can be made from recycled waste plastics such as cling wrap and poly-gloves. Electrodes were fabricated using polyethylene glycol (PEG) coated magnetite as the active material, PPBT polymeric binder, and LLDPE Carbon. Electrodes made with 14.3 wt.% LLDPE did not cycle well and exhibited a poor morphology with a cracked surface and large aggregates. Simple conductivity testing using a 4-point probe and profilometry measurements showed that Super-P was orders of magnitude more conductive than LLDPE. Increasing the carbon loading to 33.3 wt.% LLDPE in the electrode drastically improved rate capabilities and capacity retention. SEM analysis showed that a higher carbon loading of LLDPE had a better morphology overall and demonstrated less cracking. However, when compared to Super-P, the electrode had larger aggregates and a higher density of clumping. EDX SEM imaging and elemental (Fe, O, C) image mapping confirmed the presence of  $\text{Fe}_3\text{O}_4$  nanoparticles, carbon additives, and PPBT binder. XPS analysis after 100 cycles confirmed the presence of an SEI layer in the LLDPE electrode. XPS on electrode slurries showed the presence of satellite peaks which confirm interactions between the polymeric binder and active material surface, regardless of carbon used. EIS testing provided information on the charge transfer resistances of Super-P and LLDPE which was consistent with the cycling trends. Overall, use of LLDPE in Li-ion batteries has been shown to work in a half-cell assembly. The performance of LLDPE does not beat the current industry standard, Super-P, but demonstrates promise for use after further optimization and analysis.

## Introduction

Lithium ion batteries are one of the most commonly used batteries in the world, known for lightweight configurations and high energy density. These batteries use lithium and carbon to store energy in atomic bonds for on demand use. Li-ion batteries consist of an anode, cathode, and electrolyte. The anode and cathode are known as electrodes, which are made of active material, conducting agents, polymeric binders, and a current collector. A large amount of research has gone into developing new materials for electrode components to increase lifetime, durability, power density, and energy density of Li-ion batteries.

Recently, there has been a focus on developing environmentally friendly materials for use in batteries to reduce carbon footprints. Use of brown algae for use in high capacity Li-ion batteries drew attention for the remarkably stable battery anode.<sup>1</sup> In this system, alginate extracted from brown algae contains a high number of carboxylic groups, unlike most polysaccharides, creating more options for binder-Si bonds. This increase in bonds leads to better Si electrode stability. Electrodes were prepared using Si nanopowder, carbon additives, and an alginate compound, and demonstrated uniform structure and smooth surface structures in addition to high capacity and stable cycling performance. This research highlighted the importance of strong active material and binder interactions and showcased an opportunity to use a renewable material in battery technology.

Linear low-density polyethylene (LLDPE) has been explored as a potential carbon source in Li-ion batteries.<sup>2</sup> Waste plastics such as cling wrap, and poly-gloves were converted into an ordered carbon linear low-density polyethylene (LLDPE) showing promise for converting waste products to valuable carbon. High quality conductive agent was created from the ‘upcycling’ of waste LLDPE; however this material has not been implemented in Li-ion anodes. Many materials

show promise as alternatives to commercially available components but run into difficulty during half-cell testing. We propose varying anode recipes to incorporate LLDPE as a carbon source as an alternative to Super-P while maintaining capacity and cycling performance.<sup>2</sup>

### Literature Review

Lithium ion batteries have transformed the world of energy storage and battery technology. However, there remains room for optimizing these batteries and adding elements to make higher performing batteries. Anode performance is at the heart of many of the performance issues of Li-ion batteries. Testing different anode materials and configurations in coin cells provides information on how the electrode cycles in a battery.

Electrodes fabricated are made of four groups: an active material, a conducting agent, a polymeric binder, and a current collector. Creating high performance anodes presents several challenges. Performance is measured by capacity, reaction potential, and volume change. High capacity and reaction potential are desired because they enhance electrical pathways while high volume changes are undesired because they lead to electrical contact loss and poor cycling.<sup>3</sup>

There have been many modifications to the traditional Li-ion battery. One such example is the use of transition-metal oxide nanoparticles for negative electrodes, demonstrating high electrochemical capacities and long capacity retention for 100 cycles with high recharging rates.<sup>4</sup> This study focused on reducing the limitation of Li-alloying agglomeration on reversible insertion of Li ions into electrodes. The reaction mechanism of lithium was different than in traditional batteries, as  $\text{Li}_2\text{O}$  is formed during the reduction of metal nanoparticles and decomposes during the oxidation of metal nanoparticles. Surface electrochemical reactivity was enhanced and led to strong improvements in Li-ion batteries.



Currently, a synthetic graphite, Super-P, is the most widely used carbon for conducting agents in electrodes. Alternative carbon sources for Li-ion batteries have been investigated. The carbon conducting agent must exhibit structured ordering and have a high electrical conductivity. More specifically, good mechanical and electrical properties come from well-packed hexagonal ladder planes in the carbon structure, leading to long range conjugation and ordered stacking of planes through pi bond interactions.<sup>4</sup> Linear low density polyethylene, or LLDPE, has been made into a graphitizable product that demonstrates these desired properties at the same level or exceeding Super-P. Use of LLDPE over Super-P is desired as cling wrap and other waste made from LLDPE can be converted to an ordered carbon, ‘upcycling’ waste products and providing a clear environmental advantage. LLDPE has not been used in electrode fabrication or cycled in a battery. Other attempts at using an environmentally conscious carbon source include a mix of Si nanopowder with alginate, a brown algae extract, which also exhibited stable cycling and high capacity.<sup>1</sup>

Active material in the electrode is another important factor in performance. A polyethylene glycol (PEG) coating on Fe<sub>3</sub>O<sub>4</sub> (magnetite) nanoparticles has been shown to help ionic conduction and increase magnetite dispersion, further increasing electron transport properties.<sup>5,6,7,8</sup> PEG reduced aggregate size and reduced electrode resistance, stabilized cycle performance, and improved rate capabilities.

The binder used in the anode system also has an effect on performance. Poly[3-(Potassium-4-Butanoate)Thiophene-2,5-diyl] (PBBT) is a carboxylic group functionalized polythiophene, and is water soluble.<sup>9</sup> Because of this, PBBT tends to form porous structures leading to enhanced electron transport in comparison to other binders such as poly(vinylidene difluoride) (PVdF), a binder widely used in Li-ion batteries. Coin cells integrating both PEG coated magnetite and

PBBT have had great cycling performance and rate capabilities, but have only used Super-P as a carbon source. Integrating a recycled carbon source that reduces plastic waste with PEG coated magnetite and PBBT could provide a new battery solution with high performance and positive environmental impacts.

### Materials and Methods:

#### **Electrode Preparation:**

Electrodes are composed of active material, a conducting agent, a polymeric binder, and a current collector. In this system, the active material is polyethylene glycol (PEG) coated  $\text{Fe}_3\text{O}_4$  (magnetite), the conducting agent is linear low-density polyethylene (LLDPE), and the polymeric binder is Poly[3-(Potassium-4-Butanoate) Thiophene-2,5-diyl] (PBBT). Copper foil is used as the current collector.

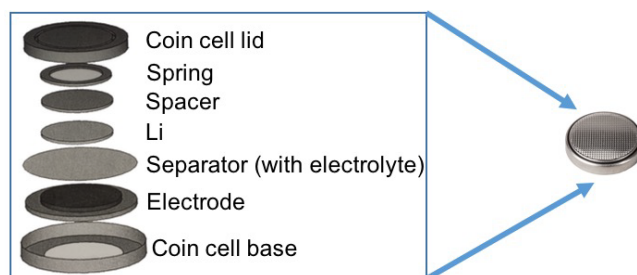
To coat magnetite with PEG, a slurry with PEG, magnetite, and water is probe sonicated for 30 seconds three times. The solution is then centrifuged for 5 minutes and rinsed with acetone 3 times. Vials with solution are dried in an oven overnight at 80 °C and a powder product is collected.

A 10 wt. % solution of PBBT in water is used as the polymeric binder. This solution is stirred on a hotplate at 80 °C for 24 hours.

To fabricate electrodes, PEG coated magnetite and LLDPE are ground in a mortar with a pestle to mix and break apart large pieces. PBBT and water are added and mixed well. This slurry is placed on copper foil and blade coated using a doctor blade to standardize thickness. The copper foil with electrode is dried at 110 °C overnight. Electrodes are punched out for coin cell assembly using a 7/16 inch cutter.

### Coin cell Assembly:

Figure 1 shows the assembly routine for making coin cells. To begin, three drops of electrolyte are placed around the coin cell base edge. The electrode is placed in the center of the coin cell base and a separator is placed above it. Five drops of electrolyte are placed on the separator. Lithium metal is centered on the separator, and a spacer and spring are put on top. A lid is placed on top and the coin cell is crimped to 1500 psi. The coin cell is then checked with a handheld voltammeter for the open circuit voltage. If during coin cell assembly the coin cell short circuited, the voltmeter would read 0 volts and the battery would not be cycled.



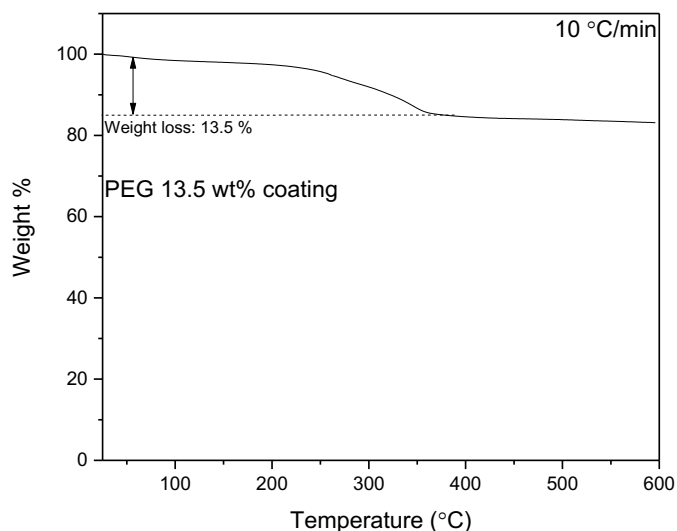
**Figure 1:** Coin cell assembly diagram

### Analysis:

SEM images and EDX analysis of sample electrodes were taken to analyze the morphology of the electrode and gain insight into potential deformities and structural issues with the electrode. XPS data was gathered on LLDPE and compared to the industry standard Super -P to confirm that similar energies and peaks were found. Coin cells fabricated were cycled to test for rate capability and capacity retention. A four-point probe was used to measure the conductivity of LLDPE/PBBT samples and Super-P/PBBT samples.

## Results and Discussion

The active material in this system is  $\text{Fe}_3\text{O}_4$ , or magnetite, has been coated with polyethylene glycol (PEG) to reduce aggregate size. Bare magnetite particles are known to aggregate in aqueous systems which leads to very unstable dispersions due to their strong Van der Waals interactions and magnetite attractions between particles. The addition of polymers and other organic molecules have been shown to stabilize the particles through steric and electrostatic forces when attached to the charged particle surface. Carbon coatings have been shown to improve the cyclic performance of magnetite by serving as a buffer to relieve volume changes to preserve electrode integrity, enhancing electrical conductivity and electrochemical performance. PEG is one of the most commonly used polymers for surface modification of magnetite nanoparticles due to its high solubility in water. PEG was physically introduced onto the  $\text{Fe}_3\text{O}_4$  particle surface to reduce aggregate size and improve particle dispersion in the system.<sup>7,8,9,10</sup> Thermogravimetric analysis (TGA) showed that PEG- $\text{Fe}_3\text{O}_4$  particles were comprised of about 13.5 wt.% PEG (Figure 2).



**Figure 2:** TGA profile of PEG coating, which is carried out in nitrogen in the temperature range of 25–600°C at a heating rate of 10°C/min, confirming ~13.5 wt.% PEG coating.

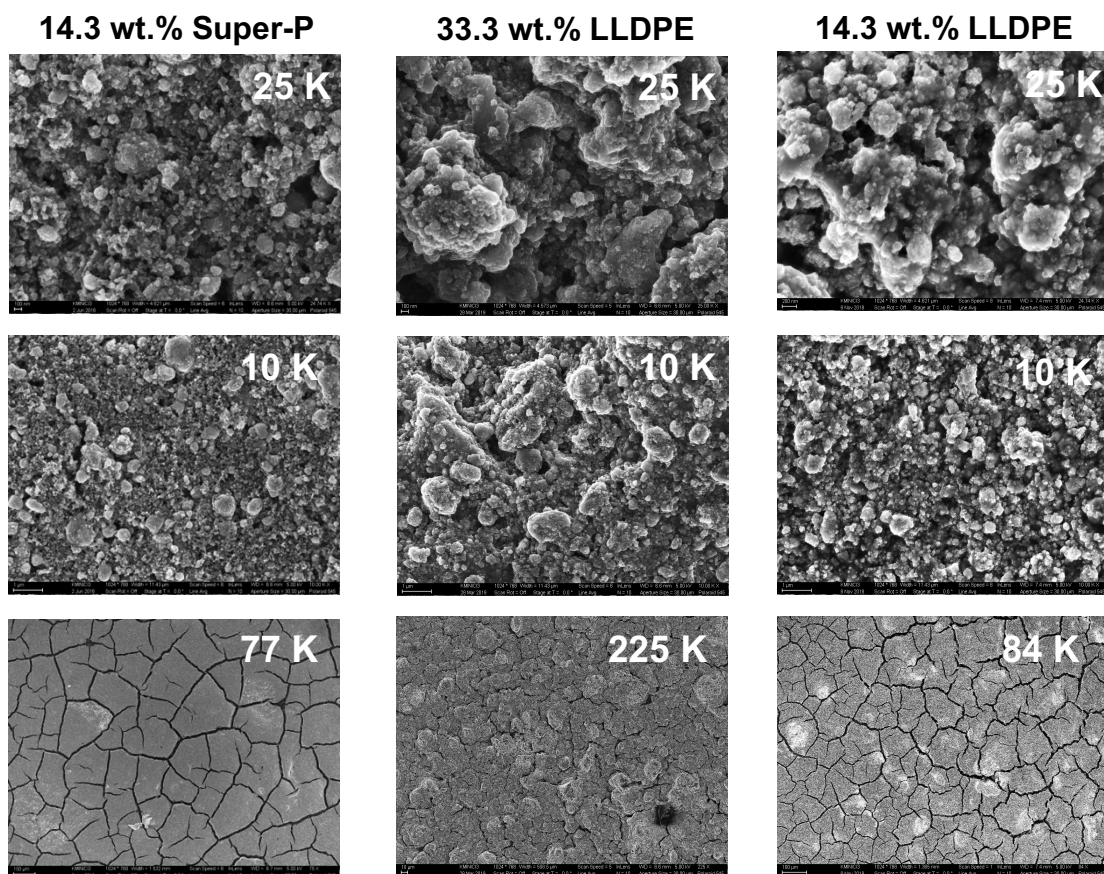
LLDPE has been shown to have higher conductivity than Super-P and similar physical properties.<sup>2</sup> Electrodes were initially prepared with PEG-Fe<sub>3</sub>O<sub>4</sub> as the active material, carbon additives (LLDPE or Super P), and a polymeric binder in a weight ratio of 71.4:14.3:14.3, respectively, to optimize conductivity. Initial electrochemical characterization of the 14.3 wt.% LLDPE demonstrated low capacity retention and poor rate capability performance, which can be seen in Figure 3. To understand differences between Super-P and LLDPE, a simple version of conductivity testing was investigated to confirm that the correct carbon loading of LLDPE was being used in the electrode. A higher carbon loading could potentially improve performance if conductivity was worse than that of Super-P. Conductivity samples were prepared at 0.2, 0.5, and 0.6 ratios of carbon to polymer for both Super-P and LLDPE to assess for any major differences. Conductivity was determined using a 4-point probe and profilometry measurements. The results are shown in Table 1. Super-P was found to be orders of magnitude higher in conductivity than LLDPE. This provided evidence for increasing the carbon loading LLDPE in the electrode.

**Table 1:** Conductivity Measurements for LLDPE and Super-P samples

LLDPE	Conductivity (S/m)
0.2	0.000513457959
0.5	0.0687140184
0.6	0.03748727598
Super-P	
0.2	3.094359365
0.5	5.058061453
0.6	9.496802078

Composite morphology was evaluated through SEM and Figure 3 compares the 14.3 wt.% LLDPE electrode, 33.3 wt.% LLDPE electrode, and 14.3 wt.% Super-P electrode. SEM images of the 14.3 wt.% LLDPE showed large amounts of cracking on the surface and a higher fraction of

large aggregate clumps of material on the electrode surface. The initial poor electrochemical performance was partially attributed to the poor morphology. SEM imaging of the higher weight percent carbon showed a much better morphology, with fewer large aggregates and little cracking on the surface of the electrode. When compared to images of 14.3 wt.% Super-P, both LLDPE samples have larger aggregates. This is most easily seen in the 25K magnification image. However, overall the 33.3 wt.% LLDPE shows a more ideal morphology over the 14.3 wt.% LLDPE.

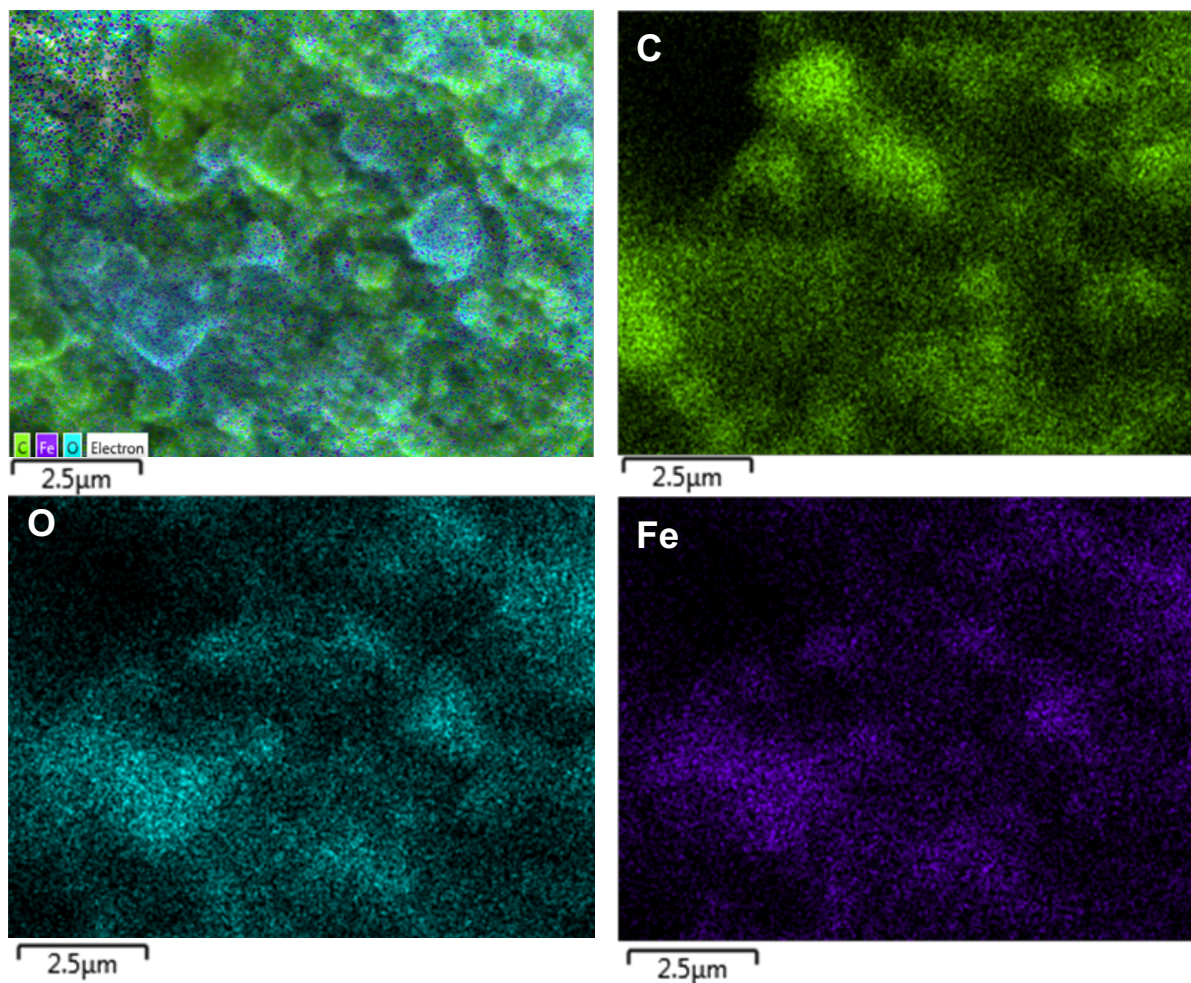


**Figure 3:** SEM images of electrode surfaces

Energy Dispersive X-Ray Spectroscopy (EDX) was used in conjunction with the SEM analysis to characterize the elemental composition of the 33.3 wt.% LLDPE electrode and determine the level of materials dispersion. The results are shown in Figure 4, with the expected



elements of carbon, oxygen, and iron, appearing in the highest magnitude.<sup>14</sup> The iron and oxygen are more heavily concentrated in small aggregates of active material, as expected, and can be seen

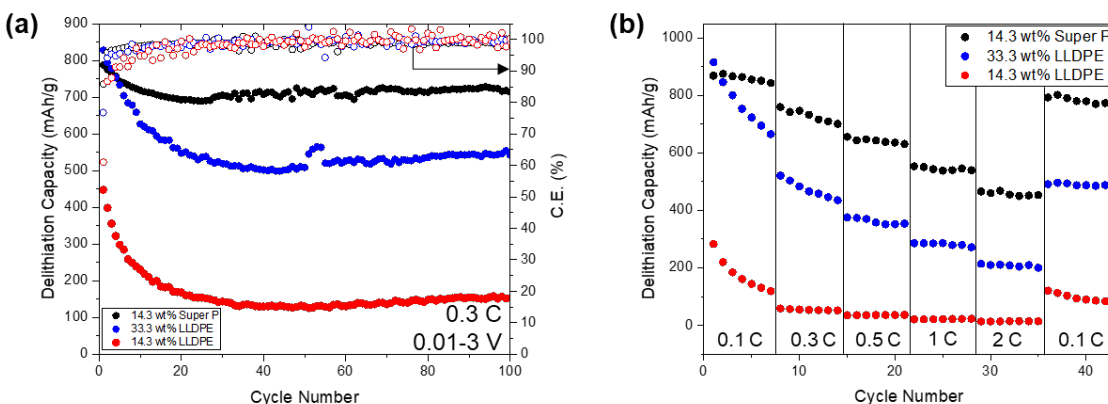


more clearly in the survey image. EDX SEM imaging and elemental (Fe, O, C) image mapping confirmed the presence of  $\text{Fe}_3\text{O}_4$  nanoparticles, carbon additives, and PPBT binder.

**Figure 4:** EDX SEM of 33.3 wt.% LLDPE electrode with elemental mapping of C, O and Fe.

Based on the aforementioned conductivity measurements, the carbon loading was increased to 33.3 wt.%, while the active material stayed at 55.6 wt.% and the polymeric binder was at 11.1 wt.%. Resulting cycling and rate capability data (Figure 5) was much improved and shows promise for use of LLDPE as a conducting agent in batteries. As shown below, the jump in

delithiation capacity for the coin cell made with 33.3 wt.% carbon vs 14.3 wt.% carbon is large. The 33.3 wt.% carbon still does not reach the prime performance of Super-P, the industry standard, but is within a workable range for Li-ion batteries. The coulombic efficiencies of all samples are within the appropriate range of 90-99%. The starting coulombic efficiency is observed to be proportional to the starting delithiation capacity. Repeatability over two to three separate coin cells is demonstrated in Table 2. For every metric of electrochemical testing, 33 wt.% LLDPE performs much better than 14.3 wt.% LLDPE but does not have as of a high performance as 14.3 wt.% Super-P.



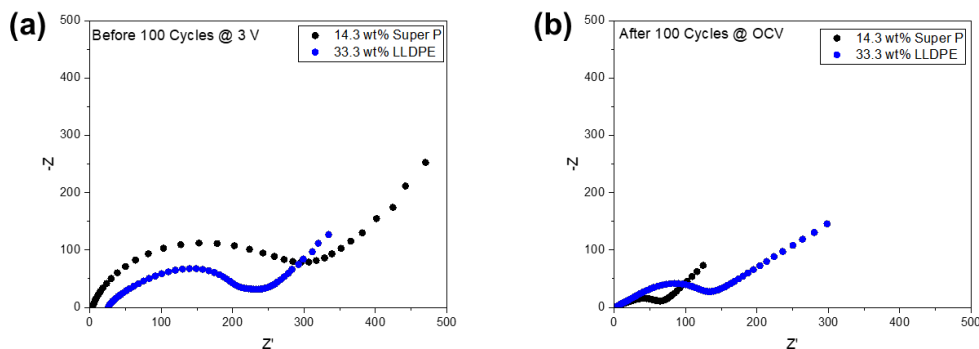
**Figure 5:** (a) Cycling performance (capacity retention as a function of cycle number) comparing Super P to varied LLDPE loadings at 240 mA g<sup>-1</sup> (~0.3 C) between 0.01 and 3 V. (b) Delithiation rate capability, where cells are lithiated at a constant current density of 80 mA g<sup>-1</sup> (0.1 C) and delithiated at different current densities between 0.01 and 3 V.



**Table 2.** Composite anodes comparing Super P to varied LLDPE loadings (averaged over 2-3 coin cells)

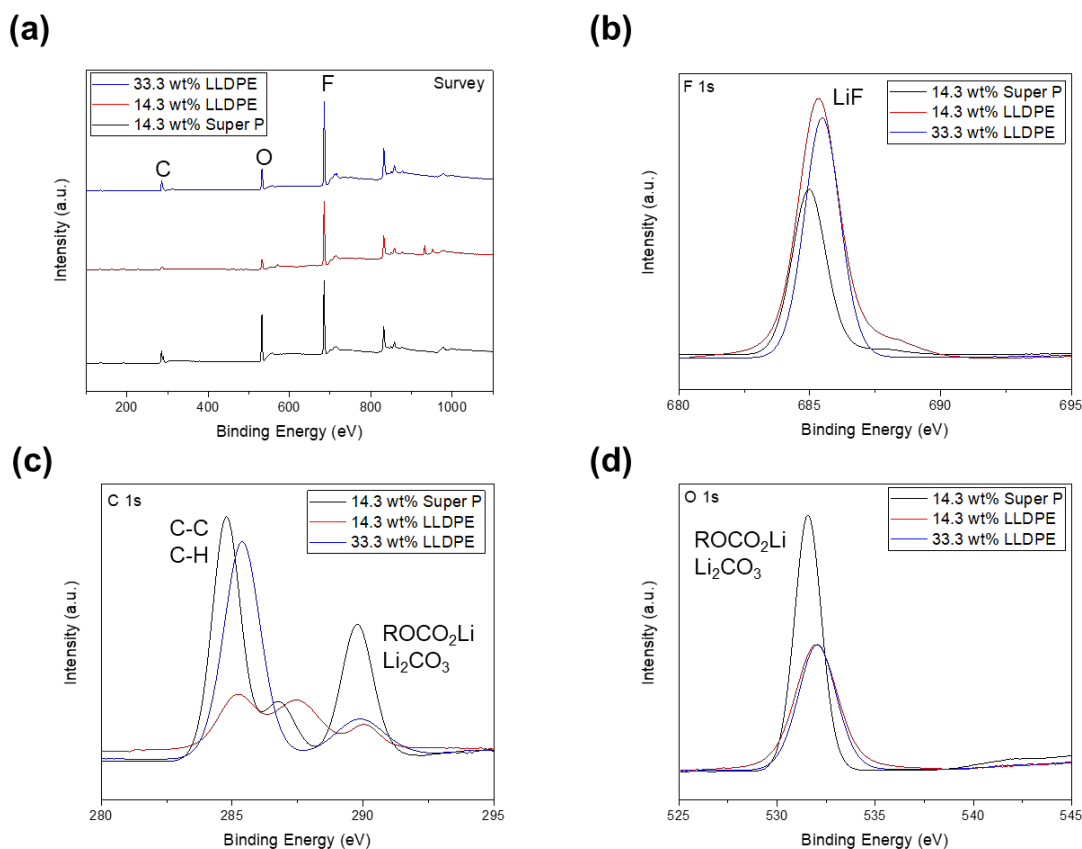
<b>Anode System</b>	<b>Initial Delithiation Capacity (mAh/g)</b>	<b>100<sup>th</sup> Delithiation Capacity (mAh/g)</b>	<b>Initial C.E. (%)</b>	<b>100<sup>th</sup> C.E. (%)</b>	<b>Capacity Retention (%)</b>
14.3 wt.% Super P	826 ± 40	739 ± 43	85.6 ± 0.4	99.1 ± 0.3	89.4 ± 2.4
33.3 wt.% LLDPE	731 ± 85	517 ± 22	76.0 ± 2.1	99.4 ± 1.3	71.1 ± 5.1
14.3 wt.% LLDPE	447 ± 32	154 ± 10	61.0 ± 0.7	100.2 ± 1.1	34.3 ± 3.5

Electrochemical Impedence Spectroscopy (EIS) can provide information on the charge transfer resistances of different materials. The cells used here correspond to those cycled at 0.3 C between 0.01 and 3V (Figure 6a). The below Nyquist plots (Figure 6) show EIS data gathered before and after cycling. The area under the semi-circle plotted correlates to the charge transfer resistance with a smaller semi-circle indicating reduced charge transfer resistance and higher expected performance. Before cycling, the coin cell containing 33.3 wt.% LLDPE had lower charge transfer resistance, which correlates well with a higher initial delithiation capacity. After cycling, the trends reversed and the coin cell containing 14.3 wt.% Super-P had lower charge transfer resistance. This is consistent with the above cycling data as Super-P has better capacity retention over 100 cycles than LLDPE.



**Figure 6:** Impedance spectra measured **(a)** at 3V before cycling and **(b)** at open-circuit voltage (OCV) after 100 cycles in the frequency range from 0.1 MHz to 0.1 Hz.

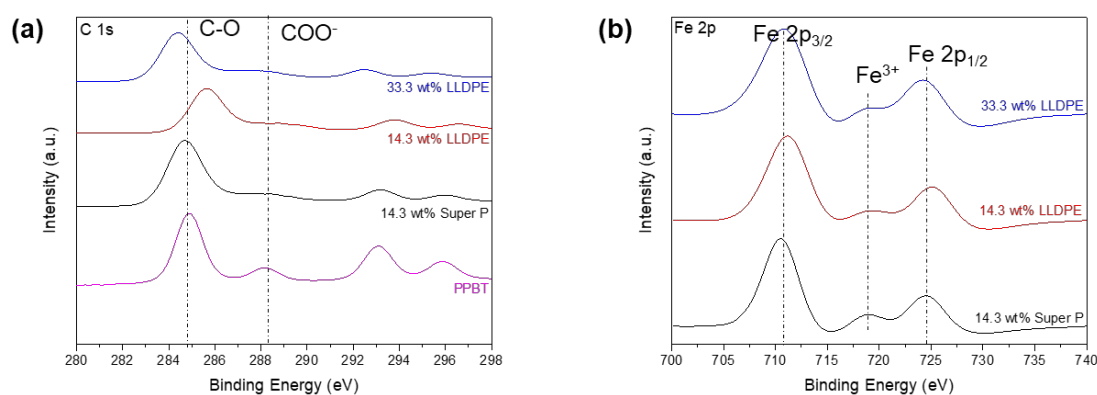
X-Ray photoelectron spectroscopy (XPS) spectra after 100 cycles (Figure 7) revealed information regarding SEI layer formation. The survey scan (Figure 7a) illustrates that only C, O and F remain on the electrode surface after cycling. The F1s scan (Figure 7b) reveals information regarding the stability of the SEI layer through the LiF peak<sup>9,10</sup>. The highest intensity peak correlates with SEI layer formation. Interestingly, the LLDPE electrodes have a higher intensity peak when compared to the Super P electrode. Although the Super P electrode demonstrates more stable electrochemical performance, the information from the LiF peak could indicate that the LLDPE positively impacts SEI layer formation and a higher carbon loading is needed for enhanced electrochemical performance. Further evidence of a stable SEI layer is derived from C 1s and O 1s data (Figure 7c, d), where a higher proportion of organic compounds is desired. Here, we see that Super P has a significantly higher organic fraction when compared to both LLDPE systems, which would confirm its stable electrochemical performance.



**Figure 7:** XPS analysis of SEI layer after 100 cycles: **(a)** Survey scan, **(b)** F 1s scan, **(c)** C 1s scan, and **(d)** O 1s scan.

X-Ray Photoelectron Spectroscopy (XPS) provides information about composition of the surface layers of a material by sending X-rays to the surface and measuring the energy of photoelectrons emitted. The binding energy and intensity of each peak provides information on the composition and chemical states of the compounds present.<sup>13</sup> The below XPS data (Figure 8) was gathered on a 33.3 wt.% LLDPE slurry, 14.3 wt.% LLDPE slurry, 14.3 wt.% Super-P slurry, and PPBT polymeric binder. The XPS spectra of the C 1s core level (Figure 8a) peaks show that the same carbon bonds are present in each slurry and carbon prior to any cycling. The peaks and relative intensity are almost identical for each slurry. The PPBT has a higher intensity COO<sup>-</sup> peak than the slurry's indicating that the slurry compositions have a higher fraction of C-O bonds. The

XPS spectra of the Fe 2p core level (Figure 8b) provides preliminary information on chemical interactions present in the various carbon systems. The peaks at  $\sim 712$  and  $\sim 724$  eV correspond to Fe  $2p_{3/2}$  and Fe  $2p_{1/2}$  orbitals, respectively. The absence of the middle peak, associated with Fe<sup>3+</sup>, confirms a pure magnetite phase.<sup>12</sup> The presence of this satellite peak illustrates interactions between polymeric binder and active material surface, as demonstrated in all electrode systems, regardless of carbon used.



**Figure 8:** (a) C 1s high resolution XPS spectra and (b) Fe 2p high resolution XPS spectra of electrode slurries and PPBT polymeric binder.

### Conclusions

Fabrication of LLDPE presented an exciting opportunity to integrate a recycled carbon into the electrode of a Li-ion battery in the form of a conductive agent. Previous testing on the carbon showed high conductivity and optimal properties rivaling that of Super-P, the current industry standard. PEG coated magnetite was used as the active material in the electrode, and PPBT was used as the polymeric binder. TGA testing showed that the PEG-Fe<sub>3</sub>O<sub>4</sub> system is about 13.5 wt.% PEG.

Initial testing of LLDPE in electrodes using the same weight fractions as Super-P yielded poor results. Low capacity retention and rate capabilities was consistent with images of a cracked surface and large aggregates in the electrode. Conductivity measurements to compare the properties of LLDPE and Super-P showed that a higher carbon loading may be required for LLDPE electrodes. Increasing the weight percent from 14.3 wt.% to 33.3 wt.% led to drastically improved electrochemical performance. Results for initial delithiation capacity and columbic efficiency, 100<sup>th</sup> cycle delithiation capacity and columbic efficiency, and capacity retention were much larger than that of 14.3 wt.% LLDPE. Performance was still below that of Super-P, but within a workable range for Li-ion batteries.

SEM analysis on 14.3 wt.% Super-P, 14.3 wt.% LLDPE, and 33.3 wt.% LLDPE was consistent with electrochemical results. SEM imaging of the higher weight percent LLDPE carbon showed a much better morphology, with fewer large aggregates and little cracking on the surface of the electrode. However, when compared to images of 14.3 wt.% Super-P, both LLDPE samples have larger aggregates. EDX SEM imaging and elemental (Fe, O, C) image mapping confirmed the presence of Fe<sub>3</sub>O<sub>4</sub> nanoparticles, carbon additives, and PPBT binder.

XPS after 100 cycles showed that although Super -P electrode demonstrated a more stable electrochemical performance, LLDPE electrodes have a higher intensity LiF peak which correlates to a more stable SEI layer. When looking at C 1s and O 1s data, Super P has a significantly higher organic fraction when compared to the LLDPE systems which backs up its stronger electrochemical performance. XPS on electrode slurries show that each electrode has a pure magnetite phase. The presence of satellite peaks confirm interactions between the polymeric binder and active material surface, regardless of carbon used. Additionally, spectra of the C 1s core level shows that the same carbon bonds are present in each slurry and carbon prior to cycling.

EIS testing provided information on the charge transfer resistances of the different materials. Nyquist plots indicated that 33.3 wt.% LLDPE had a lower charge transfer resistance before cycling, which correlates to a higher initial delithation capacity. After cycling trends were reversed and 14.3 wt.% Super-P had a lower charge transfer resistance. This matches the electrochemical data gathered as Super-P has better capacity retention over 100 cycles than LLDPE.

Overall, use of LLDPE in Li-ion batteries has been shown to work in a half-cell assembly. The performance of LLDPE does not beat the current industry standard, Super-P, but demonstrates promise for use after further optimization and analysis. Future studies will experiment with the optimal carbon loading in electrode fabrication and attempt to better understand how LLDPE differs from Super-P when electrochemically tested.

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