

Investigation of the Species Responsible for the Dramatic Capacity Fade Improvement Caused by the Addition of Fullerene Soot to Li-ion Anodes

Robert E. Doe, Louis J. Rendek, Jr.,
and Michael J. Wagner
Department of Chemistry
The George Washington University
Washington, DC 20052

Recently we have shown that the addition of arc-generated fullerene soot can dramatically decrease the capacity fade observed in lithium-ion battery negative electrodes upon extended cycling. Fullerene soot is a complex mixture containing thousands of species. This talk will focus on our investigations of which of these species are actively responsible for the improved performance observed.

Our active materials were prepared by blending graphite with arc generated fullerene soot, either raw or extracted, by means of ball milling. In some cases these were further heat treated prior to use in electrodes. The fullerene soot was separated into component species predominantly by means of soxhlet extraction with various organic solvents. Electrodes were formulated using 95% carbon and 5% PVDF binder in a slurry with *n*-methyl-pyrrolidinone (NMP). The working electrode material was coated onto copper current collectors in a custom doctor blade setup. The NMP was evaporated and the working electrodes dried in a vacuum oven at 130°C for 1 hour. After cooling, the electrodes were pressed at 5,000 lbs, compacting them to a final thickness of between 70 and 150 μm .

Charge/discharge cycling was done inside an argon filled glove-box, using custom made, three-electrode, spring-loaded glass "T-cells". Lithium metal coated on stainless steel rods was used for both the counter and reference electrodes. Ethylene carbonate (EC) / dimethyl carbonate (DMC) (2:1 v/v ratio) was employed as the non-aqueous solvent with 1M LiPF_6 as the supporting electrolyte. The cycling regime consisted of loading (intercalation) of the graphite electrode with lithium at a constant current of 0.71 mA/cm^2 to a cut-off voltage of 20 mV vs. lithium metal, holding at 20 mV for 3 h, resting for 1 h, unloading (de-intercalation) at constant current (also 0.71 mA/cm^2) and resting for 1 h prior to the subsequent cycle. Considering the average electrode mass (~6 mg), a rate of approximately C/2 was used for both loading and unloading.

While some components of fullerene soot are responsible for the stabilization of anode capacity observed, others may have no effect or even a negative influence. For instance, the capacity fade improvement is seen regardless of whether the anodes contain low mass fullerenes C_n , where $n < 100$ (Figure 1). Electrodes made by blending these low mass fullerenes with graphite, without inclusion of the higher mass components of fullerene soot, show capacity fade that is identical to pure SFG-15 graphite electrodes. Thus, these lighter fullerenes are inert additives, have little effect on electrode performance other than diminishing specific capacity though the addition of inactive mass. The results of our investigations of which species in the fullerene soot are responsible for capacity fade stabilization of graphitic lithium-ion negative electrodes will be presented.

Figure 1. Reversible capacity for electrodes composed of SFG-15 graphite (crosses) and 80/20 blended SFG-15 graphite with arc fullerene soot either "raw" (triangles) or with fullerenes $\text{C}_{<100}$ removed (circles).

