Structural and Electrochemical Characterizations of Carbon Electrodes using Pure and Lithium Ion Containing Ionic Liquids

Thomas E. Sutto[1,2*], Doug M. Fox[1], Paul C. Trulove[1,3],
and Hugh C. De Long[1,3]

[1]Naval Research Laboratory, Code 6170, Chemistry
Division, Washington D. C., 20375

[2]Chemistry Department, US Naval Academy, Annapolis
MD 21402

[3]Air Force Office of Scientific Research, 4015 Wilson
Blvd., Rm 713,
Arlington, VA 22203-1954

The development of ionic liquid based electrolytes[1] and polymer gel electrolytes[2] for use in Li-ion batteries necessitates the use of either pure metallic lithium as the anode or a carbon based anode, such as amorphous carbon or graphite. Previous work has already shown that the use of the $AlCl_4$ anion, one of the more common counter anions in the ionic liquids, results in the formation of a Li-Al alloy along the graphite-electrolyte surface, rather than true intercalation.[3] Therefore, the ionic liquids studied for use in Li-ion batteries have moved to incorporate those which use fluorinated anions, such as BF_4^-, PF_6^- and bis-trifluoromethanesulfonylimide ($TFSI^-$).

Ionic liquids themselves, because they are composed of discreet cations and anions unlike the more common organic carbonate electrolytes, can potentially intercalate and deintercalate graphite, thus leading to secondary reactions in addition to the intercalation and deintercalation of lithium.[4,5] Therefore, using several different ionic liquids based upon the tri-substituted imidazolium cations 1,2-dimethyl-3-n-propylimidazolium and 1,2-dimethyl-3-n-butylimidazolium, liquid battery cells were assembled in which the ionic liquid with 1.0 M lithium present served as the electrolyte, and solid state batteries in which the electrolyte was an 80% 1.0 molar lithium containing ionic liquid and 20%$ PVdF-HFP polymer gel were assembled. Two electrode cells, in which the anode was lithium metal and the cathode was graphite or a high surface area amorphous carbon, were charged and discharged to determine the efficiencies, as well as the structural changes which occur upon intercalation by either the pure ionic liquid, or by lithium. Results indicate that for graphitic types of electrodes, the Li and imidazolium cations cointercalate graphite (Figure 1), while only Li ion intercalation is observed for amorphous carbon. Also, no structural modification is observed for attempts to intercalate the anions into amorphous carbon, while the TFSI anion especially resulted in the formation of well defined staged graphite intercalates (Figure 2).

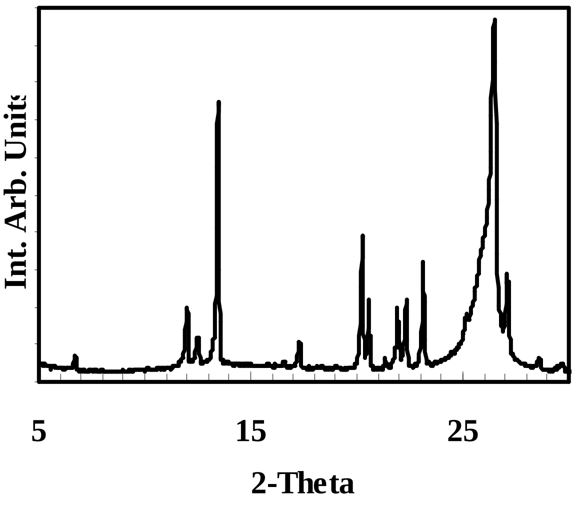

Figure 1. Staged Li-ion 1,2-dimethyl-3-n-butylimidazolium graphite intercalate.

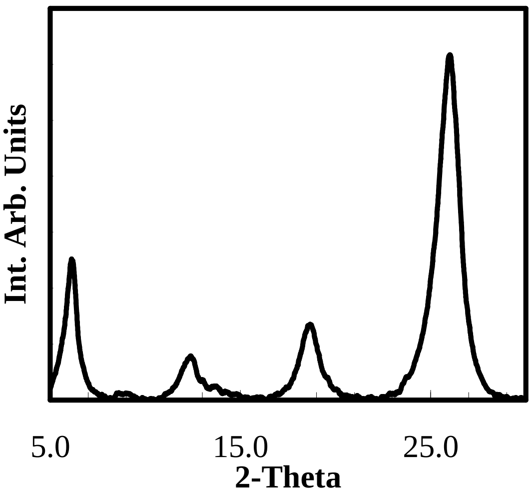

Figure 2. 1st stage $(TFSI^-)_x$Graphite intercalate.

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