

Unraveling the Origin of Failure Mechanism in Graphite-Rich Silicon/Graphite Composite Electrodes with Poly(vinylidene fluoride) (PVDF) Binder

Xiuyun Zhao^a, Svetlana Niketic^a, Chae-Ho Yim^a, Jigang Zhou^b, Jian Wang^b, Yaser Abu-Lebdeh^{a*}

^a National Research Council Canada, Ottawa, ON, K1A 0R6, Canada

^b Canadian Light Source Inc., Saskatoon, S7N 2V3, Canada

E-mail: Xiuyun.Zhao@nrc-cnrc.gc.ca

Graphite-rich Si/graphite composites have the potential to improve the practical energy density of lithium ion batteries. [1] Our research group at the National Research Council Canada has screened a number of natural or synthetic polymer binders for the design and optimization of this composite electrode in the past few years. During this process, we were surprised to find that when the graphite-rich Si/graphite composite electrode is used with conventional PVDF binder it loses most of its capacity very quickly and after 5 cycles the capacity is even lower than that of graphite-only electrode and the differential curves of the electrode show no peaks characteristic of Li ions electrochemically intercalating into graphite or alloying with silicon.

Herein, we shed light on the failure mechanism by using Scanning Electron Microscope (SEM), energy dispersive spectroscopy (EDS), X-ray photoelectron spectroscopy (XPS), and X-ray photoemission electron microscopy (X-PEEM) [2] methods respectively. It is found that this capacity loss could be attributed to passivation of both silicon and graphite particles: in addition to conventional degradation of silicon, some graphite particles in the composite lose their connection with carbon black and PVDF binder that is caused by the expansion and contraction of neighboring silicon particles. Surprisingly, it is also found that the chemical decomposition of PVDF binder takes place even in single electrodes (Si only or graphite only) despite the excellent battery performance of graphite-only electrode. Both EDS and X-PEEM results show that this may be due to the uniform dispersion of both PVDF binder and decomposition products (e.g. LiF) in cycled graphite-only electrode. This study shows that the most suitable binder for the composite electrode is a polymer with good chemical interaction with both graphite and silicon.

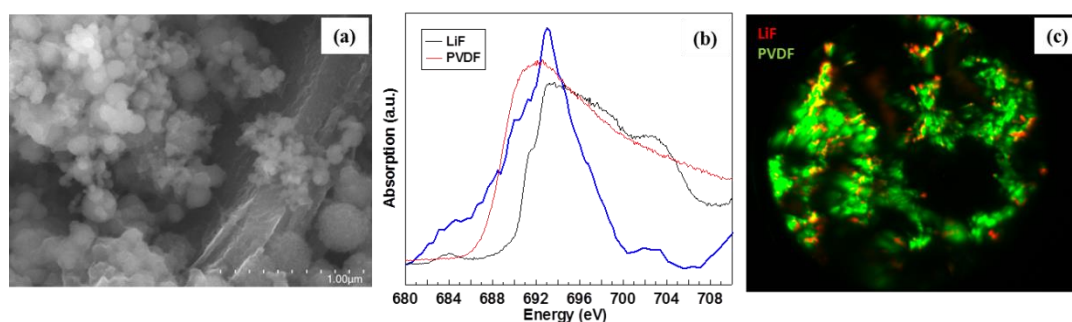


Figure 1. (a) SEM image, (b) fluorine K-edge X-ray absorption near edge structure (XANES) spectra, and (c) fluorine chemical mapping of cycled graphite-rich Si/graphite composite electrode.

References:

- [1] C.H. Yim, S. Niketic, N. Salem, O. Naboka, Y. Abu-Lebdeh, J. Electrochem. Soc., 164 (2017), A6294–A6302.
- [2] J. Zhou, J. Wang, Y. Hu, M. Lu, ACS Appl. Mater. Interfaces, 9 (2017), 39336–39341.