IN-SITU NEUTRON DIFFRACTION STUDIES OF SONY 18650 LITHIUM-ION CELLS

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Abstract:
In-situ measurements yield the best information about the function of battery materials because the data is collected on an actual functioning cell as it undergoes charge/discharge. We, as well as other groups, have recognized the utility of these in-situ measurements, and have studied a wide variety of materials in experimental cell designs.[1-3] We were interested in extending these in-situ structural studies by using neutron diffraction because 1) this technique can be performed on full cells encased in metal canisters, i.e. commercially available cells, and 2) the stronger relative scattering of neutrons by lithium makes neutron diffraction a good probe for lithium occupancy in the structure.

A Sony 18650 cell was placed on the High Intensity Powder Diffraction (HIPD) neutron spectrometer at Los Alamos Neutron Scattering Center (LANSCE) for data collection in the uncharged state. Data was collected overnight to obtain the best possible pattern. Next, the cell was charged up to ~4.1 V and allowed to equilibrate (~ 1 hour) before a second spectra was collected (again overnight for the best possible signal). The resultant diffraction signal from the $LiCoO_2$ inside the battery was very weak and somewhat swamped by the iron signal from the material making up the outside canister. However, it was possible to locate the diffraction peaks of $LiCoO_2$ in the spectra, as well as to refine the data to obtain lattice parameters for the material in the fully charged and discharged state. Table 1 shown below summarizes these results.

Table 1. Refined lattice parameters from Neutron diffraction spectra.

Sample	$LiCoO_2$ a-axis (Å)	$LiCoO_2$ c-axis (Å)	$LiCoO_2$ volume (Å^3)	Aluminum a-axis (Å)
Ex-situ cathode	2.8135(4)	14.240(4)	97.62(3)	-
In-situ uncharged	2.8120(13)	14.253(14)	97.60(10)	4.0525(2)
In-situ charged	2.8090(12)	14.332(12)	97.94(9)	4.0527(2)

This in-situ experiment demonstrates the feasibility of investigating commercial cells via neutron diffraction. Clear structural changes were observed between the charged and uncharged states of the cathode. However, due to the very small peak intensities for the $LiCoO_2$, it was not possible to refine the lithium site occupancy from these spectra. It may be possible to boost the cathode signal by replacing the canister material of the battery. If this were to be performed, it must be done in the least intrusive way as to maintain similar performance from the battery. Several possible alternatives are under consideration. Another necessity is to reduce the data collection time so as to make real-time experiments possible. It is possible that modification of the cell canister, coupled with experiments on the new Hi Pressure Preferred Orientation (HIPPO) spectrometer may improve count rates sufficiently to monitor cells under typical charge cycles.

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