Unravelling the behaviour of layered $x \operatorname{Li}_2\operatorname{MnO}_3 \cdot (1-x) \operatorname{LiMO}_2$ high capacity cathode material through correlated electrochemistry and characterization of single particles

<u>Dean J. Miller¹</u>, Christian Proff¹, J.G. Wen¹, Lynn Trahey² ¹ Electron Microscopy Center

¹ Electron Microscopy Center ² Chemical Sciences and Engineering Division Argonne National Laboratory, Argonne, IL 60439 (USA)

The layered oxide cathode material x $Li_2MnO_3 \cdot (1-x) LiMO_2$ is of great interest due to its potential for high capacity. However, long term performance is limited by voltage fade - a progressive reduction in voltage as a function of cycle. It is widely presumed that voltage fade is a result of structural evolution of the composite layered structure to a "spinel-like" structure. However, identifying a clear correlation between voltage fade and any such structural change is complicated by the fact that working batteries and test cells consist of ensembles of millions of individual particles. Thus most measurements of performance provide average properties that are difficult to correlate with structural characterization on a much smaller length scale. We approach this challenge by carrying out electrochemical cycling on single particles upon which detailed characterization can be carried out before and after various periods of cycling. The electrochemical cycling data of single particles show the same characteristic signatures of voltage fade those observed in measurement of coin cells, confirming the validity of these measurements. By using a measurement platform compatible with a variety of characterization approaches, we are able to characterize the same particle subjected to cycling, providing an improved opportunity to correlative changes in structure with changes in electrochemical performance. This presentation will detail our approach and the insights gained from these correlated measurements.

*Research sponsored by the U.S. DOE Vehicle Technologies Program and by the U.S. DOE, Office of Science – Basic Energy Sciences under contract DE-AC02-06CH11357. The Electron Microscopy Center at Argonne is supported by the Office of Science – Basic Energy Sciences.