Abstract Submitted
for the MAR14 Meeting of
The American Physical Society

First-principles analysis of phase stability in layered-layered composite cathodes for lithium-ion batteries[1] HAKIM IDDIR, ROY BENEDEK, Argonne National Laboratory, VOLTAGE FADE TEAM — The atomic order in layered-layered composites with composition xLi2MnO3·(1-x)LiCoO2 is investigated with first-principles calculations at the GGA+U level. This material, and others in its class, are often regarded as solid solutions, however, only a minute solubility of Li2MnO3 in a LiCoO2 host is predicted. Calculations of Co-vacancy formation and migration energies in LiCoO2 are presented, to elucidate the rate of vacancy-mediated ordering in the transition-metal-layer, and thus determine whether low vacancy mobility could result in slow equilibration. The Co-vacancy formation energy can be predicted only to within a range, because of uncertainty in the chemical potentials. Predicted migration energies, however, are approximately 1 eV, small enough to be consistent with rapid ordering in the transition metal layer, and therefore separated Li2MnO3 and LiCoO2 phases. The relatively small (of the order of a few nm) Li2MnO3 domain sizes observed with TEM in some xLi2MnO3·(1-x)LiMO2 composites may result from other factors, such as coherency strain, which perhaps block further domain coarsening in these materials.

[1] Office of Science of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

Hakim Iddir
Argonne National Laboratory

Date submitted: 13 Dec 2013 Electronic form version 1.4