Computational modeling of the layer-spinel interface properties for cathode materials in lithium ion batteries

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The research on Li-excess materials has been intense as this family of the materials can deliver 250mAh/g capacity routinely and has a voltage around 4.2V. With the high energy density resulting from the excellent capacity and voltage, Li-excess materials are considered to be promising candidates for HEV and EV battery applications.[1] However, the rate capability and cycling performance of this family of materials is not adequate due to the surface phase transformation during cycling.[2] To overcome this problem, layer-spinel composite materials have been proposed recently to get both high energy density and power density.[3] Only by understanding the interface properties, can we successfully optimize Li-excess material as well as the layer-spinel composite.

In our study, first principles calculations are carried out in the generalized gradient approximation with Hubbard U correction (GGA+U) to the Density Functional Theory (DFT) to study the surface and interface properties of layer and spinel phases. Coherent interfaces based on the lattice parameters and our experimental observations are investigated to study the stabilization mechanism of spinel forming on layered surface. Figure 1 shows the computational model of {110} spinel on {010} layered phase. From our preliminary results, it shows that the surface energy for the spinel-layer model (summation of spinel surface and spinel-layer interface energy) is lower than the surface energy of layer surface. The charge transfer at the interface is the main driving force for the stabilization. Further interfacial properties including transition metal mobility as well as oxygen vacancy formation will be discussed.

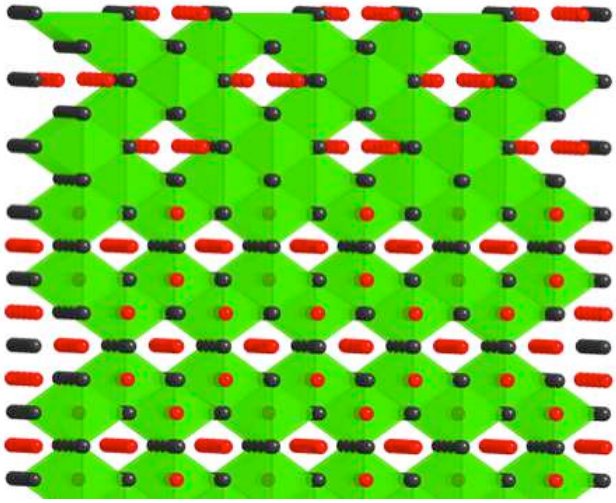

Figure 1. Computational model for the interface calculation with spinel on layer phase. Green octahedral: TM; Red: Li; Black: O. Do not show unpublished data.

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Reference

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