Tackling Oxygen Defects in Lithium Titanium Oxide (LTO) using First Principle Simulation Methods

Matthias Kick¹, Cristina Grosu¹,², Arobendo Mondal¹, Harald Oberhofer¹, Josef Granwehr² and Christoph Scheurer¹

¹TU München, Lehrstuhl für Theoretische Chemie, Lichtenbergstr. 4, 85748 Garching
²FZ Jülich IEK-9, Ostring O10, 52425 Jülich

Motivation
Lithium titanium oxide (LTO) is an intriguing anode material promising particularly long lived batteries, due to its remarkable phase stability during (dis)charging of the cell. However, its usage is limited by its low intrinsic electronic conductivity. Introducing oxygen vacancies can be one method to overcome this drawback, possibly by altering the charge carrier transport mechanism. Getting detailed insight in its defect chemistry is a non trivial task which needs to be tackled by theory and experiment. Here, we present extensive theoretical two component density functional theory (TCDFT) calculations in order to interpret positron annihilation measurements used to reveal defect influences on Li mobility. In addition we used Hubbard corrected DFT (DFT+U) calculations to gauge electron mobility to shed a light on the experimentally observed increase in electronic conductivity. By explicitly calculating hopping barrier heights our simulations indeed show that a polaron hopping mechanism can be the source for the increased electronic conductivity.

Experimental and Theoretical background: Application to LTO

How to produce defects in LTO?

Positron annihilation spectroscopy

DFT computation of positron lifetimes

Results

Polaron Hopping in LTO

Acknowledgements:

References:


We are using Hubbard corrected DFT to simulate possible polaron hopping in order to calculate the theoretical increase in LTO’s electronic conductivity.

Standard semi-local DFT is not able to capture the physics of localized polarons as the large amount of self-interaction error (SIE) leads to over-delocalization of the electron density.

PBE+U (U = 2.65 eV)

Hopping barrier height: 186 meV indicates polaron hopping already at room temperature.

Top view: Only nearest neighbor oxygen atoms relax during hopping. Indeed, this indicates a small polaron hopping mechanism.

Member of the Helmholtz Association