

# Enhanced Lithium Transport in Epitaxial Thin Films for Lithium-ion Batteries

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Lithium-ion batteries are the most popular rechargeable batteries nowadays, as they have become the main power source for many applications, such as portable electronics, power tools, and hybrid/full electric vehicles. None of the current rechargeable batteries can fully satisfy all the challenging requirements for our current energy storage. Although tremendous research effort has been devoted to investigate the electrochemical performance of a wide variety of active lithium-based materials, current rechargeable batteries exhibit energy density, lifetime and safety still far below their theoretical capabilities.

Essential for all high-performance energy applications are processes that happen at the interfaces between the different components. Key problems include slow electrode process kinetics with high polarization and low ionic diffusion or electronic conductivity, particularly at the electrode-electrolyte interfaces. Epitaxial engineering is used to control the crystal orientation of electrode thin films, which enables a unique insight into the relation between electrochemistry and crystal directionality of such chemically complex inorganic interfaces, not obtainable in single crystals or polycrystalline samples.

In this lecture I will show the lithium diffusion behavior in  $\text{LiMn}_2\text{O}_4$  cathode, and  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  anode, thin films, which are epitaxially grown by pulsed laser deposition on single crystalline Nb-doped  $\text{SrTiO}_3$  substrates. Control over the specific crystal orientation of the full thin film enables detailed analysis of the lithium diffusion along specific crystal planes ( $\{001\}$ ,  $\{110\}$  and  $\{111\}$ ). Single phase films show enhanced cyclability and faster charging speed, as compared to studies on polycrystalline materials. The achieved capacity reached >90% of the theoretical limit and minimal capacity reduction was observed when measured over 1000 cycles.

- T.A. Hendriks, D.M. Cunha, D.P. Singh, M. Huijben, 'Enhanced Lithium Transport by Control of Crystal Orientation in Spinel  $\text{LiMn}_2\text{O}_4$  Thin Film Cathodes', ACS Applied Energy Materials 1, 7046–7051 (2018).
- D.M. Cunha, T.A. Hendriks, A. Vasileiadis, C.M. Vos, T. Verhallen, D.P. Singh, M. Wagemaker, M. Huijben, 'Doubling Reversible Capacities in Epitaxial  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  Thin Film Anodes for Microbatteries', ACS Applied Energy Materials, DOI: 10.1021/acsaem.9b00217 (2019).