

## 3D Structuring of Thin Film Solid State Batteries for High Power Demanding Applications

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**Abstract :** High energy and power density are the main requirements of today's high demanding applications in consumer electronics. Lithium ion batteries (LIB) have the highest energy density of all known systems and are thus the best choice for rechargeable micro-batteries. Liquid electrolyte LIBs present limitations in safety, size and design, thus thin film all-solid state batteries are predominantly considered to overcome these restrictions in small devices. Although planar all-solid state thin film LIBs are at present commercially available they have low capacity ( $<1\text{mAh/cm}^2$ ) which limits their application scenario. By using micro-or nanostructured surfaces (i.e. 3D batteries) and appropriate conformal coating technology (i.e. electrochemical deposition, ALD) the capacity can be increased while still keeping a high rate performance. The main challenges in the introduction of solid-state LIBs are low ionic conductance and limited cycle life time due to mechanical stress and shearing interfaces. Novel materials and innovative nanostructures have to be explored in order to overcome these limitations. Thin film 3D compatible materials need to provide with the necessary requirements for functional and viable thin-film stacks. Thin film electrodes offer shorter Li-diffusion paths and high gravimetric and volumetric energy densities which allow them to be used at ultra-fast charging rates while keeping their complete capacities. Thin film electrolytes with intrinsically high ion conductivity ( $\sim 10^{-3}$  S.cm) do exist, but are not electrochemically stable. On the other hand, electronically insulating electrolytes with a large electrochemical window and good chemical stability are known, but typically have intrinsically low ionic conductivities ( $< 10^{-6}$  S cm). In addition, there is the need for conformal deposition techniques which can offer pinhole-free coverage over large surface areas with large aspect ratio features for electrode, electrolyte and buffer layers. To tackle the scaling of electrodes and the conformal deposition requirements on future 3D batteries we study  $\text{LiMn}_2\text{O}_4$  (LMO) and  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  (LTO). These materials are among the most interesting electrode candidates for thin film batteries offering low cost, low toxicity, high voltage and high capacity. LMO and LTO are considered 3D compatible materials since they can be prepared through conformal deposition techniques. Here, we show the scaling effects on rate performance and cycle stability of thin film cathode layers of LMO created by RF-sputtering. Planar LMO thin films below 100 nm have been electrochemically characterized. The thinnest films show the highest volumetric capacity and the best cycling stability. The increased stability of the films below 50 nm allows cycling in both the 4 and 3V potential region, resulting in a high volumetric capacity of  $1.2\text{Ah/cm}^3$ . Also, the creation of LTO anode layers through a post-lithiation process of  $\text{TiO}_2$  is demonstrated here. Planar LTO thin films below 100 nm have been electrochemically characterized. A 70 nm film retains 85% of its original capacity after 100 (dis)charging cycles at 10C. These layers can be implemented into a high aspect ratio structures. IMEC develops high aspect Si pillars arrays which is the base for the advance of 3D thin film all-solid state batteries of future technologies.

**Keywords :** Li-ion rechargeable batteries, thin film, nanostructures, rate performance, 3D batteries, all-solid state

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