Final Report

Project acronym: *PLARASBAT*

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Publishable project summary

The idea of building a Li - ASSB based on a the novel architecture of large area planar Sheet - like battery is based on the possibility to produce micro structured electrodes (MEE) that can be sintered on the surface of a thick film solid electrolyte (SE). The sintered MEE in principle should keep the electronic and ionic connection within its ceramic volume. Herewith the sintered interface between the MEE and SE should not present interdifusion such as it prevents the ionic conduction through it. To produce the battery different building blocks must be prepared and assembled in a way that the ionic connection along Anode/electrolyte/Cathode structure keeps its continuity to giverise to the electrochemical chain. The ionic wire must be continuous along the two electrode/ electrolyte interfaces and as short as possible to reduce the internal resistance of the battery. The materials for electrodes (anode and cathode) and electrolyte were studied and selected, novel methods to produce slurries based on the preparation of sol-gel solutions of the electrode compounds were tested as well as different methods for the deposition (tape casting, screen printing, spin casting, robotic deposition system with syringe) and sintering to produce the materials (thick films, interdigital micro-electrodes IDE- MEE) with clean interfaces to give rise to desired battery architecture.
The base of the proposed battery is a large area (50x50 mm^2) thick film (100- 200 μm thick) solid electrolyte. After an exhaustive study of Lithium lanthanum Titanate perovskites and Li-NASICON compositions (including different powder preparation procedures, RMN, DRX, Broadband impedance and PFG-NMR ^{7}Li diffusion studies) the $Li_{1.3}Al_{0.3}Ti_{1.7}(PO_4)_3$ Li-NASICON (LATP) solid electrolyte was selected. Both self-supported (SSP) and supported in α-Al_2O_3 (SP) thick films were prepared with success (high Li conductivity and flatness) by tape casting and screen printing of optimized slurries. For electrodes; $Li_4Ti_5O_{12}$ (LTO) anode, $LiCoO_3$ (LCO) and $LiFePO_4$ - LATP composite cathodes were studied. Sol-gel method combined with commercial powder was used to formulate slurries for deposition of the electrodes. The strong interaction between LCO, LTO and LATP electrolyte made impossible to obtain clean interfaces even in the mildest sintering conditions. LFP-LATP composite showed electrochemical activity after sintering onto LATP thick film. For assembling the Li- ASSB battery SP LATP thick film was selected as it presented the best mechanical properties. One cell battery made with the IDE electrode with dimensions: 30 pairs of LTO anode and LCO or LFP-LATP composite cathode with fingers 200 m wide and 30-40 m thick separated by 250 m with an overlap of 18mm. The electrodes were deposited by a robotic deposition method using syringes filled with the corresponding optimized slurry. After sintering, using the two step schedule, current collectors made by covering the IDE electrodes with first, a layer of C paint and then a second layer of Ag paint to improve electronic conductivity were deposited with the deposition robot. The CC was sintered at 200ºC. This cell planar architecture of ASSB was not electrochemically active. Wetting the ASSB with a liquid electrolyte activated the ionic response indicating that as expected the ionic wire was broken in sintered interfaces and it is mandatory to repair it using an electrolyte that can be processed in liquid form.