Press Release M13

The project: The SOLiD project aims to create a sustainable and cost-efficient pilot scale manufacturing process for a high energy density, safe and easily recyclable solid-state Li-metal battery (LMB). A dual-layer polymer design I selected for Roll-to-roll (R2R) production. A dry extrusion process will be developed to produce the positive electrode composite layer, the latter including the positive electrode material (NMC811), the Solid Polymer Electrolyte (SPE) and conductive additives. The so-produced layer will then be coated by slot-die to yield a separator SPE layer. A protected, thin-film Li-metal anode is produced by PVD. This cell design will be long-lasting and safe due to the presence of the solid electrolyte and protective layers.

Materials for Lithium and NMC811 protection

For formulation of initial specifications for the SOLiD project, and within the process of related experiments planning, a literature search was conducted among the reports on lithium-ion battery electrode coating with various materials. Based on the extracted data, the screening of protective layer coatings for anode (***metallic lithium***) and cathode (***NMC811***) materials produced by atomic layer deposition (**ALD**) was made to achieve the **Milestone 9**. These coatings are needed to improve electrochemical performance, increase capacity, and extend lifespan of the batteries under development. In turn, the **ALD** method was chosen as it is ideal for precise, low-temperature, nanoscale coating due to its self-limiting process, ensuring excellent coverage and conformal deposition. As a result, the most effective and, thus, the highest priority coating compositions for further experiments were selected.

For coating **lithium metal anodes**, aluminum oxide was the first and most widely used material. It improves electrochemical performance, increases capacity, and extends lifespan. Similar to ALD, MLD is a versatile technique that allows to deposit pure polymeric or hybrid thin films at lower temperatures. The most popular MLD coating is alucone, which is also the only metallic lithium coating material, reported previously as efficient in solid-state batteries. After assessing the materials' performance in symmetric cells and the technical feasibility of coating approaches for selected materials, it was decided to focus solely on binary materials, the deposition methods of which are currently under development.

For powder ALD coating of **NMC811 cathode active material** the titanium oxide based materials were chosen due to existing expertise of consortium members to implement the corresponding coating processes. At the same time, niobium-based materials were chosen as lithium niobium oxide was found as only reported NMC811 coating material studied in solid state batteries.

The initial specifications for the materials intended to use in SOLiD batteries, their processing approaches, as well as further corrections have been collaboratively agreed upon by the consortium.

Polymer electrolyte materials

The targets are to develop SPEs that provide high-voltage, safe and easily recyclable Li-metal batteries (LMBs), validate these electrolytic materials through electrochemical and structural characterisations and optimise the technological solutions for processability. The binder SPE (BSPE), is integrated during the positive electrode extrusion, therefore it needs to be extrudable, thermoplastic and with low glass transition temperature (Tg). The separator SPE (SSPE) is coated on the positive electrode composite, therefore, it must have low viscosity, low molar mass and can be cross-linked. The types of carbonates considered for both SPEs, are aliphatic main-chain polycarbonates and polymers with cyclic carbonate side groups.

After further testing, it became evident that there was a requirement to explore alternative methods to boost ionic conductivity and to make the polar group more mobile. The design of new monomers in ongoing as well as the exploration of new co-polymer formulations.

Achievements - milestone

Over the past year, research has been conducted on particle detection using Laser Speckle Photometry (LSP) and dark-field light scattering. LSP proved to be extremely sensitive to environmental conditions and was unreliable for particle identification. On the other hand, dark-field scattering proved to be effective, revealing particles appearing larger in images than their actual size, thereby reducing the required camera resolution for detecting small particles. Additionally, laboratory validation was carried out to demonstrate the feasibility of Electrochemical Impedance Spectroscopy (EIS)-inline inspection, focusing on small-scale samples manufactured in-house. The validation itself involved producing different SPE samples and correlating their ionic conductivity with the impedance spectra of the positive electrode composite. This strong correlation confirms that EIS measurements conducted on the cathode composite, are directly connected to fundamental material properties like ionic conductivity, enabling real-time monitoring during cathode composite production. Furthermore, optical and X-ray measurements were used to detect various kinds of defects and characterise material thickness and internal material distribution inconsistencies. Preliminary spectral measurements were inconclusive, but they should be re-examined when final materials are available. So far, the measurement principles have been tested on reference materials and further development and validation are necessary when real samples become available. Moreover, a camera-based approach for meniscus monitoring was explored, with estimated resolution capabilities for spatial and depth directions of 0,9 µm and 1,55 µm respectively, indicating feasibility based on the target used for resolution estimation.

For more information about the SOLiD project, please, visit: www.thesolidproject.eu

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