High-performing, sustainable and safe Li-based batteries: Another world is possible?

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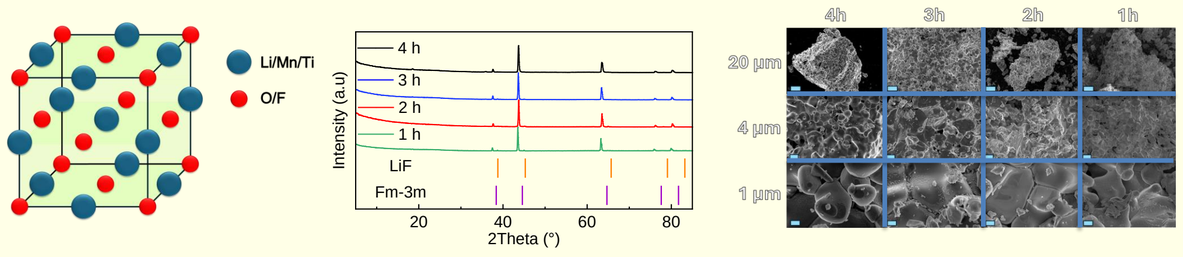
Energy and power density of battery cells for automotive applications are required to be higher and higher to satisfy user demands and compete with conventional vehicles in terms of driving range and cost. On the other hand, European regulation asks for more sustainable batteries over all the value chain. It is not easy to address all the key-points contemporarily [1]. High-voltage cathodes often show limited compatibility with conventional electrolytes, while high-Ni materials enhance capacity at the expense of safety. The use od solid-state electrolytes also contributes to battery safety. To increase sustainability, cathode production should be entirely based on water, which is already applied for anodes, as well as the introduction of bio-based polymer separators could be important for recycling. This communication presents the work carried out by researchers and PhD students within several collaborative projects with industrial partners.

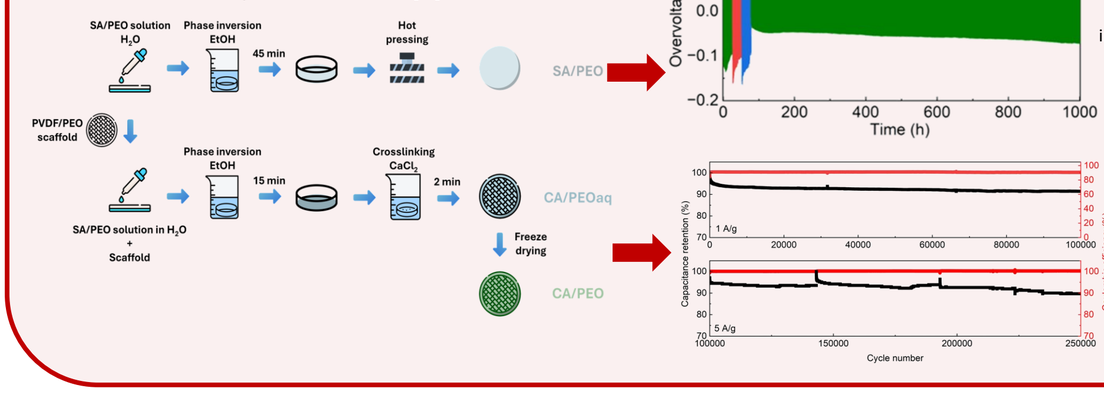
Cathodes: Li-excess disordered rocksalts

In **disordered rocksalt**, lithium and transition-metal cations are randomly distributed, enhancing lithium-ion transport. **Co-free** compositions reduce costs and ethical concerns.
Solid-state synthesis is a well-established and widely used method for preparing new compounds. XRD and SEM are used to evaluate the outcome of the synthesis.



Separators: Alginate-based separators

Sodium alginate and **polyethylene oxide** are used for the preparation of a bio-based separator for electrochemical power devices [4].



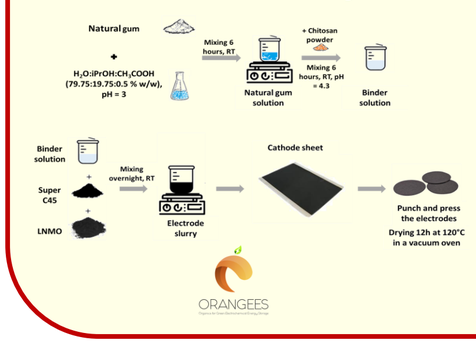
Li//Li
in organic solvent

AC//AC
aqueous
supercapacitor

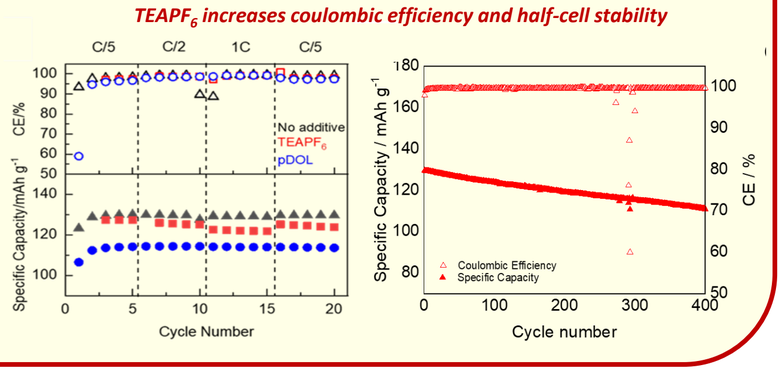
$LiNi_{0.5}Mn_{1.5}O_4$ from aqueous slurry and chitosan-based binder

Development of sustainable high-performance Li-ion cells using Co-free LNMO cathodes with chitosan binders, aqueous processing, and optimized electrolytes with additives to suppress Li dendrites and stabilize interfaces [2, 3].

Green Processing



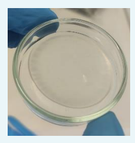
$TEAPF_6$ increases coulombic efficiency and half-cell stability



Blend polymer solid-state electrolytes

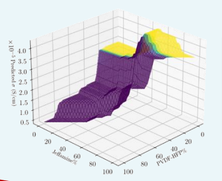
By blending two polymers with complementary properties, i.e. Jeffamine® and PVDF-HFP, into a single solid-state electrolyte and **optimizing their composition**, we achieved a balance of **high ionic conductivity** and **mechanical robustness**, enabling safer and efficient lithium metal batteries [5].

Jeffamine : PVDF-HFP 1:1



Robust but **low σ_{ion}**

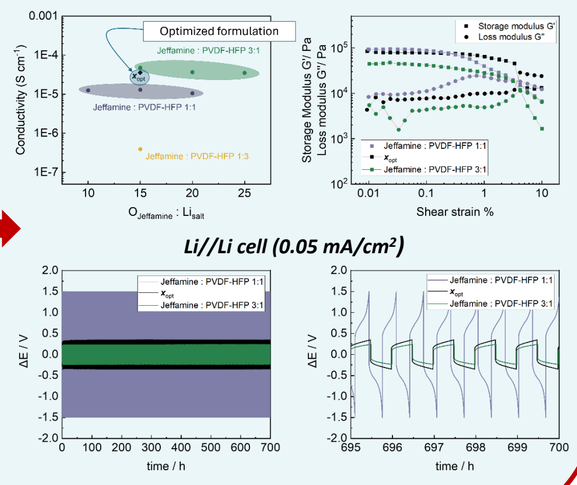
Modeling mechanical feasibility with a **classifier** and σ_{ion} with **machine learning algorithms**



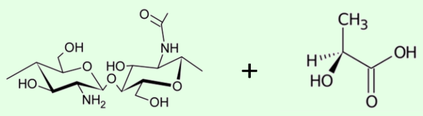
Jeffamine : PVDF-HFP 3:1

High σ_{ion} but **weak** and **cracked** membrane

Optimization of σ_{ion} with a **mechanical constraint**



Anodes: Chitosan-based binder for graphite anodes

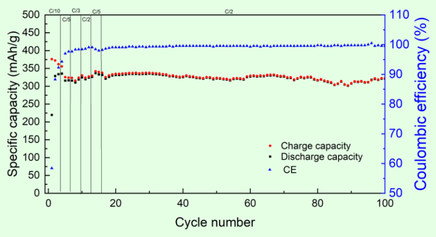


Chitosan + 5% wt lactic acid solution

Binders with high ionic conductivity for fully sustainable Li-ion cells

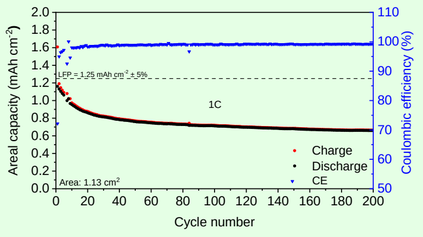
WE: Graphite : Binder : C45 (90:7:3) AM: 2.3 mg/cm²
CE: Li metal, LP30 + 2% VC + 10% FEC

HALF CELL
CE: >99%
CR: 99%



WE: Graphite : Binder : C45 (90:7:3), AM: 4.0 mg/cm²
CE: LFP (AM: 7.3 mg/cm²), LP30 + 2% VC + 10% FEC

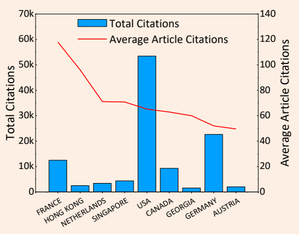
FULL CELL
CE: >99%
CR: 72%



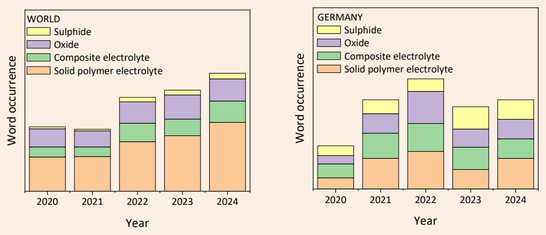
Solid state batteries: Market analysis and battery control tools

In the emerging field of SSB research, a detailed market analysis highlights the major players contributing to the advancements of solid-state electrolytes (SSEs).

$$TC = \sum_{N_{articles}} citations \qquad AAC = \frac{\sum citations}{N_{articles}}$$



The families of SSEs received different attention worldwide, which is also related to the special lab-tools required for their testing. Local trends, such as in Germany, has a more balanced study distribution among the different SSE chemistry.



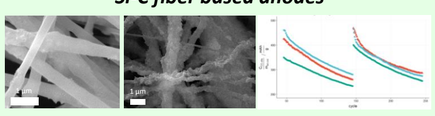
Understanding the reactivity in air of sulphide-based SSE to favour their adoption in EV industry is of foundamental importance.
LPSCl (infiltrated in non-woven scaffold) prepared at Fraunhofer ISE [6] (Freiburg) was tested. Its suitability to be scale up in mild dry room conditions was assessed through mass spectrometry analysis under different humidity level.

MARPOSS

Silicon-anodes and upscaling

Lab-scale

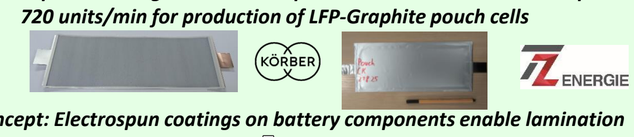
Si-C fiber based anodes



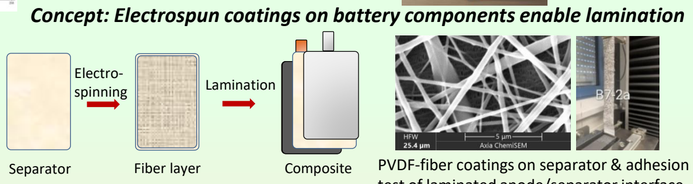
- Carbonization of polyacrylnitrile fibers with 25 w% Si content → flexible current collector-free anodes
- Stable performance in coin LFP full-cells test (~ 0.4 mAh/g) with a CE > 99% and ~300 mAh/g electrode mass @ 1C cycling,

Industrial-scale

High-speed process using lamination to produce monocell-stacks with up to 720 units/min for production of LFP-Graphite pouch cells



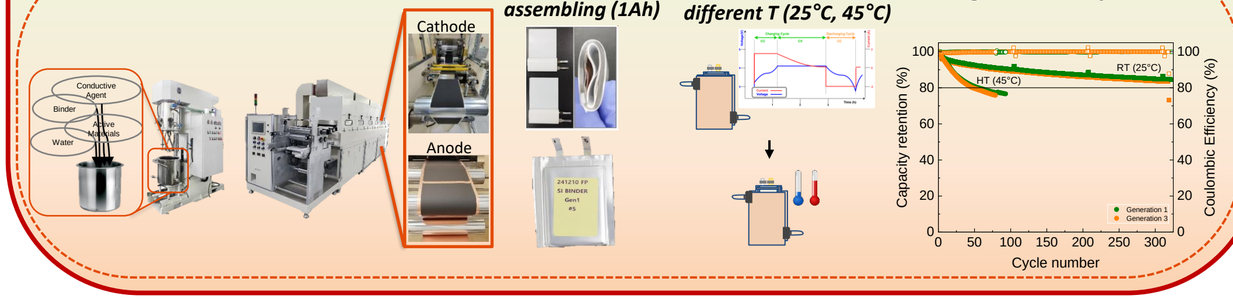
Concept: Electrospun coatings on battery components enable lamination



PVDF-fiber coatings on separator & adhesion test of laminated anode/separator interface.

Pilot-scale

Planetary Mixing ***Coating & Drying*** ***Pouch cells assembling (1Ah)*** ***Electrochemical tests at different T (25°C, 45°C)*** ***Long-term stability at 1C***



Conclusions

Producing **high-power batteries**, which are also **safe and sustainable**, is possible. This can be achieved through the selection of **Co-free cathodes** and **Si–graphite** anodes, and by using **bio-based polymer binders** that enable sustainable, water-based electrode processing. In addition, **solid electrolytes** play an important role in battery safety, specifically those with Li metal anodes. The cost of SSBs is higher than that of LIBs, partly due to their lower production volume and the greater complexity of their manufacturing processes. Nevertheless, EVs equipped with SSBs have been announced by several manufacturers and seems to be the future trend. Studies aimed at elucidating interfacial phenomena in **Li metal electrodes**, improving their safety and stability, advancing the understanding of **Li zero-excess configurations** to enhance energy density and sustainability, and **modeling** optimized electrode and electrolyte formulations, together with combined **in situ and operando characterization**, will be essential for developing the next generation of batteries.

Acknowledgements

Finanziato dall'Unione europea NextGenerationEU
Ministero dell'Università e della Ricerca
Italiadoma PIANO NAZIONALE DI RIPRESA E RESILIENZA
NEST NETWORK FOR ENERGY SUSTAINABLE TRANSITION
PRIN 2022 - Funded by EU - Next Generation EU
BIONIC
ORANGEES
INSTM
ENEA Agenzia nazionale per le nuove tecnologie, l'energia e lo sviluppo economico sostenibile
MINISTERO DELL'AMBIENTE E DELLA SICUREZZA ENERGETICA

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