



ECO2LIB Project Newsletter

Dear readers,

Welcome back to the seventh newsletter of the Horizon2020-project ECO2LIB. In this issue, we want to give you an overview of the project activities in the third reporting period of the project. In addition, for new readers, we also provide a short summary of the ECO2LIB project and some more information on the project consortium.

I hope you enjoy the newsletter and the activities we are doing!



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[ECO²LIB Project](#)

Summary of the ECO2LIB project

After the successful EU-project Sintbat, ECO2LIB aims to continue the effort by focusing on a new KPI, the cycle related costs per energy: €/kWh/cycle. This KPI very well reflects the real need of the customers in the energy storage market if a minimum volumetric energy density is added. The research and development activities will be supported by a clear recycling concept and an extended Life Cycle Assessment, to judge the environmental impact of the different options and to choose the best. To show both ECO-aspects (**ECO**logical and **ECON**omical) of our project the acronym ECO²LIB was created.

The consortium decided to continue the improvement of the well-established **Lithium-Ion system** with advanced materials, methods, and corresponding recycling-concept. So, it will be possible to directly exploit the results of ECO²LIB in an IPCEI project, which is under preparation.

Summary of the project progress

With month 42 we are entering the final year of the project. In the past six months, our main focus was on:

Electrodes and Electrolytes: The preparation of Generation 2 21700 cells with improved capacity retention is slightly delayed and thus still ongoing. A lack of available materials was the reason for this delay. To reduce the impact of this delay, material properties and cycling behaviour have already been tested in CoinPower cells. Depending on the used voltage range, the cycle stability can be tuned. With discharge cut-off voltages >2.75 V, the target of 70% capacity remaining at cycle 300 will be achievable. In addition, Generation 3 of the electrolyte material has been completed. It will now be further engineered to allow the integration in pilot cells.

Recycling: The development of an innovative Li-Ion battery recycling process has been completed. The objective was to regain as much material as possible. In the designed process, it is possible to safely shred the batteries and access the inner materials. Unfortunately, some materials have still not been recoverable, but the process will be further optimised in the coming months.

Characterisation: Different in-lab and synchrotron techniques have been applied to investigate the impact of the novel fluorine-free electrolytes on the SEI properties. The three techniques are highly complementary: 1. Operando XRR measurements allowed to obtain the variations of the SEI thickness and mean composition during cycling. 2. Ex situ ssNMR spectroscopy was applied to silicon-graphite composite electrodes to analyse their SEI after cycling in coin cells with fluorinated and fluorine-free electrolytes. 3. Ex situ ToF-SIMS measurements of silicon-graphite composite electrodes allow us to detect and locate the elements composing the SEI after cycling in coin cells with fluorine-free electrolytes.

In parallel, characterisation of the developed electrolytes was performed. To avoid electrolyte degradation, the protocol for formation cycles was modified. However, the electrochemical response was still very similar in both cases. This could suggest that even with a different protocol, the interphases formed are similar and dominating the electrochemical performance or that the formation protocol is not the main problem with this type of electrolyte and other methods to improve its performance are needed. This will be further investigated. Finally, the replacement of the organic solvents with oligomers did not render the desired electrochemical performance and therefore we are working now towards the implementation and characterization of Generation III electrolyte composed of a polymeric matrix instead of oligomers.

Modelling and Simulation: A 3D microscale anode model was realised. More precisely, the development of theory, models and computational tools for in-situ modelling of anode at the microscale under the thermo-chemo-mechanical coupling. This will be used to carry out systematic computational investigations of the relationship between microstructure and chemo-mechanical response of the anode, to gain insights into the effects of various design parameters (e.g. material properties) on induced stresses within active particles and the effective matrix, and the extent of lithiation.

M42 Remote General Assembly Meeting

For the M42 meeting, it was agreed by the consortium to do a remote 1-day meeting. The meeting took place in November 2023.

Publications

Publications in journals

Multi-scale quantification and modeling of aged nanostructured silicon-based composite anodes

Vorauer, T., Kumar, P., Berhaut, C.L. et al., *Commun Chem* 3, 141 (2020)

“Advanced anode material designs utilizing dual phase alloy systems like Si/FeSi₂ nano-composites show great potential to decrease the capacity degrading and improve the cycling capability for Lithium (Li)-ion batteries. Here, we present a multi-scale characterization approach to understand the (de-)lithiation and irreversible volumetric changes of the amorphous silicon (a-Si)/crystalline iron-silicide (c-FeSi₂) nanoscale phase and its evolution due to cycling, as well as their impact on the proximate pore network.” Read more: <https://doi.org/10.1038/s42004-020-00386-x>

Surface Oxidation of Nano-Silicon as a Method for Cycle Life Enhancement of Li-ion Active Materials

Ratynski, M., Hamankiewicz, B., Buchberger, D. A. et al., *Molecules* 2020, 25(18), 4093

“Among the many studied Li-ion active materials, silicon presents the highest specific capacity, however it suffers from a great volume change during lithiation. In this work, we present two methods for the chemical modification of silicon nanoparticles. Both methods change the materials' electrochemical characteristics. The combined XPS and SEM results show that the properties of the generated silicon oxide layer depend on the modification procedure employed.” Read more: <https://doi.org/10.3390/molecules25184093>

The role of coordination strength in solid polymer electrolytes: compositional dependence of transference numbers in the poly(ϵ -caprolactone)–poly(trimethylene carbonate) system

Eriksson, T., Mace, A., Mindemark, J., Brandell, D., *Phys. Chem. Chem. Phys.*, 2021,23

“Both polyesters and polycarbonates have been proposed as alternatives to polyethers as host materials for future polymer electrolytes for solid-state lithium-ion batteries. While being comparatively similar functional groups, the electron density on the coordinating carbonyl oxygen is different, thereby rendering different coordinating strength towards lithium ions. In this study, the transport properties of poly(ϵ -caprolactone) and poly(trimethylene carbonate) as well as random copolymers of systematically varied composition of the two have been investigated, in order to better elucidate the role of the coordination strength.” Read more: <https://doi.org/10.1039/D1CP03929F>

Facile preparation of hierarchical 3D current collector for Li-ion anodes

Ratynski, M., Hamankiewicz, B., Czerwinski, A. *Electrochimica Acta*, 403 (2021)

„Beside great commercial success of Lithium-ion batteries, initiated by Sony in 1991, the cells manufacture details, active material selection, synthesis routes and further modifications are still under intense development. To increase the capacity of the whole cell, the researchers are focusing on new, high capacity, alloy type anode materials such as tin, germanium, aluminum and silicon. Apart from the great capacity, all of the alloy-type materials suffer from large volume changes during lithiation, e.g. silicon volume expansion can reach 300%.” Read more: <https://doi.org/10.1016/j.electacta.2021.139698>

A cut finite-element method for fracture and contact problems in large-deformation solid mechanics

Poluektov, M., Figiel, Ł. *Computer Methods in Applied Mechanics and Engineering*, 388 (2022)

"Cut finite-element methods (CutFEMs) belong to the class of methods that allow boundaries/interfaces to cut through the elements, which avoids any meshing/remeshing problems. This is highly convenient from a practical point of view, especially when non-stationary interfaces are considered, e.g. phase boundaries in solids, as the interfaces can move independently of the mesh. There are many research directions related to CutFEM, one of which focuses on the equations of solid mechanics." Read more:

<https://doi.org/10.1016/j.cma.2021.114234>

Carbonyl-Containing Solid Polymer Electrolyte Host Materials: Conduction and Coordination in Polyketone, Polyester, and Polycarbonate Systems

Eriksson, T., Gudla, H., Manabe, Y., et al. *Macromolecules* (2022)

"Research on solid polymer electrolytes (SPEs) is now moving beyond the realm of polyethers that have dominated the field for several decades. A promising alternative group of candidates for SPE host materials is carbonyl-containing polymers. In this work, SPE properties of three different types of carbonyl-coordinating polymers are compared: polycarbonates, polyesters, and polyketones. The investigated polymers were chosen to be as structurally similar as possible, with only the functional group being different, thereby giving direct insights into the role of the noncoordinating main-chain oxygens." Read more:

<https://doi.org/10.1021/acs.macromol.2c01683>

A Multiscale, Correlative, Air Free Workflow for the Analysis of Li Distribution in Batteries via ToF-SIMS

Kelly, S.T., White, R., Tordoff, B. et al. *Microscopy and Microanalysis*, Volume 28, Issue S1, 1 August 2022

"The ability to link the macro and nano world has long been a challenge due to correlation and sample preparation difficulties. X-Ray microscopy is widely accepted as the standard for non-destructive interior tomography imaging of large objects with high resolution. The ability to access these interior objects for further analysis has previously been impossible due to sample damage and lack of precision in correlation when manually cutting the sample. Moreover, this technique requires careful chemical handling for systems of interest such as intact batteries which may have undergone cycling. The adaptation of a FIB-SEM with a femtosecond laser in a dedicated separate chamber has made this workflow feasible." Read more:

<https://doi.org/10.1017/S1431927622003841>

Synchrotron Holotomography on Silicon-Based Anode Materials for Improved Lithium Ion Batteries

Chamasemani, F.F., Häusler, M., Vorauer, T., et al. *Microscopy and Microanalysis*, Volume 28, Issue S1, August 2022

"Lithium ion batteries are an essential part of our society. Certainly, there is a strong demand for higher energy densities with respect to future applications in energy storage and e-mobility. The use of silicon-based anode materials is considered as a promising approach due to its high theoretical capacity. The major drawback that comes along using silicon as an anode material is its huge volumetric expansion of up to 300% during lithiation, which induces mechanical stresses on the material. These stresses result in the formation of cracks and further pulverization of the Si as well as causing the formation of an in-homogeneously growing solid electrolyte interphase (SEI)." Read more:

<https://doi.org/10.1017/S1431927622001672>

Impact of solid-electrolyte interphase reformation on capacity loss in silicon-based lithium-ion batteries

Vorauer, T., Schöggel, J., Sanadhyan, S.G., et al. *Communications Materials*, 4, 44 (2023)

"High-density silicon composite anodes show large volume changes upon charging/discharging triggering the reformation of the solid electrolyte interface (SEI), an interface initially formed at the silicon surface. The question remains how the reformation process and accompanied material evolution, in particular for industrial up-scalable cells, impacts cell performance. Here, we develop a correlated workflow incorporating X-ray microscopy, field-emission scanning electron microscopy tomography, elemental imaging and deep learning-based microstructure quantification suitable to witness the structural and chemical progression of the silicon and SEI reformation upon cycling. The nanometer-sized SEI layer evolves into a micron-sized silicon electrolyte composite structure at prolonged cycles. Experimental-informed electrochemical modelling endorses an underutilisation of the active material due to the silicon electrolyte composite growth affecting the capacity." Read more:

<https://doi.org/10.1038/s43246-023-00368-1>

Role of Filler Content and Morphology in LLZO/PEO Membranes

Mehraj Ud Din, M., Häusler, M., Fischer, S.M. et al. *Front. Energy Res.*, 12 October 2021

"Polymer electrolytes containing Li-ion conducting fillers are among the extensively investigated materials for the development of solid-state Li metal batteries. The practical realization of these electrolytes is, however, impeded by their low Li-ion conductivity, which is related to the filler and the interplay between the filler and the polymer. Therefore, we performed an in-depth analysis on the influence of the filler content (0, 10, and 20 wt%) and filler morphology (particles and nanowires) on the electrical and electrochemical properties of the PEO-based composite electrolyte using a wide spectrum of characterization techniques, such

as 3D micro-X-ray computed tomography, cross-sectional scanning electron microscopy, X-ray diffraction, and differential scanning calorimetry, impedance spectroscopy, and galvanostatic cycling. The studies reveal that the filler materials are well distributed within the membranes, without any indications for the formation of agglomerates.” Read more: <https://doi.org/10.3389/fenrg.2021.711610>

Articles

Ageing of nanostructured silicon-based composite anodes: Morphology changes and inhomogeneous lithiation

Brunner, R. (2020)

“Alloy systems like Si/FeSi nano-composites have great potential as stable anode materials in Li-ion batteries, but their characterization at different scales and throughout their ageing remains challenging due their complex architecture.” Read more: <https://go.nature.com/3IWmRuC>

Innovatives Materialdesign für hocheffiziente Energiespeicher [German]

Brunner, R. (2020)

“Ohne effiziente Energiespeicher und einhergehende Kosteneffizienz wird es keine Energiewende geben. Wie kann Energie möglichst effizient und über eine Vielzahl von Lade- und Entladezyklen gespeichert werden?” Read more: <https://www.just-magazin.com/innovatives-materialdesign-fuer-hocheffiziente-energiespeicher/>

Improving the Design of Anode Materials in Lithium Ion Batteries

Interview with R. Brunner on the ZEISS Blog

“Researchers use advanced imaging methods to understand the structure-property relationship

Dr. Roland Brunner is a Group Leader for Material and Damage Analytics in the Microelectronics Division at the Materials Center Leoben (MCL) in Austria. The group strongly focuses on 3D nano/micro-structure image- based characterization and analysis with respect to innovative materials used in microelectronics and energy, to trigger improved functional material design for industrial applications.” Read more: <https://blogs.zeiss.com/microscopy/en/fesem-anode-materials/>

Elektrolyt utan giftigt fluor ger elbilsbatterier samma prestanda

Kristensson, J. (2020)

Article on the development of fluorine-free electrolytes at Uppsala University

<https://www.nyteknik.se/batterier-premium/elektrolyt-utan-giftigt-fluor-ger-elbilsbatterier-samma-prestanda/1177032>

Conferences, seminars, lectures

Date	Presentation title	Event	Speaker
26.05.2020	Operando synchrotron experiments and porous-electrode modeling: a combined approach. Case study: sequential lithiation mechanisms in Silicon-graphite blended anodes.	Battery 2030+ workshop	S. Lyonnard (CEA)
03.09.2020	Elimination of Fluorination: The Influence of Fluorine-Free Electrolytes on the Performance of Si-based Li-ion Batteries	ISE meeting	G. Hernández (Uppsala)
15.10.2020	Monitoring Li-ion batteries by advanced operando neutron techniques	ILL-ESS user meeting satellite workshop	S. Lyonnard (CEA)
11.2020	Elimination of Fluorination: The Influence of Fluorine-Free Electrolytes on the Performance of Si-based Li-ion Batteries	2020 Virtual MRS Spring/Fall Meeting & Exhibit	G. Hernández (Uppsala)
03.12.2020	Ecologically and Economically viable Production and Recycling of Lithium Ion Batteries	H2020 Low TRL Smart Grids and Storage Projects clustering event	B. Achzet (VARTA Storage)
25.01.2021	Akkutechnologien / Methoden der Materialcharakterisierung speziell im Bereich Energiespeicherung	Workshop Technical High School Leoben	R. Brunner (MCL)
03.2021	Improvement of the Cell Performance: Si/graphite coin cell interactive workshop to develop DOI for characterization workflow	Seminar Workshop with pupils from the Technical Highschool Leoben (HTL)	R. Brunner (MCL)
25.-29.07.2021	CutFEM approach for handling non-stationary interfaces in large-deformation solid mechanics: Application to fracture	16th US National Congress on Computational Mechanics	M. Poluektov, L. Figiel (UoW)

07.2021	Silicon-based Composite Anodes for Li-ion Batteries: Morphology Changes and Ageing	Zeiss Workshop	R. Brunner (MCL)
18-20.08.2021	Multiscale Investigation of an Si-Fe Alloy Anode Material for Storage Applications with Improved Aging Performance	American Association for Advanced Functional Materials	R. Brunner (MCL)
10.2021	Multiscale characterization of advanced Si-based composite anodes	KMM-VIN Workshop	R. Brunner (MCL)
09.12.2021	From Imaging to Knowledge: Towards More Advanced Li-Ion Batteries	Workshop Oslo on Batteries	R. Brunner (MCL)
17.12.2021	Ecologically and Economically viable Production and Recycling of Lithium Ion Batteries	H2020 Battery Projects Online Workshop	N. Bucher (VARTA Microbattery)
WS2021/22	Solar cells lecture	Lecture: Montanuniversität Leoben "Solar Cells" Communication with students Energy relevant topics and storage for PVs.	R. Brunner (MCL)
2021	Operando characterization of battery anodes using mXRD and combined SAXS/WAXS	iUCR, Invited Talk.	S. Lyonnard (CEA)
2021	Operando characterization of silicon-based battery anodes by neutron & synchrotron techniques	MRS, Boston. Invited Talk.	S. Lyonnard (CEA)
04.-06.04.2022	Modelling of chemo-mechanical processes in heterogeneous materials for energy storage	18th European Mechanics of Materials Conference (EMMC18)	M. Poluektov, A. Morozov, A. B. Freidin, L. Figiel (UoW)
11.04.2022	Creating Synergies Through R&D Projects	LiPLANET Expert Group	D. Ott (EurA)
04.05.2022	Multi-Method Characterization Workflow for Advanced Si-based Anodes	14th annual FIB SEM meeting	R. Brunner (MCL)
05.05.2022	Machine Learning Assisted Analysis of Chemical & Micro-Structure Properties in Advanced Si-based anodes	MPPE 2022	T. Vorauer (MCL)
05-09.06.2022	A cut finite-element method for fracture and contact problems in large-deformation solid mechanics	18th European Congress on Computational Methods in Applied Sciences and Engineering (ECCOMAS2022)	M. Poluektov, L. Figiel (UoW)
19.06.2022	Beyond Sweep Voltammetry to Assess Electrochemical Stability of Electrolytes	32nd Topical Meeting of the International Society of Electrochemistry (ISE)	G. Hernández (Uppsala)
28.06.2022	Uncovering the impact of coordination chemistry on cation transport in polymer electrolytes	EPF 2022	J. Mindemark (Uppsala)
28.06.2022	Gel Polymer electrolytes based on Methacrylate End-Capped Poly(trimethylene) carbonate oligomers for lithium batteries	EPF 2022	V. Vijayakumar (Uppsala)
05.07.2022	Safe and Sustainable Electrolytes for Lithium-Based Batteries	POF22 (Power Our Future)	G. Hernández (Uppsala)
31.08.-02.09.2022	Modeling silicon-graphite composite negative electrodes in lithium-ion batteries	8th International Conference on Advanced Computational Methods in Engineering (ACOMEN)	S. G. Sanadhya (UoW)
31.08.-02.09.2022	Multiscale modelling of chemo-mechanical processes in heterogeneous materials for energy storage	8th International Conference on Advanced Computational Methods in Engineering (ACOMEN)	M. Poluektov, L. Figiel (UoW)
19.09 - 21.09.2022	Advanced Imaging and Analysis of Si-Based Li-Ion Cells: Unravel the Microstructure – Material Property Relationship	Battery Days 2022	R. Brunner (MCL)
03.10.2022	Elucidating the Electrochemical Stability of Polymer Electrolytes	ISPE-17	G. Hernández (Uppsala)
WS2022/23	Solar cells lecture	Lecture: Montanuniversität Leoben "Solar Cells"	R. Brunner (MCL)

		Communication with students Energy relevant topics and storage for PVs.	
06.10.2022	To hop or not to hop: How to accurately describe the diversity of ion transport modes in solid polymer electrolytes	ISPE-17	J. Mindemark (Uppsala)
Sept. 2022	Lithiation and aging mechanisms in nanostructured group-IV materials by operando techniques at Large Scale Facilities	ISE (online) meeting	S. Lyonnard (CEA)
01.03.2022	Accelerating battery characterization using neutron & synchrotron techniques: why and how ?	Battery2030+, Excellence Seminar on-line.	S. Lyonnard (CEA)
01.08.2022		ECM33, Versailles. Invited Talk.	S. Lyonnard (CEA)
2023	Bessere Batterien dank Künstlicher Intelligenz	Article Just magazine	R.Brunner (MCL)
2023	Machine learning revealed statistics of material microstructure features and failures for model development and calibration	Impulsvortrag Workshop AVL	R.Brunner (MCL)
2023	Talente: Praktika für Schülerinnen und Schüler 2023	GreenEnergy3	R.Brunner (MCL)
2023	in context to: Commun Mater 4, 44 (2023). https://doi.org/10.1038/s43246-023-00368-1	Nature Community Post	R.Brunner (MCL)
2023	in context to: Commun Mater 4, 44 (2023). https://doi.org/10.1038/s43246-023-00368-1	LinkedIn Post	R.Brunner (MCL)
02.2023	Accelerating and standardizing battery characterization workflows	ESRF Users Meeting, Plenary Talk	S. Lyonnard (CEA)
03.2023	Multimodal characterization of heterogeneities in Li-ion batteries	IBA, Austin, Texas. Invited Talk	S. Lyonnard (CEA)

The project consortium

VARTA Microbattery GmbH (Germany)

VARTA Microbattery (VMB) is an internationally leading and globally active manufacturer of retail and OEM batteries and has been operating for more than 125 years. [Read more](#)

CEA (France)

CEA is a French government-funded technological research organization. With more than 15,000 researchers and co-workers, its activities cover four main areas: Energy, Defence & security, Health & information technologies, and Fundamental research. Two institutes from CEA, both located on the CEA Grenoble centre, are involved in the ECO²LIB project. [Read more](#)

Warwick Manufacturing Group (UK)

WMG is a world leading research and education group, transforming organisations and driving innovation through a unique combination of collaborative research and development, and pioneering education programmes. [Read more](#)

VARTA Innovation GmbH (Austria)

VARTA Innovation GmbH (VI), with registered office in Graz, was founded in 2009 as a joint venture between VARTA Microbattery and Graz University of Technology. Within VARTA Innovation both, the industrial fabrication know-how from VARTA Microbattery and the basic research know-how from Graz University of Technology for various electrochemical energy storage systems are merged. [Read more](#)

EurA AG (Germany)

EurA AG has been established in Ellwangen (Baden-Württemberg, Germany) in 1999. The company currently employs more than 140 persons on 9 locations in Germany, Portugal, and Belgium. As an innovation service provider, EurA advises more than 1,500 mainly medium-sized companies in Germany, covering all industrial sectors. [Read more](#)

Uppsala University (Sweden)

Uppsala University, founded in 1477, is the oldest University in the Nordic countries, and generally ranked among the top 100 universities in the world. Today, it trains more than 43,000 students, and employs 6,000 people. There are about 2,500 active graduate students; 44% of these are women. Each year, the University awards some 270 doctoral degrees. [Read more](#)

Materials Center Leoben Forschung GmbH (Austria)

The Materials Center Leoben Forschung GmbH (MCL) is the leading Austrian institution in the field of applied materials science with around 150 employees. [Read more](#)

VARTA Storage GmbH (Germany)

VARTA Storage GmbH (VS) is a developer and manufacturer of stationary battery storage systems based in Nördlingen, Germany. The company has substantial know-how in the field of energy storage by using long-life lithium-ion batteries and conducts in this context innovative research and development activities. [Read more](#)

University of Warsaw (Poland)

University of Warsaw (UW) was founded in 1816. The University brings together scholars from a variety of disciplines. It is the place of a diversity of scientific research. Nearly 60,000 people study at the University of Warsaw every year. [Read more](#)

ACCUREC Recycling GmbH (Germany)

Accurec is a German SME company, founded in 1995 with its primary target to constitute the consumer battery recycling market in Germany. [Read more](#)



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