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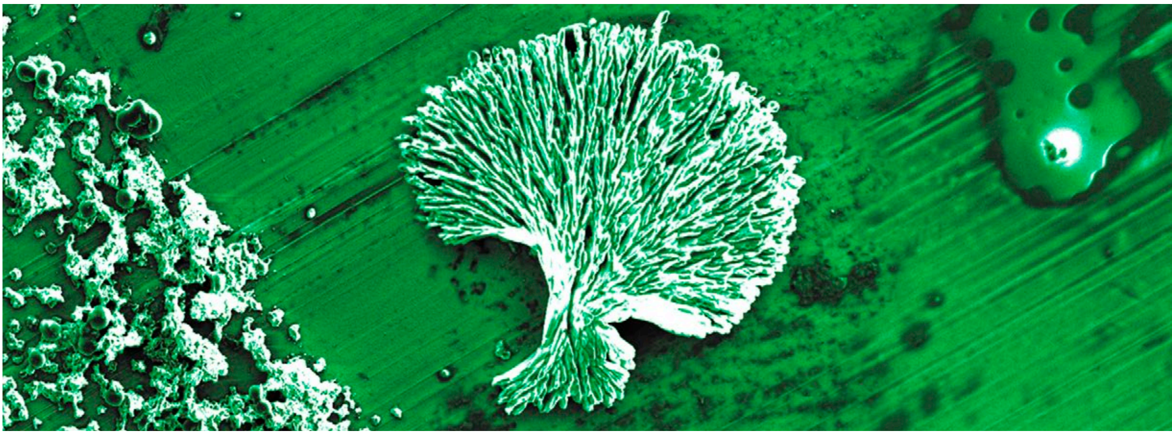

Annual Report 2021

Electrochemistry Laboratory

Cover

Scanning electron micrograph of crystallised lithium salt on a copper current collector.

PAUL SCHERRER INSTITUT

Annual Report 2021

Electrochemistry Laboratory

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Publisher
Electrochemistry Laboratory
Paul Scherrer Institut
5232 Villigen PSI
Switzerland

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Layout
Peter Lutz, LUTZdocu, Uster

Printing
Paul Scherrer Institut

ISSN 1661-5379
DOI: 10.3929/ethz-a-007047464
PSI Electrochemistry Laboratory :: Annual Report 2021

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EDITORIAL

PSI's Electrochemistry Laboratory is the leading center for research in electrochemical energy storage and conversion in Switzerland. The Lab bridges fundamental electrochemical science with applied electrochemical engineering, which is unique even on international standards. This allows us to keep a systemic view even when addressing basic scientific questions.

With the pandemic situation, also 2021 has again been a special year for the Electrochemistry Laboratory. Still, we were able to have a strong scientific output. In this Annual Report 2021, you will not only find details on PSI's Electrochemistry Laboratory, but 12 highlights on outstanding results of the Lab, contributing to the grand challenges of electrochemical energy storage and conversion. Further the Laboratories key facts and figures for 2021 are given, including a full list of our 52 peer reviewed publications and numerous talks demonstrating the leading role of PSI's Electrochemistry Laboratory on the national and international level.

Due to the pandemic situation, the annual 37th Swiss Electrochemistry Symposium «Prospects of Future Transport Electrification» was held successfully on April 21, 2021 as online event with world-class scientists and engineers as speakers, attracting an audience of more than 100 participants. Details of this event you will find also in this annual report on page 60.

2021 was also a successful year for seven of our PhD students who successfully defended their theses. An overview on their work is given in this report. The new PhDs mostly continued their careers directly either in an academic or industrial environment.

At this point it needs to be mentioned that all our projects and results could not have been achieved without the funding we received over the years from industrial partners and the different funding agencies in Switzerland and abroad. We, therefore, would like to take this opportunity to thank all our funding sources hoping to continue our successful collaborations in the years to come.

Felix N. Büchi

THE ELECTROCHEMISTRY LABORATORY AT A GLANCE

Our Mission

Advancing electrochemical energy storage and conversion by

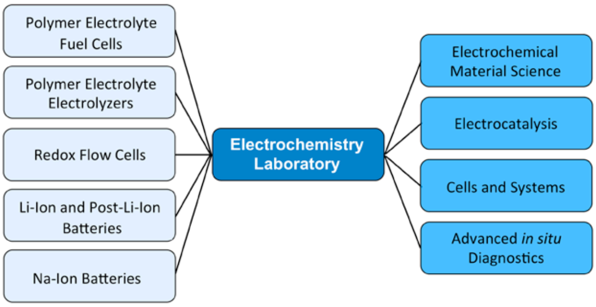
- **developing novel electrochemical materials, cells and devices;**
- **using novel, advanced *in situ* and *operando* methods;**
- **performing fundamental, applied and industry-oriented research.**

PSI's Electrochemistry Laboratory is Switzerland's largest Center for Electrochemical Research with about 70 employees. Our mission is to advance the scientific and technological understanding and implementation of electrochemical energy storage and conversion, specifically in the context of a sustainable energy system. This requires renewable energy to be stored in secondary batteries or chemicals such as hydrogen and be (re-)converted into electricity. The Laboratory's R & D, is hence focused on secondary batteries – specifically Li-based systems –, polymer electrolyte fuel cells and electrolyzers, as well as redox flow cells.

As a research institute's laboratory we are bridging the gap between fundamental science and applied engineering by combining both academically and industrially relevant questions. For the technologies under research, we not only develop fundamental understanding of materials on an atomic and molecular scale (electrochemical materials sciences and electrocatalysis), but also in the applied development of technical cells and devices.

On many technical complexity levels, we are developing and utilizing advanced *in situ* and *operando* diagnostic tools to gain insights in properties and processes from the nanometer to the centimeter scale, respectively, making intensive use of PSI's unique large scale facilities such as the Swiss Light Source (SLS) and the Swiss Neutron Spallation Source (SINQ).

Topics of Electrochemistry Laboratory.



Electrochemical Energy Storage

The vision in Electrochemical Energy Storage is to make significant contributions to the most advanced electrochemical energy storage systems.

The work is focused on rechargeable batteries, which are mainly lithium based. The scientific goal is a profound understanding of the electrochemical processes in complex non-aqueous systems. In particular, of utmost scientific interest are the numerous interactions of all components determining the safety and life time of such systems.

The work equally considers the synthesis of novel materials for electrochemical energy storage and the modification of known materials (e.g., carbon), and material characterization, keeping in mind the entire span from basic science to industrial applications. To answer the scientific questions, we develop various sophisticated *in situ* and *operando* methods for use in the field of non-aqueous solid-state electrochemistry and investigate the physical and electrochemical properties of insertion and conversion materials and electrochemical interfaces *in situ*. Also, we do electrochemical engineering work on three-dimensional electrodes and characterize industrial batteries.

Dr. Thi Thu Diêu Nguyen testing life time of commercial battery cells.

Electrochemical Energy Conversion

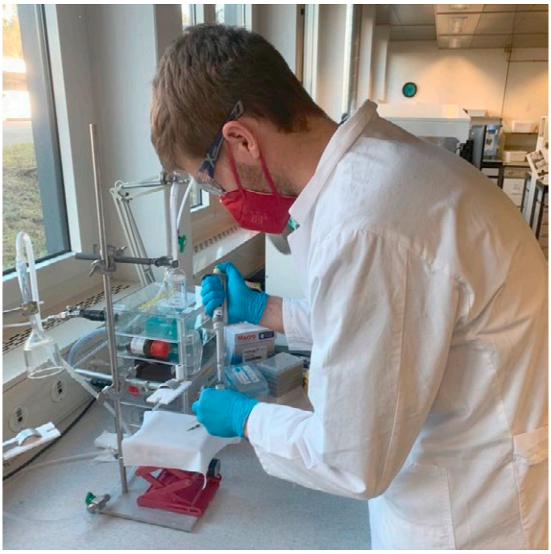
The Electrochemical Energy Conversion focuses on the development and in-depth understanding of materials, processes and devices for the conversion of renewable power to hydrogen (or syngas through the electrochemical reduction of carbon dioxide) and back to power. Especially in the context of a sustainable energy system utilizing hydrogen as an energy carrier, these electrochemical energy conversion steps are of particular importance.

In this topical context the work is focused on Polymer Electrolyte Fuel Cells (PEFC) and Polymer Electrolyte Water Electrolyzers (PEWE) for water electrolysis and the co-electrolysis of CO_2 and water, respectively. In addition, work is devoted on the materials development for Redox Flow Cell systems.

The R & D strategy involves activities on these pathways:

- the development and application of advanced *in situ* and *operando* diagnostic imaging tools on stack, cell and component levels (X-rays and neutrons) with strong use of the large installations at PSI (Swiss Light Source and Neutron Spallation Source);
- research in electrocatalysis and the reaction kinetics of the relevant reactions (e.g., the oxygen electrode reactions) for developing new electrocatalysts and improved understanding of intrinsically limiting factors;
- development of new porous materials for PEFC and PEWE;
- membrane development for next generation devices;
- system, stack and cell testing.

On the level of technology demonstration, we have designed and we develop and operate the so-called hydrogen path on PSI's Energy System Integration (ESI) Platform on the 100 kW level, i.e., operate a technical scale PEWE System including product gas clean-up and a H_2-O_2 PEFC reconversion system, which is based on a joint development with our collaboration partner Swiss Hydrogen SA.



PhD student Justus Diercks preparing an electrode to test the activity of a catalyst to reduce carbon dioxide into useful chemicals.

CURRENT SCIENTIFIC TOPICS

SCIENTIFIC HIGHLIGHTS

Efficient Water Electrolysis at Elevated Temperature Using Commercial Cell Components

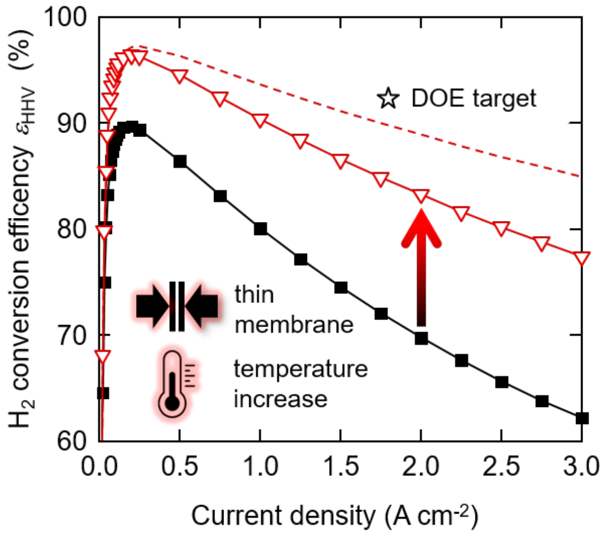
Decarbonization of the energy system across different sectors using power-to-X concepts relies heavily on the availability of low-cost hydrogen produced from renewable power by water electrolysis. Polymer electrolyte water electrolysis (PEWE) is a promising technology for hydrogen (and oxygen) production for distributed as a well as centralized operation. The total cost of hydrogen is dominated by the electricity cost. Therefore, increase of conversion efficiency is pivotal in improving the commercial viability of electrolytically produced hydrogen. In this study, we investigate the prospects of improving conversion efficiency by reducing the membrane thickness from 200 to 50 micron and increasing the cell temperature from 60 to 120°C.

State of the art polymer electrolyte water electrolyzers typically use a perfluorinated proton exchange membrane of the Nafion type with a thickness of 150 to 200 micron and are operated at a cell temperature of up to 60°C. This conservative choice of materials and operating conditions ensures that the lifetime is in the range of ten thousands of hours.

With a view to increasing the conversion efficiency, which corresponds to a decrease of the operating cell voltage, we investigated the effect of using a thinner membrane and increasing the cell temperature up to 120°C. We chose a Nafion membrane of 50 micron thickness. This is expected to reduce the ohmic resistance of the cell. The temperature increase is expected to lead to a reduction of the activation overpotential, in particular for the oxygen evolution reaction (OER), and an increase of the membrane conductivity. The figure shows the conversion efficiency of the water splitting reaction as a function of current density. The poor efficiency at low current density is a result of low faradaic efficiency due to gas crossover through the membrane. The curves go through a maximum and then decrease, which is a consequence of increasing overpotentials. As shown, with a combination of a 50 µm membrane and 120°C cell temperature, the efficiency can be increased by 15 % at a current density of 3 A/cm^2. Further reduction of the membrane thickness is limited by the gas diffusion through the membrane, which poses the risk of the formation of an explosive gas mixture (the lower explosion limit of H_2 in O_2 is 4 %). The main overpotentials at 120°C

Figure 1
Conversion efficiency based on the higher heating value (HHV) of hydrogen for the electrochemical water splitting reaction. Efficiency increase through reduction of membrane thickness from 200 to 50 micron, and increase of cell temperature from 60 to 120°C. The dashed line indicates further expected improvement through elimination of electrical and contact resistances. The star indicates the target of the US Department of Energy (DOE).
Cell: 25 cm^2 active area, Nafion® type membrane, IrO_2-TiO_2 anode catalyst, Pt/C cathode catalyst.

with the thin membrane are related to the OER (0.34 V) and the ohmic resistance (0.36 V). The ohmic resistance is caused by the membrane, the interface between the anode catalyst layer and the titanium based porous transport layer, and electronic/contact resistances in the cell. If we could eliminate the latter, for example by a suitable coating on the titanium parts of the cell, the efficiency could be further enhanced (dashed line in the figure). The star indicates the development target by the US Department of Energy (DOE). We see that further development is necessary on the cell components to reach this goal. Moreover, the durability of the cell at elevated temperature needs to be studied in detail. Degradation phenomena are expected to be accelerated, which needs to be quantified to allow estimates of lifetime. This is the subject of ongoing studies.



Acknowledgement

Swiss Federal Office of Energy (SFOE, Grant No. SI/501603) for funding of this project.

Publication

Insight into Elevated Temperature and Thin Membrane Application for High Efficiency in Polymer Electrolyte Water Electrolysis.
Steffen Garbe, Jonas Futter, Thomas J. Schmidt, Lorenz Gubler.
Electrochimica Acta 377, 138046 (2020).
DOI: 10.1016/j.electacta.2021.138046

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Temperature Dependent Water Transport Mechanism

Subsecond and submicron *operando* X-ray tomographic microscopy (XTM) was applied to reveal the water dynamics inside the gas diffusion layer (GDL) of polymer electrolyte fuel cells (PEFC). Utilizing the instrumental advancements in *operando* XTM of PEFCs the contribution of capillary-fingering and phase-change-induced flow on water transport in GDLs was quantified, for the first time during fuel cell startup at different operation temperatures.

The product water of polymer electrolyte fuel cells (PEFCs) has to pervade the gas diffusion layer (GDL) and can lead to shortages of reactant gas diffusion pathways to the catalyst layer, hence to significant performance losses.

Recent efforts enabled subsecond and submicron *operando* X-ray tomographic microscopy (XTM) over a wide range of cell operating temperatures. The short XTM scan times allowed to quantify the water transport inside the cathode GDL at elevated operating temperatures during a current ramp-up process. Two major water transport mechanisms for water emerging from the catalyst layer (CL) towards the flow fields (FF) are commonly considered in the literature, namely capillary-fingering of liquid water through the pores of the gas diffusion layer, and gas phase transport of water vapor.

The water distribution during a current ramp up process was investigated and analyzed for several locations in the GDL, including MPL cracks, GDL and GDL binder pores. At both temperatures of 40°C and 80°C, capillary-fingering as well as phase-change-induced water transport were observed, though with different contributions. At 40°C the through-plane water distribution was «downhill-like» from the MPL to the flow field similar to observations in *ex situ* liquid injection experiments. The water distribution was initially driven by bottom-connected water clusters located at the GDL-MPL interface, while already after 50 s fully-connected clusters that span from the GDL-MPL interface to the GDL-flow field were dominating. At a typical automotive PEFC operating temperature of about 80°C, at the initial low saturation level, phase-change-induced flow efficiently works for water removal without impeding reactant gas pathways. The through-plane water distribution was more «valley-like» with the saturation increasing mainly at the GDL-MPL and GDL-flow field interface in the first 25 s. Top-connected water clusters located at the GDL-FF interface dominate the water distribution in the first 30 s. These clusters are initially small and disconnected, but grow and merge over time. As soon as the top-connected phase-change-induced water clusters merge with the capillary-fingering driven bottom-connected clusters, fully-connected water clusters dominate the water distribution after only about 40 s.

The possibility of investigating water dynamic phenomena with both high spatial and temporal resolution utilizing synchrotron XTM facilities enables a broad field of future studies of liquid water dynamics in porous transport layers in PEFCs.

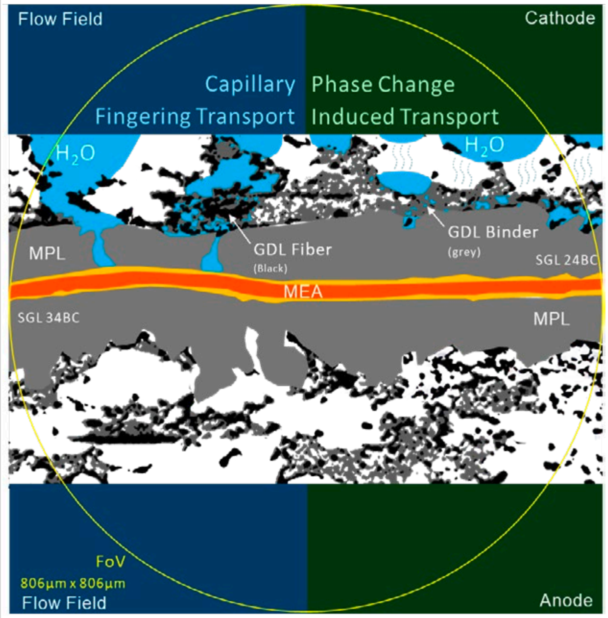

Figure 2
Schematic of the water transport mechanisms in GDL: capillary-fingering driven water transport (left) and phase-change-induced (PCI) water transport (right).

Publication

Temperature Dependent Water Transport Mechanism in Gas Diffusion Layers Revealed by subsecond *Operando* X-ray Tomographic Microscopy.
Hong Xu, Shinya Nagashima, Hai P. Nguyen, Keisuke Kishita, Federica Marone, Felix N. Büchi, Jens Eller.
Journal of Power Sources 470, 229492 (2021).
DOI: 10.1016/j.jpowsour.2021.229492

Acknowledgement

Swiss Federal Office of Energy (SFOE, Grant No. 200021 166064) for funding of this project.

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Scientific Highlight

3

Using X-ray Emission Spectroscopy to Study the Electronic Properties of Single Atom Catalysts

Single atom catalysts hold great promise as O_2- or CO_2-reduction electrocatalysts, but a deeper understanding of their active sites' structure and electronic properties is needed in order to render them sufficiently active and stable. To this end, we have used X-ray emission spectroscopy to determine these catalysts' electronic configuration, and performed *in situ* measurements that unveil the effect of potential on this key feature.

The rollout of renewable energy conversion technologies like fuel cells and electrolyzers requires inexpensive electrocatalysts for their electrodes' reactions. In this context, materials featuring single metal atoms coordinated by nitrogen functionalities in a carbon matrix (so called single atom catalysts, or SACs) offer tremendous potential as catalysts for the reduction of O_2 or CO_2. However, the needed improvements in these materials' catalytic features (i.e., activity, stability and selectivity) require a better understanding of the structure and electronic properties of their active sites.

With this motivation, we have used X-ray emission spectroscopy (XES) to determine the electronic configuration (or spin state) of the active centers in two of these catalysts. To do so, we first collected XES spectra of several compounds with metal coordination environments similar to those of the active sites in SACs and well-defined electronic properties. We then found a correlation between these compounds' spin states and the intensity of their low energy, Kβ X-ray emission spectral feature (at ≈ 7.04 keV), and used this relation to quantify the catalysts' average spin state.

Finally, we performed additional XES measurements on one of these SACs under electrochemically relevant conditions, and proved for the first time that this average spin state changes as a function of the applied potential. In doing so, our results pave the road for the extended use of XES to study the electronic properties of SACs.

Figure 3

In situ Kβ X-ray emission spectra of an Fe-based single atom catalyst, whereby the gray spectrum was acquired at the open circuit voltage (OCV, of ≈ 0.8 V vs. the reversible hydrogen electrode (RHE)), and the green spectrum was recorded at 0.2 V vs. RHE. The loss of intensity in the low energy feature at ≈ 7.04 keV denotes a decrease in the catalysts' average spin state.

Publication

Surface segregation acts as surface engineering for the oxygen evolution reaction on perovskite oxides in alkaline media.
Anthony Boucly, Emiliana Fabbri, Luca Artiglia, Xi Cheng, Daniele Pergolesi, Markus Ammann, Thomas J. Schmidt.
Chemistry of Materials, 32(12), 2020.
DOI: 10.1021/acs.chemmater.0c01396

Acknowledgement

The Swiss National Science Foundation (Ambizione Energy grant nr. PZENP2_173632) is gratefully acknowledged for the financial support.

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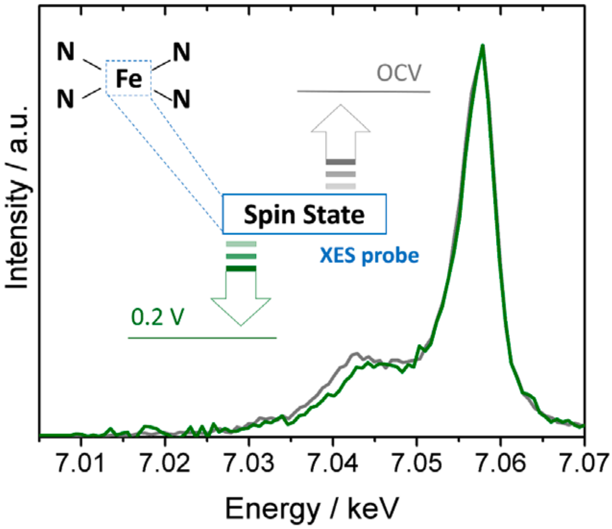

Scientific Highlight

4

Impact of Micro-porous Layers (MPL) on Two-phase Flow in Electrolyzers

Polymer Electrolyte Water Electrolyzers (PEWE), due to their excellent dynamic characteristics, can provide an economical solution to the intermittent nature of new renewable sources, by converting the excess electricity into hydrogen. However, improvements in efficiency and in capital cost are still required for the large-scale deployment of this solution. In this context, we studied whether the efficiency improvements observed when using porous structures featuring a micro-porous layer (MPL) can be attributed to a better distribution of the water.

In fact, the water supplied and the gas produced by the electrochemical reaction share the same pathway, resulting in so-called two-phase flow phenomena. If the volume occupied by the gas hinders the supply of water, this can have a detrimental impact on efficiency.

To elucidate the impact of adding a MPL – an additional layer with a similar porosity, but a finer grain size as the base porous structure –, we used high resolution neutron radiography to measure the water and gas distribution across the structure of operating PEWE cells. An important characteristic of this method is that it provides an important contrast for water, while the neutron beam can penetrate through large thicknesses of relatively dense materials. The conducted measurements demonstrated that the presence of a MPL has a significant impact on the water and gas distribution. Near the catalyst layer where the reaction takes place, a larger water saturation is observed – meaning a much lower fraction of the pores is occupied by gas – than when no MPL is used. This effect is particularly strong when using the MPL with the smallest pore size (6 µm). On the other hand, using a MPL results in a much lower water saturation (higher gas fraction) in the bulk of the coarse porous layer. The strong contrast in saturation between the MPL and the region with coarser pores can be explained by the fact that each gas transport pathway through the small pores of the MPL emerges in a large pore, resulting in a significant increase of the volume occupied by gas at this interface. The observation that the presence of a MPL strongly affects the water distribution in a positive way near the electrode may lead to conclude that the improved water supply is responsible – at least

Figure 4

a) Images of water distribution across the structure of PEWE cells without and with microporous layers (MPLs).
b) Corresponding quantitative water distribution profile.

in part – for the improvement of performance observed when using MPLs. However, this hypothesis is put in perspective by the fact that the magnitude of the impact of MPL on water distribution is a function of the MPL pore size, with a stronger impact for lower pore sizes. On the other hand, the performance improvement is observed when using a MPL, independently on the pore size. Therefore, another explanation for the performance improvement is that the MPL provides a closer contact to the electrode, removing the dependency on the ionic conductivity of the catalyst layer.

Finally, it is remarkable that the performance improvement when using a MPL occurs despite a very strong increase of the gas saturation in the coarse part of the porous layer. Obviously, the remaining water pathways in this case are sufficient to supply the electrochemical reaction without detrimental effects. This highlights the fact that an important potential remains for more economical structures for water supply and gas removal, as long as the interface to the catalyst layer is carefully designed.

Publication

Elucidation of fluid streamlining in multi-layered porous transport layers for polymer electrolyte water electrolyzers by *operando* neutron radiography.
Mateusz Zlobinski, Tobias Schuler, Felix N. Büchi, Thomas J. Schmidt, Pierre Boillat.
J. Electrochem. Soc. 168, 014505 (2021).
DOI: 10.1149/1945-7111/abcf19

Acknowledgement

The authors would like to thank the Swiss Federal Office of Energy (SFOE, for Grant Nos. SI/501331-01 and SI/8100066). T.J. Schmidt thanks Innosuisse and the Swiss Competence Center for Energy Research (SCCER) Heat & Electricity Storage.

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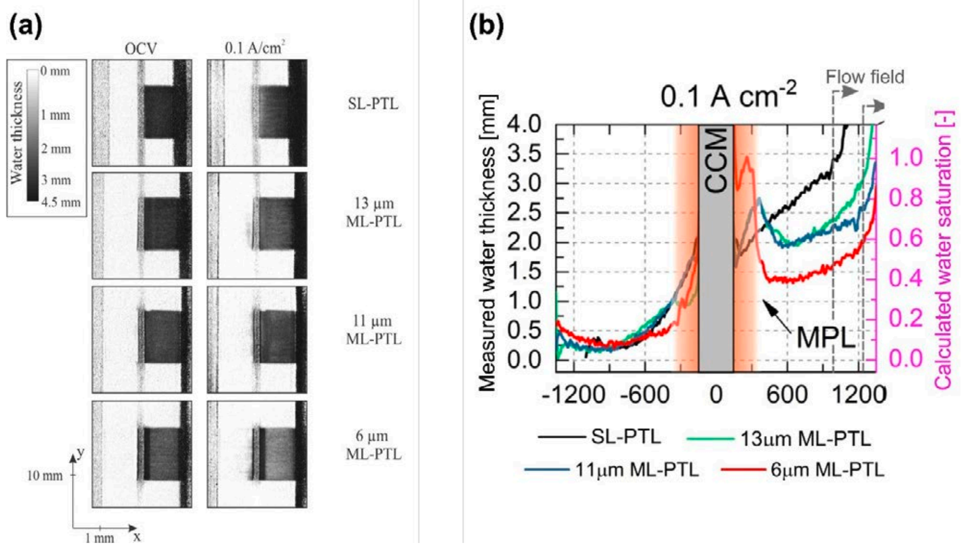


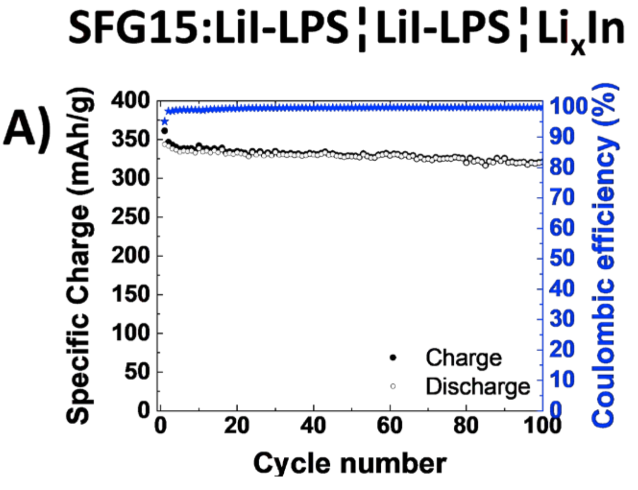
Excellent Cycling Stability of Graphite in All-solid-state Battery Using Sulfide Solid Electrolyte

All-solid-state lithium ion batteries represent a promising battery technology for boosting the volumetric energy density and promising a superior safety. In this study excellent cycling stability of graphite anode material has been demonstrated in combination with a sulfide-based solid electrolyte. Furthermore, we evaluated the stability of the graphite-electrolyte interface by analyzing the normalized cumulative irreversible charge during cycling experiments.

All-solid-state lithium ion batteries could offer the great opportunity of enhanced safety, longer lifetime and higher energy density thanks to the replacement of the volatile and flammable organic liquid electrolyte by a non-flammable solid electrolyte. The sulfide based materials are among the most attractive solid electrolytes as they offer the highest ionic conductivities at room temperature and have low grain boundary resistance. Besides, the sulfide solid electrolytes can be cold pressed; thus, they can be more easily processed than their oxide or phosphate counterparts. However, their very narrow electrochemical stability window limits the ion transport across the solid electrolyte–active materials interfaces. In fact, for potentials negative to 1.7 V vs. Li^+/Li, a reductive process occurs leading to their decomposition into Li_2S and Li_3P species.

In this study, we report for the first time long-term electrochemical cycling of graphite in two different sulfide-based solid electrolytes, $0.75Li_2S$-$0.25P_2S_5$ and $0.3LiI$-$0.7(0.75Li_2S$-$0.25P_2S_5)$ denoted (LPS) and (LiI-LPS), respectively, with high confining pressure. We also evaluate the stability of the graphite–solid electrolyte interface by analyzing the normalized cumulative irreversible charge during cycling experiments. We examine how, over several months of cycling, the electrochemical performance of graphite is affected by the type of the sulfide electrolyte and the particle size, as well as the type of graphite (different size and shape). For different electrolyte-electrode combinations, we report the specific charge, the coulombic efficiency, and the capacity retention during cycling.

Our results demonstrate excellent cycling stability and high utilization of graphite electrodes in cells with LPS based solid electrolytes. At a given



rate, the utilization of graphite can be improved by decreasing the size of the sulfide electrolyte particles. The interface of LPS to graphite is however unstable. The layer of decomposition products at the interface increases rapidly during the first cycles and then does not stop over time, following a kind of root type function. A noticeable irreversible charge consumption exists even after several months of cycling. We show that coulombic efficiencies close to 100 % in a particular cycle are not sufficient to assume good long-term cycling behavior of a cell. To this point, we propose to compare the cumulative irreversible charge for the entire cycling to avoid misinterpretation of the electrochemical cycling data.

Finally, we demonstrated that optimizing the graphite electrode morphology is important to enhance the power performance of the cells.

Publication

Study of Graphite Cycling in Sulfide Solid Electrolytes.
Laura Höltschi, Franziska Jud, Camelia Borca, Thomas Huthwelker, Claire Villevieille, Vincent Pelé, Christian Jordy, Mario El Kazzi, Petr Novák.
J. Electrochem. Soc. 167 (11), 110558 (2021).
DOI: 10.1149/1945-7111/aba36f

Acknowledgement

The authors acknowledge the financial support from SAFT.

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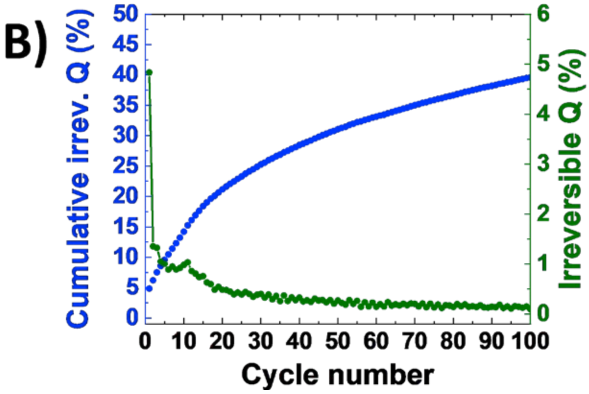

Figure 5
A) Specific charge with respect to the graphite mass and B) normalized cumulative and per cycle, respectively, irreversible charge (Q) of a half-cell with the LiI-LPS solid electrolyte layer and the working electrode SFG15:cLiI-LPS (50:50 wt%). The cell is cycled at 25°C at C/20 rate between –0.61 and 1.50 V vs. Li_xIn.

Cross-talk-suppressing Electrolyte Additive for Li-ion Batteries

Control of interfacial reactivity at high-voltage is a key to high-energy-density Li-ion batteries. 2-aminoethyldiphenyl borate was investigated as an electrolyte additive to stabilize surface and bulk of both NCM851005 and graphite in the cell with upper cut-off voltage of 4.4 V vs Li$^+$/Li. AEDB almost completely eliminated the «cross-talk» in the cell, by significantly reducing metal leaching from the cathode, preventing their deposition at the anode, and further electrolyte decomposition.

Energy density of Li-ion cells can be increased by using materials with higher capacity, extending operating voltage window, or both. The classical cathode active materials, based on intercalation chemistry have limited capacity of below 300 mAh/g, most often even below 200 mAh/g. Therefore, here the strategy is to increase battery cell's upper cut-off potential, resulting not only in higher voltage of the cell but also extracting additional capacity from the cathode. The hurdles in following this strategy often are related to electrolyte instability at high potentials and absence of stable interface formation at the cathode surface. Control of electrode-electrolyte interfacial reactivity at high-voltage, therefore, is a key to successfully obtain high-energy-density lithium-ion batteries. In this study, 2-aminoethyldiphenyl borate (AEDB) was investigated as a multifunctional electrolyte additive in stabilizing surface and bulk of both Ni-rich $LiNi_{0.85}Co_{0.1}Mn_{0.05}O_2$ (NCM851005) and graphite electrodes with elevated upper cut-off voltage of 4.4 V vs Li$^+$/Li. The AEDB improved performance of both

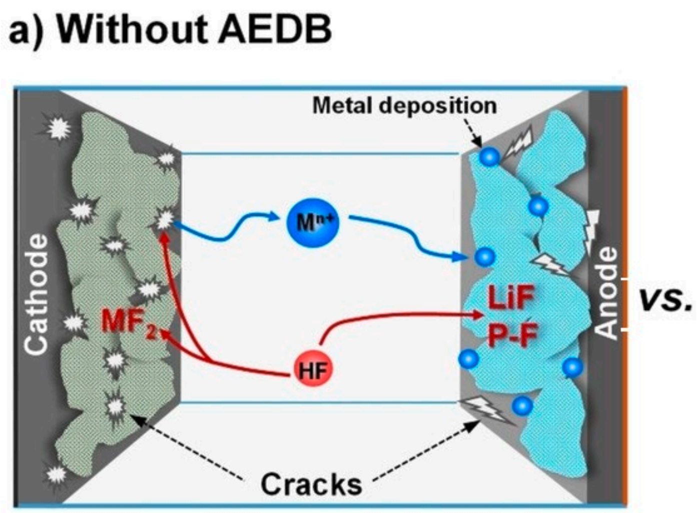

Figure 6
Schematic representation of AEDB electrolyte additive's effects on processes within Li-ion battery cells:
a) processes without additive and b) with AEDB.

cathode and anode materials in a full-cell, suppressing crack formation of cathode particles and helping to preserve the structure of oxide and graphite after multiple cycles. Moreover, even if the effect of AEDB in half-cells was rather modest, in full-cells its addition resulted in tremendous performance improvement. The graphite ǁ NCM851005 full-cell in the presence of AEDB has a capacity retention of 88 % after 100 cycles, even when the upper cut-off voltage was 4.35 V, corresponding to 4.4 V vs Li^+/Li, whereas with standard electrolyte under the same conditions the retention was only 21 %. Most importantly, AEDB almost completely eliminated so called «cross-talk» between the two electrodes, by significantly reducing metal dissolution from the cathode, preventing them being deposited at the anode, where they could catalyse further electrolyte decomposition. Noteworthy is the fact that the effect of the AEDB in half-cells is rather unimpressive, while in the full-cell it shows the full potential, showing the importance of additive testing to be performed in the environment of their intended use – the full-cell.

Publication

Cross-Talk-Suppressing Electrolyte Additive Enabling High Voltage Performance of Ni-Rich Layered Oxides in Li-Ion Batteries.
Hieu Quang Pham, Minh Tri Nguyen,
Mohamed Tarik, Mario El Kazzi, Sigita Trabesinger.
ChemSusChem 14, 1–15 (2021).
DOI: 10.1149/1945-7111/ab9b9a

Acknowledgement

This work was financially supported by the BASF Scientific Network on Electrochemistry and Batteries, and the SCCER Heat and Electricity Storage

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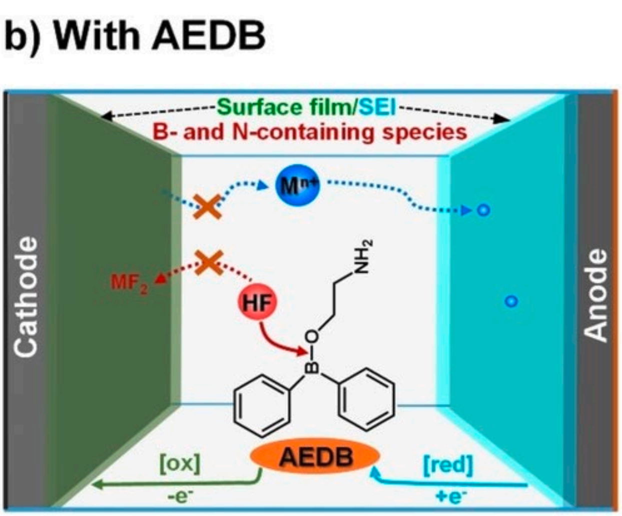

Correlation between Oxygen Vacancies and Oxygen Evolution Reaction Activity for a Model Electrode: $PrBaCo_2O_{5+\delta}$

The role of the oxygen stoichiometry of perovskite catalysts in the oxygen evolution reaction (OER) is systematically studied in the $PrBaCo_2O_{5+\delta}$ family. The reduced number of physical/chemical variables combined with in-depth characterizations such as neutron diffraction, O K-edge X-ray absorption spectroscopy (XAS), electron energy loss spectroscopy (EELS), magnetization and scanning transmission electron microscopy (STEM) studies, helps investigating the complex correlation between OER activity and a single perovskite property, such as the oxygen content. Larger amount of oxygen vacancies appears to facilitate the OER, possibly contributing to the mechanism involving the oxidation of lattice oxygen, i.e., the lattice oxygen evolution reaction (LOER). Furthermore, not only the number of vacancies but also their local arrangement in the perovskite lattice influences the OER activity, with a clear drop for the more stable, ordered stoichiometry.

In recent years, great efforts have been dedicated to the understanding of the water-splitting mechanism and to the improvement of electrocatalyst materials. In particular, the anodic reaction, the oxygen evolution reaction (OER), is affected by slow kinetics and by significant overpotentials, even with the state-of-the-art electrocatalysts such as IrO_2. The broad commercialization of green-hydrogen technologies urges for the use of highly active, stable and cost-effective electrocatalyst materials.

Significant interest and promising results in alkaline environment came from perovskite-type oxides, where the large chemical flexibility allows for a broad variety of elemental compositions and fine properties tuning. Despite the great scientific interest and efforts, however, there is still no univocal consensus on the oxygen evolution reaction (OER) mechanism and on the perovskite electro-chemical contribution in the OER. The systematic study of the correlation between oxygen stoichiometry and OER activity in the $PrBaCo_2O_{5+\delta}$ materials shows an increase of OER activity as the lattice oxygen vacancies increase (see Figure 7). Furthermore, the trend of OER activity vs. oxygen vacancies content is not linear, indicating that other aspects should be considered to fully understand the key parameters of perovskite oxide catalysts affecting the OER. A clear activity drop is present for $\delta \sim 0.5$ (see Figure 7), where the oxygen-vacancies in the Pr plane form alternating $[CoO_6]$ octahedral and $[CoO_5]$ square pyramids, with clear changes in the Co-O coordination and O-Co-O plane buckling. This suggests that oxygen vacancy ordering is detrimental for the OER activity of perovskites. The OER activity was correlated also with other physico-chemical properties of the perovskite materials, such as electronic conductivity, Co 3d–O 2p orbitals hybridization and electronic configuration, but with none of them, a clear correlation was observed.

Publication

Temperature Dependent Water Transport Mechanism in Gas Diffusion Layers Revealed by subsecond *Operando* X-ray Tomographic Microscopy.
Elena Marelli, Jaume Gazquez, Emiliya Poghosyan, Elisabeth Müller, Dariusz J. Gawryluk, Ekaterina Pomjakushina, Denis Sheptyakov, Cinthia Piamonteze, Dino Aegerter, Thomas J. Schmidt, Marisa Medarde, Emiliana Fabbri.
Angew.Chem. Int. 60, 14609–14619 (2021).
DOI International Edition: 10.1002/anie.202103151

Acknowledgement

The authors gratefully acknowledge Paul Scherrer Institut CROSS initiative.

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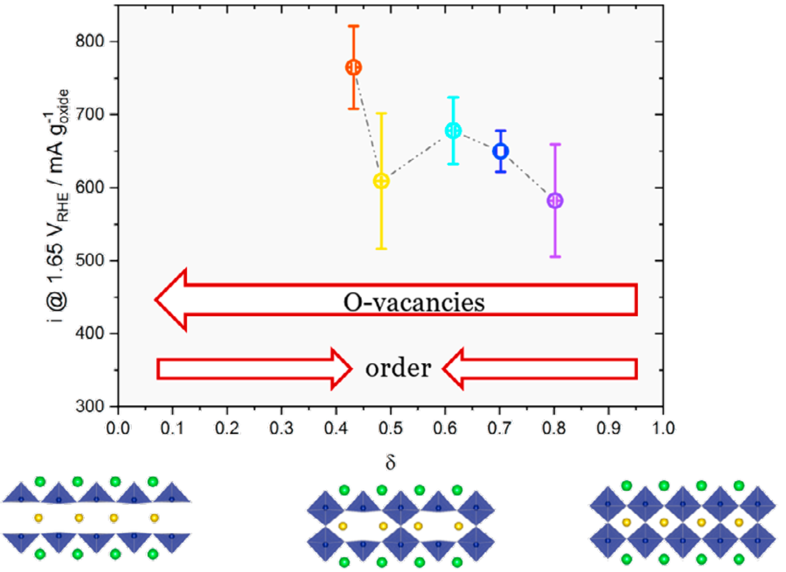


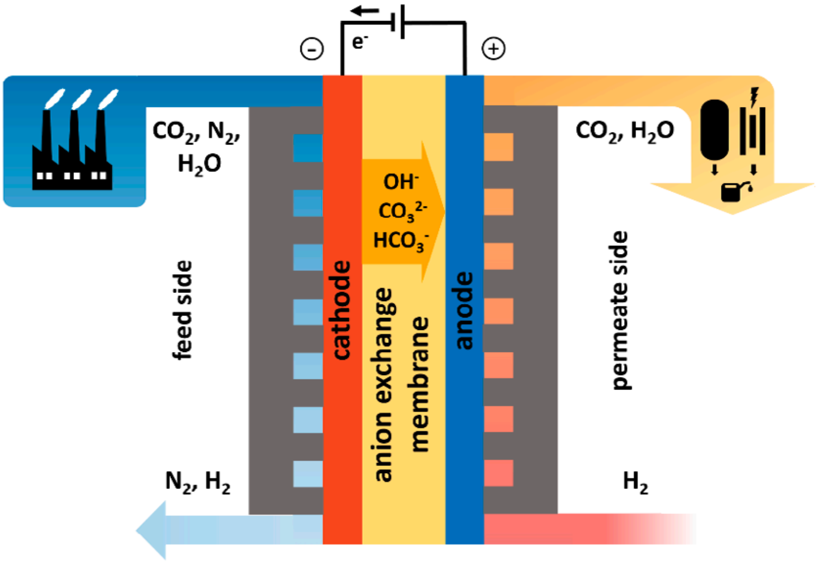
Figure 7
Oxygen evolution reaction (OER) activity variation as function of oxygen stoichiometry in the PrBaCo$_2$O$_{5+\delta}$ samples.
The oxygen vacancies distribution in the ordered PrBaCo$_2$O$_{5+\delta}$ structures with $\delta = 0$, 0.5 and 1 are depicted at the bottom.

An Electrochemical Membrane Process for CO_2 Capture

CO_2 capture from dilute gas mixtures (e.g., combustion flue gases, air) is increasingly recognized as a critical technological pathway towards stemming catastrophic climate change. Conventional thermal-based processes for removing CO_2 from flue gas (e.g., amine scrubbing) are energy intensive and significantly reduce power plant efficiency. Electrochemical separation approaches have the potential to reduce these power requirements considerably by using electrons to transport CO_2 as (bi)-carbonate ions across an alkaline membrane.

Electrochemically driven CO_2 capture is a rapidly growing field of research that represents a wide yet largely unexplored parameter space.

We have developed a first-of-its-kind methodology that uses an anion exchange membrane sandwiched between Pt/C catalyst-coated gas diffusion electrodes within a cell, as shown in Figure 8. OH^- ions that are generated on the feed (cathode) side through the hydrogen evolution reaction (HER) react with CO_2 molecules present in the feed gas stream to form HCO_3^- and CO_3^{2-} ions (carbonation). These ions then electromigrate across the membrane to the permeate (anode) side, where they react with a stream of H_2 to form a mixture of CO_2 and H_2O through the hydrogen oxidation reaction (HOR). The net result is the transport of CO_2 from the feed side to the permeate side of the cell, where the resulting mixture of CO_2 and re-



sidual H_2 could be used for a downstream fuel synthesis process, such as methanol synthesis. We performed cell polarization experiments with current densities up to 50 mA·cm^{-2} using 0.1–100 % CO_2 in N_2 as the feed gas, and permeate CO_2 concentrations were monitored using on-line gas analysis. The bar plot in Figure 8 shows the molar specific energy consumption of the CO_2 separation process (log scale) at the current densities of 5 and 10 mA·cm^{-2} for various feed gas concentrations, with the minimum theoretical separation work for the given gas mixture shown for reference.

As shown in Figure 8, the energy consumption is heavily dependent on the feed gas concentration, with values of 16–26 kJ·mol^{-1} for 15 % CO_2 (similar to the concentration found in coal flue gas) and 189–390 kJ·mol^{-1} for 0.1 % CO_2 (similar to what is found in air). This is largely an effect of increased fraction of OH^- pumping at low CO_2 concentrations, which limits the applicability of the approach for sub-percent concentrations of CO_2. Further experiments successfully demonstrated the feasibility of electrochemical CO_2 pumping against concentration and pressure gradients. Finally, we employed a simplified techno-economic model to examine the cost dynamics of the system, showing that current densities of >50 mA·cm^{-2} and faradaic efficiencies of 50 % are necessary for the process to be economically viable.

The results represent an important step in mapping out the current capabilities and limitations of a membrane electrochemical process for CO_2 capture, showing where efforts should be directed in future experimental campaigns.

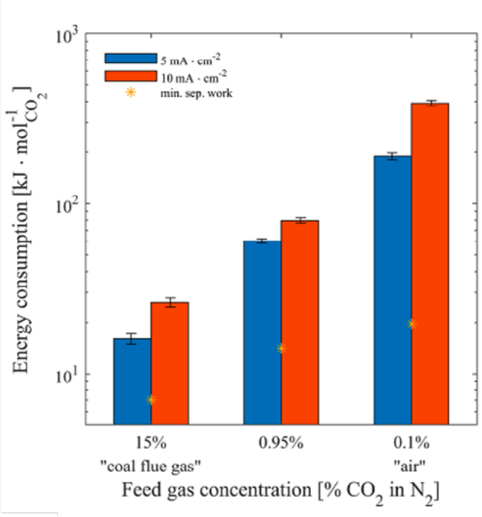


Figure 8

Schematic of the anion exchange membrane CO_2 concentration device and specific energy consumption for various concentrations of CO_2 in N_2.

Publication

Separation of CO_2 from Dilute Gas Streams Using a Membrane Electrochemical Cell.
Alexander P. Muroyama, Alexandra Beard,
Bernhard Pribyl-Kranewitter, Lorenz Gubler.
ACS ES&T Engineering 1(5), 905–916 (2021).
DOI: 10.1021/acsestengg.1c00048

Acknowledgement

The authors would like to acknowledge Shell Global Solutions BV for providing the funding for this work.

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First Direct Observation of the Oxygen Transport in Polymer Electrolyte Water Electrolysis

PSI researchers have developed a new methodology for studying the complex transport processes in polymer electrolyte water electrolysis (PEWE). Using advanced *operando* X-ray tomographic microscopy, we were able to observe for the first time the formation of oxygen pathways in the porous transport layer, in three dimensions. Understanding oxygen transport is crucial for improving PEWE technology and this work provides precious insights for the design of future, better-performing PEWE cells.

Based on the development of a new imaging methodology, the nature of the complex two-phase transport occurring in polymer electrolyte water electrolysis (PEWE) is studied.

With a novel 3D-printed *operando* cell, the use of advanced *operando* X-ray tomographic microscopy at the TOMCAT beamline of SLS, and innovative data analysis techniques, we were able to make the first observation of the oxygen transport in a representative titanium anodic porous transport layer (PTL) during operation. Results show how oxygen forms stable pathways inside the bulk of the PTL, but the pathways get disrupted in the proximity of the water channel. From the correlation with electrochemical data, it is shown that the oxygen saturation in the cell does not increase considerably with current density and that the majority of the oxygen accumulates near the catalyst layer. Furthermore, the water tends to occupy the smaller pores of the PTL and oxygen

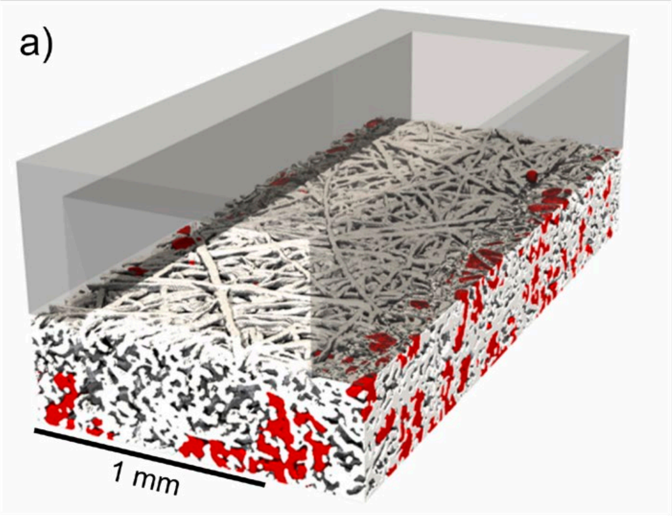

Figure 9
a) Three-dimensional rendering from the tomographic data acquired while the cell was operating at 0.5 Acm^{-2} (the color of the Ti fibers is white while the oxygen phase is depicted in red).
b) Oxygen saturation measured as a function of current density.

the larger ones. The emerging oxygen pathways are organized in many distinct and unconnected clusters. However, with increasing current density, small bridges are formed and more clusters get connected.

Learning from these unique findings, we proposed a set of guidelines for realizing better performing PTL materials, able to operate more efficiently at high current densities. Water occupies the smaller pores, and to avoid the oxygen invasion into these pores, which deteriorates transport in the water network, we propose the realization of anisotropic PTLs having a smaller in-plane throat size distribution than through-plane throats. The presented results and future experiments using different PTL types will pave the way for the development of more rationally designed and better performing PEWE PTLs.

Publication

Unraveling two-phase transport in porous transport layer materials for polymer electrolyte water electrolysis.
Salvatore De Angelis, Tobias Schuler, Margarita A. Charalambous, Federica Marone, Thomas J. Schmidt, Felix N. Büchi.
J. Mater. Chem. A 9, 22102–22113 (2021).
DOI: 10.1039/D1TA03379D

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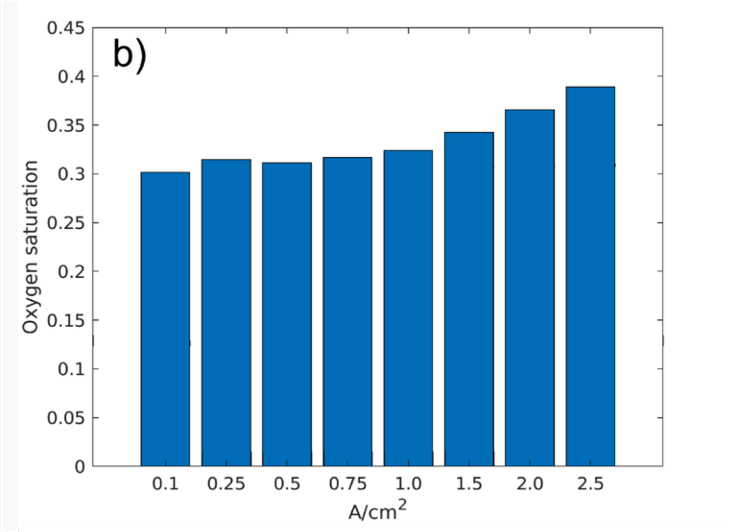

Scientific
Highlight

10

Towards *In situ* Imaging of Electrolyte Physical and Chemical Changes in Li-ion Batteries

The Lithium conducting electrolyte is a critical component of Li-ion batteries, as it has an important impact on performance and its durability, in particular in case of extreme environmental conditions. At low temperatures, a partial solidification can occur, while high temperatures can promote the degradation of the electrolyte. Using time-of-flight (ToF) neutron radiography, the possibility of imaging such changes in a fully non-invasive manner was demonstrated for the first time.

The hydrogen contained in Li-ion battery electrolytes has a very large neutron scattering cross-section, resulting in a strong contrast of the electrolyte in neutron imaging. Moreover, due to inelastic scattering interactions, the intensity of transmitted neutrons does not only depend on the concentration of hydrogen atoms but also on their motions due to diffusion or molecular vibrations. These contributions can be evidenced using ToF neutron radiography, which allows to measure the transmission as a function of the neutron wavelength. The impact of both chemical composition and aggregate state on the neutron transmission spectrum were identified in measurements on solvent mixtures used for battery electrolytes. In particular, in an ethylene carbonate (EC) / i-ethyl carbonate (DEC) mixture at 17°C exhibiting sedimentation, an important difference in neutron transmission spectrum was observed between the solidified and liquid regions, with a much flatter wavelength dependency at long

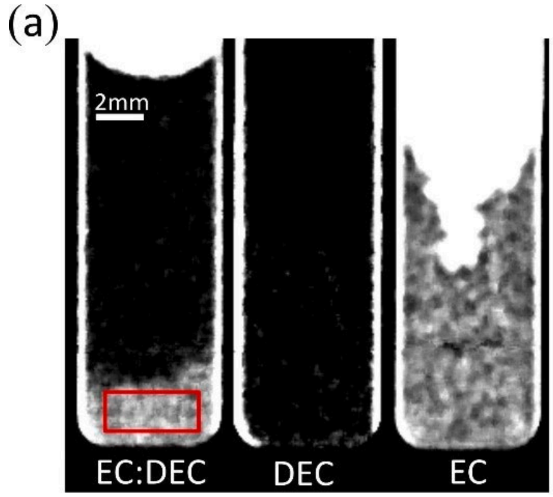

Figure 10
(a) Neutron transmission image of three different solvents in cuvettes. On the left side (EC:DEC sample), sedimentation is visible at the bottom of the cuvette. (b) Neutron scattering cross-sections as a function of wavelength, extracted from the measurements. For a better comparison of the fine differences between the cross-sections, the values are normalized to the cross section of polyethylene, and to their values at 3 Å.

wavelengths. The reason for this change is both a composition change – as the sediment region is expected to be enriched with the EC component – and a suppression of the diffusion in the solidified component.

This result illustrates the ability of ToF neutron radiography to image changes occurring to battery electrolytes as a result of extreme conditions or of degradation. In contrast to other methods for electrolyte analysis such as Fourier Transform Infrared Spectroscopy (FTIR), the interest of neutron imaging is its fully non-invasive aspect, as neutrons can be transmitted through non-transparent materials and even metallic casings. In summary, the obtained results pave the way for *in situ* analysis of electrolyte characteristics in Li-ion batteries.

Publication

Study of Phase Changes in Lithium-Ion Battery Electrolytes via Spectroscopic Neutron Imaging.
R. Carreon-Ruiz, G. Burca, M. Cochet, P. Kadletz, J. Lee, A.S. Tremsin, E. Winter, R. Woracek, M. Zlobinski, L. Gubler, P. Boillat.
240th meeting of the Electrochemical Society, October 10–14, 2021 (digital meeting).

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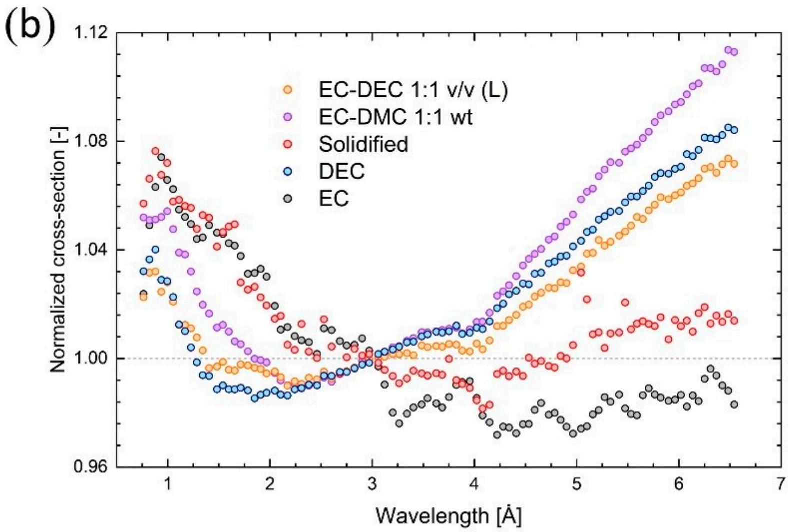


Deciphering the Mechanism of FEC-induced SEI Formation in Li-ion Batteries

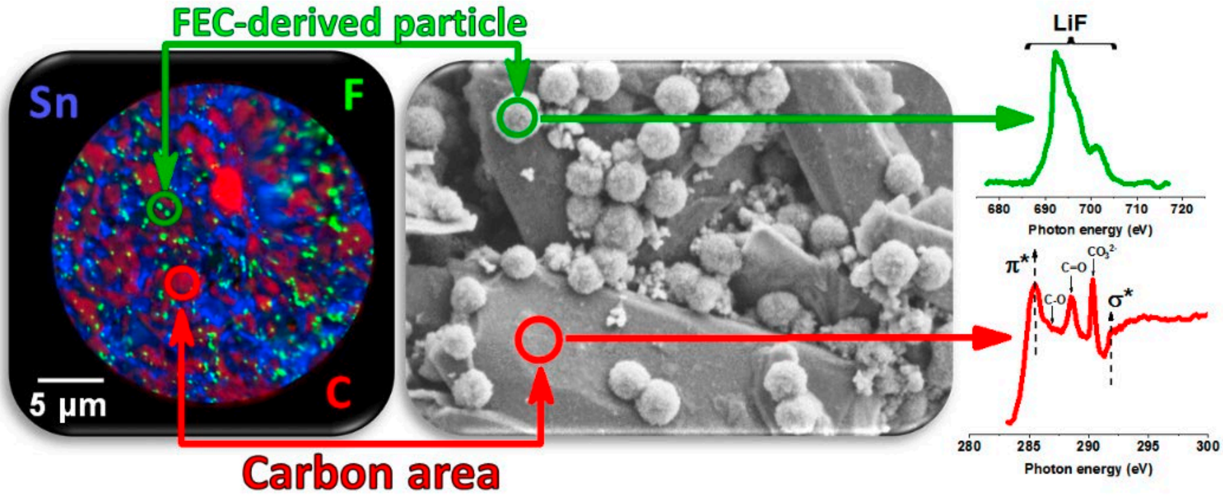
Fluoroethylene-carbonate is often referred to as a film-forming electrolyte additive for Li-ion batteries, resulting in high quality Solid–Electrolyte-Interphase on the negative electrode. However, the underlying formation mechanism, even if thought to be known, has been only clarified due to our targeted experimental design, combining systematic electrochemical, chemical and microscopy characterization techniques. We have shown that first the formation of inorganic LiF-rich particles takes place and only later the carbonate-rich film is actually formed.

High energy density is one of the clear trends in future battery technologies, especially in portable devices and mobility sectors. Energy density of Li-ion cells can be increased by using materials with higher capacity and by extending the operating voltage window of the cell.

Despite the fact that positive electrodes have lower specific capacities than the negative one, the increase of negative electrode capacity also leads to significant improvement of energy density. Besides the most-commonly used graphite, there is a number of negative electrode active materials, whose capacity is many times higher than that of graphite, such as silicon, tin oxide and others. However, these materials often exhibit volume expansion upon lithiation and therefore a SEI formed on their surface is unstable. The most popular strategy for improving their cycling stability is the use of electrolyte additives that are reduced at the potentials more positive than electrolyte solvents. Their decomposition products are incorporated in the SEI, leading to a more stable electrode/electrolyte interface.

One of the most-investigated electrolyte additives is fluoroethylene carbonate (FEC), because it has been shown to lead to a significant performance enhancement of Si- and Sn-based anodes. Many reaction pathways have been proposed but there is currently no agreement on the exact type of chemical compounds constituting the decomposition products, as well as on the mechanism of FEC decomposition.

For this reason we embarked on a systematic study, tracking morphological and chemical changes during electrochemical decomposition of FEC, and found that despite FEC often being referred to as film-forming additive, in the first stage of its decomposition, spherical particles, mainly consisting of lithium fluoride, are formed. And only later the carbonate-rich film covers the entire electrode, including the LiF-rich spheres. This phenomenon has been overlooked by researchers earlier due to a very simple reason – the imaging by electrons, using SEM, is often done at high accelerating voltages, i.e. 10–20 kV, as it allows simultaneous EDX analysis, at least in older generations of characterization tools. At these acceleration voltages, the LiF particles become invisible, as is shown in the supplementary information of our publication. And only acceleration voltages below 5 kV allow to visualize the spherical particles of LiF, formed by FEC reduction.



We have also found that the size and amount of particles strongly depend on the testing cell medium, where electrolytes with higher dielectric constant lead to larger particle size, as do the high-capacity active electrode materials, both contributing to the final properties of FEC-derived SEI. The results of this study provide a deeper understanding of how fluorine additives work and enable tuning of the SEI properties by using laws of simple crystal-growth theory to fit the conditions leading to the desired morphological and chemical outcome. This is an important step to enabling the rational design of fluorine-rich electrolytes and fluorine-based sacrificial additives towards the stabilization of high-specific-capacity anodes and, consequently, of batteries with high energy density.

Figure 11
XPEEM combined image (left) and spectroscopic signals (right) show clear differences if recorded from FEC-derived spherical particles or from the particle-free surface, as shown in SEM image (middle).

Publication

Evidence for Stepwise Formation of Solid Electrolyte Interphase in a Li-ion Battery.
Yuri Surace, Daniela Leanza, Marta Mirolo, Lukasz Kondracki, C.A.F. Vaz, Mario El Kazzi, Petr Novák, Sigita Trabesinger.
Energy Storage Materials 44, 156–167 (2022).
DOI: 10.1016/j.ensm.2021.10.013

Acknowledgement

InnoSuisse (Swiss Innovation Agency, project number 18254.2) and PSI CROSS initiative are acknowledged for financial support. Part of the work was performed at the Surfaces/Interfaces Microscopy (SIM) beamline of the Swiss Light Source (SLS), Paul Scherrer Institute (PSI).

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Reactivity and Potential Profile across the Electrified $LiCoO_2$-Li_3PS_4 Interface Probed by *Operando* X-ray Photoelectron Spectroscopy

All-solid-state lithium batteries are a promising alternative for next generation safe energy storage devices, provided that parasitic side reactions and the resulting hindrances in ionic transport at the electrolyte-electrode interface can be overcome. Motivated by the need for a fundamental understanding of such interface, we present here real-time measurements of the (electro-)chemical reactivity and local surface potential at the electrified interface Li_3PS_4 and $LiCoO_2$ using *operando* X-ray photoelectron spectroscopy.

While state-of-the-art lithium-ion batteries (LiBs) are reaching their inherent performance limits with respect to energy density and safety, all-solid-state batteries (ASSBs) are a promising alternative for boosting the volumetric energy density, with superior safety characteristics. In the past decades, significant progress has been made in the discovery of solid-state ion conductors capable of achieving room temperature ionic conductivity comparable to their liquid homologues. However, poor mechanical contact and parasitic (electro-)chemical side reactions limit ion transport across the solid electrolyte (SE)-active materials (AMs) interfaces. The chemical and electronic nature of the latter is distinctively different in ASSBs from the conventional LiBs.

For the liquid electrolyte system, an excellent understanding of electrified liquid-solid interfaces has been achieved theoretically and experimentally through the detailed investigation of the electrochemical double layer and the solid-electrolyte-interphase. Today, this fundamental knowledge provides important guidance towards improving performance parameters, such as reducing charge-transfer resistances of energy storage and conversion devices. Similarly, interactions at the electrified SE-AMs interfaces play a critical role in the performance of ASSB devices, but the full picture of the exact mechanisms associated with the (electro-)chemical reactions and the electronic properties, monitored in real time upon cycling, has not been fully achieved yet.

To bridge this gap, we investigate the (electro-)chemical and electronic processes occurring at a representative interface between a cathode AM $LiCoO_2$ (LCO) and the glassy SE Li_3PS_4 (LPS). Despite the high room temperature ionic conductivity of LPS (approx. 0.4 mS cm^{-1}), it suffers from a narrow electrochemical stability window, which is a drawback for practical applications. As shown in several post-mortem X-ray photoelectron spectroscopy (XPS) studies, delithiation of LPS occurs with polymerization of the PS_4^{3-} tetrahedra via polysulfidic bridges upon cycling the cell up to high voltages. In addition to the intrinsic instability of LPS at the operation potentials of common cathode AMs, a mutual diffusion region, where both ions and electrons are mobile, has been suggested to occur. The formation of an interlayer through inter-

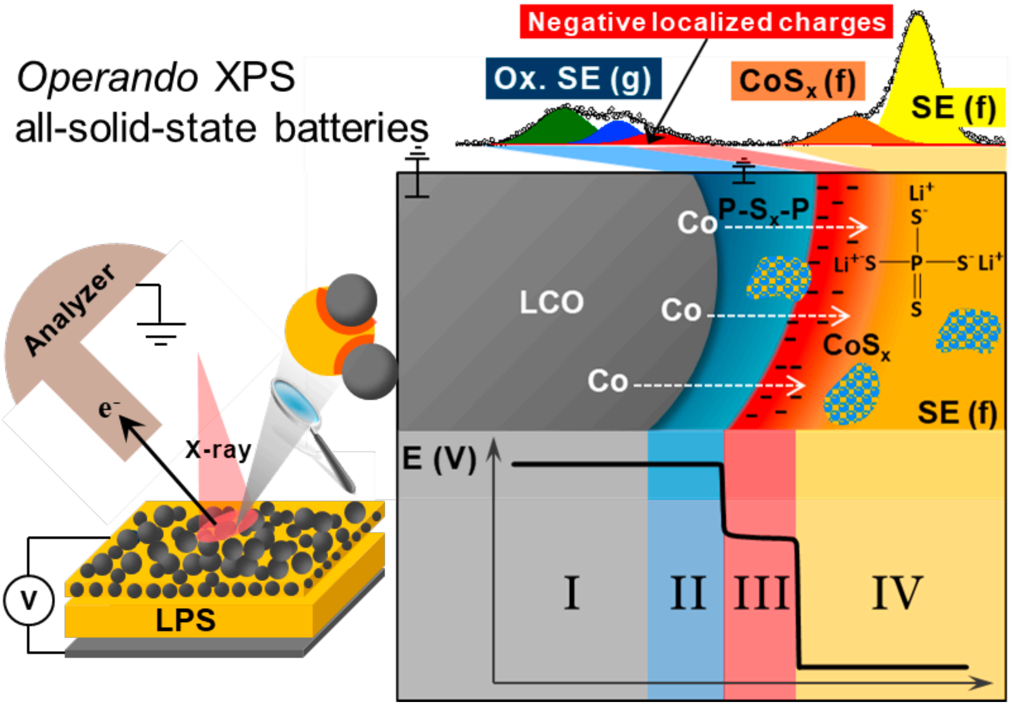

Figure 12

Schematic representation of the degradation mechanism at the LCO-LPS interface and the potential profile based on the operando XPS data. Four regions describe the potential profile across the electrified LCO-LPS interface:

(I) electronically conductive LCO particles which are grounded to the analyzer,

(II) oxidized SE in intimate contact with LCO and thus grounded to the analyzer,

(III) formation of negative localized charges and

(IV) electronically insulating SE containing diffused Co species.

diffusion of species across the SE-AMs interface is known to increase further the charge-transfer resistance.

Concerning the electronic properties of the interface, the fundamental difference between electrified solid-solid and liquid-solid interfaces lies in the electrical potential profile. In a standard model, an electrified liquid-solid interface is characterized by the existence of an electrochemical double layer leading to the formation of a potential drop region ranging from several atomic layers to tens of nm depending on the electrolyte composition and concentration.

However, although expected, little is known whether there is an analogous distribution of charges occurring across the electrified cathode AMs-SE interface. Important parameters, such as the potential profile across the interface, charge distribution, and the impact of defects have been difficult to access experimentally. Nevertheless, during battery cell cycling outside the electrochemical thermodynamic stability window of the SE, the chemistry and electronic properties of the interface become very complex, in particular, since SE degradation byproducts and transition metals inter-diffusion are likely to occur.

In this study, we address the evolution of the (electro-)chemical reactions and electronic properties of the interface in a compressed powder mixture of LCO and LPS by using the *operando* XPS electrochemical cell developed for ASSB. We carried out real-time measurements of an operating working electrode to obtain a detailed picture of the reactions occurring at the interface of an LCO-LPS. Specifically, we identify the onset of oxidation of the LPS at 2.3 V vs. $InLi_x$, which is followed by Co-ion diffusion into the LPS at 3.3 V vs. $InLi_x$ to form CoS_x species. After LPS oxidation, a localized negative charge layer is detected at the LPS-LCO interface causing a local surface overpotential of approximatively 0.9 V. Furthermore, we demonstrate that an abrupt potential variation is present at the interface between the AMs and SE, confirming previous theoretical predictions of a narrow space charge layer at the interfaces of ASSBs. Finally, with the support of X-ray photoemission electron microscopy (XPEEM), we reveal the existence of chemical reactivity between LCO and LPS, leading to the formation of reduced Co in a +2 oxidation state at the LCO surface occurring right after mixing the powder.

Publication

Reactivity and Potential Profile across the Electrochemical $LiCoO_2$–Li_3PS_4 Interface Probed by *Operando* X-ray Photoelectron Spectroscopy.
Xiaohan Wu, Marta Mirolo, Carlos A.F. Vaz,
Petr Novák, Mario El Kazzi.
ACS Appl. Mater. Interfaces 13, 42670–42681 (2021).
DOI: 10.1021/acsami.1c09605

Acknowledgement

Part of this work was performed at the Surfaces/Interfaces Microscopy (SIM) beamline of the Swiss Light Source (SLS), Paul Scherrer Institut (PSI), Villigen, Switzerland.

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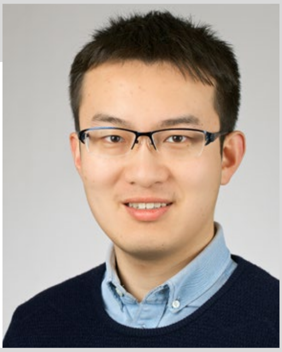
DISSERTATIONS 2021

Subsecond *Operando* X-ray Tomographic Microscopy of Liquid Water in Polymer Electrolyte Fuel Cells

Hong Xu



Ph.D. Thesis No. 27351, ETH Zürich, January 2021.

Examiners:
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Prof. Dr. M. Stampanoni, PSI Villigen/ETH Zürich
Dr. J. Eller, PSI Villigen

Abstract

Hydrogen fed polymer electrolyte fuel cells (PEFC) are expected to play a major role in a future decarbonized energy system, in particular in the mobility sector. Water management is a major limiting factor in PEFC for further increasing power density. In particular, understanding the liquid water dynamics in the GDL, which defines how the liquid water spreads in the GDL and limits the performance, is a key step to improve the performance, but also the stability and the durability of PEFC.

In his thesis, Hong explored sub-second X-ray tomographic microscopy (XTM) of PEFC for visualizing and ultimately understanding the liquid water dynamics in the GDL in close collaboration with the TOMCAT team of the Swiss Light Source. Different microscope combinations were quantified in terms of water detectability at different scan times and X-ray beam configurations, though higher magnification levels required higher X-ray intensities to be used and thus lowered the possible number of scans due to X-ray dose limitations. Thanks to these efforts, the scan time of *operando* XTM scans could be lowered by more than a factor of 10 achieving 95 % liquid water detection in the GDL at a scan time of 0.2 s and a voxel size of 2.75 µm. The identified imaging conditions and image processing parameters allowed to establish subsecond *operando* XTM as routine characterization technique.

These unique imaging capabilities were used to achieve fundamental insights into the water transport modes during PEFC startup and distinguish the contributions of capillary fingering and phase-change induced water transport at different operation temperatures. In a second systematic *operando* XTM study covering dynamic processes during GDL filling and desaturation as well as at steady state water distributions were studied. Using a variety of microstructure descriptors like water cluster size, cluster connectivity and saturation dependent effective diffusivity based on direct

numerical simulations on the segmented 3D microstructures, it was possible to explain the differences of fuel cell performance and microstructure properties of different commercial GDL materials.

Model-based Assessment of Stationary Energy Storage in Batteries and Implications for Materials Research

Simon Schneider

Ph.D. Thesis No. 27357, ETH Zürich, January 2021.

Examiners:
Prof. Dr. Petr Novák, PSI Villigen/ETH Zürich
Prof. Dr. Gabriela Hug, ETH Zürich
Mr. Christian Bauer, PSI Villigen
Dr. Tom Kober, PSI Villigen

Abstract

Large-scale energy storage is one of the prerequisites for the successful integration of renewable energy sources into the electricity supply system. In the past years, storage technologies including rechargeable lithium-ion batteries (LIBs) have therefore increasingly been deployed in numerous stationary application cases. While batteries offer several benefits, they inevitably degrade over time and use until they can no longer provide the designated service. The replacement of batteries that have reached the end of their lifetime does not only cause economic costs but also presents an environmental burden, as their manufacturing is associated with the depletion of critical resources and emission of greenhouse gases.

Thus, the viability of stationary energy storage in rechargeable batteries depends on a complex interplay of multiple parameters related to the battery chemistry, battery manufacturing, and battery use phase.

This thesis presents a new assessment framework (i) to compare the technical, economic, and environmental viability of batteries on a cell level and (ii) to analyse the deployment and operation of battery systems providing grid-level energy storage. Consistency in the assessment of different battery technologies and application cases is ensured by combining and further developing modeling approaches from a wide range of research communities, including electrochemical battery performance models, semi-empirical battery degradation models, battery cost models, life cycle assessment (LCA), and energy-economic simulation and optimization models. The developed assessment framework was used to analyze multiple case studies and business cases.

Based on the model results, this thesis provides guidelines and recommendations for further research on rechargeable batteries and the materials they are composed of.

The first part of this thesis is dedicated to the assessment of battery cells from a technical, economic, and environmental point of view. To this end, an electrochemical pseudo-two-dimensional (P2D) model is employed to assess practical specific energies of intercalation-based battery cells under varying discharge rates. Subsequently, the outputs of the P2D model are used to parameterize a bottom-up battery cell cost model and to assess greenhouse gas emissions associated with the manufacturing of battery cells from a life cycle perspective.

Serving as a case study for the modelling framework, a comparative assessment of different LIBs and sodium-ion batteries (NIBs) was performed. The model results suggest that NIBs need further improvement on the materials level in order to become competitive with LIBs. A strong inverse correlation has been found between the practical specific energy of battery cells and both their manufacturing costs and environmental impacts. By contrast, other drivers, such as cost savings due to the replacement of lithium-based materials by sodium-based ones, play only a relatively minor role under current raw material costs. NIBs are therefore most likely to become competitive with LIBs if the specific energy of NIBs can be further increased. This translates into a need for anode and cathode active materials that offer higher specific charges, higher gravimetric densities, and higher voltages.

The second part of this thesis is devoted to the techno-economic assessment of battery systems providing different stationary services, including peak shaving (PS), price arbitrage (PA), and primary control reserves (PCR) under a wide range of current and prospective electricity market conditions. The model results suggest that the provision of PS and PCR services is generally profitable under current investment costs of LIBs due to the high revenue potential of these application cases. By contrast, battery systems performing PA require both technical improvement and significant cost reduction in order to reach economic viability. On the whole, the findings of this thesis support the expectation that LIBs will continue to play a vital role in modern electricity supply systems.

An evaluation of different performance targets for battery systems shows that the research priorities vary considerably depending on the application case. This underlines that there is no one-target-fits-all strategy for the further development of today's LIBs. The model results suggest, however, that the second life use of batteries, which is enabled by extending their lifetime beyond 80 % remaining capacity, is an important development target for all applications cases considered in this thesis. In addition, the following technical improvements are desirable, as they would improve the economics of stationary batteries:

1) Longer cycle lifetimes would be particularly beneficial for batteries that perform PA on electricity markets with high price volatility.
2) Higher energy efficiency would be desirable for batteries that perform PA on electricity markets with low price volatility.
3) The provision of PCR requires batteries to perform many shallow charge and discharge cycles. Hence, depending on the LIB technology used, the cycle stability should be further improved to ensure that they can sustain a high number of shallow cycles.

Overall, the holistic modeling approach presented in this thesis provides new insights into desirable improvements of today's rechargeable batteries.

Liquid and Vapour Phase Water Transport Mechanisms in Fuel Cell Gas Diffusion Layers

Adrian Mularczyk

Ph.D. Thesis No. 27382, ETH Zürich, January 2021.

Examiners:
Prof. Dr. T.J. Schmidt, PSI Villigen/ETH Zürich
Prof. Dr. D. Poulikakos, ETH Zürich
Dr. J. Eller, PSI Villigen

Abstract

The removal of product water of polymer electrolyte fuel cells (PEFC) can take place either in the liquid phase via capillary fingering or in vapor phase by phase change processes. A well-adjusted water management of the cell requires a balance between keeping the membrane well humidified for high proton conductivity and avoiding unwanted liquid water accumulations in the gas diffusion layer (GDL) to maintain undistorted delivery of reactants to the catalyst layers.

In his thesis, Adrian investigated the transport of liquid water through the GDL in PEFCs, both in liquid and in gas phase, by X-ray tomographic microscopy. Thanks to recent developments in 4D tomography, it was possible to shed light on the percolation of water clusters through the GDL from the catalyst layer towards the flow field channels, the formation and release of water droplets in the gas channels, both via *ex situ* and *operando* imaging experiments.

Thanks to a collaboration with researchers at Imperial College London (London, UK) and Zhejiang University (Hangzhou, China) it was possible to extract the curvature of the water-air interface on the pore scale and to determine for the first time the capillary pressure in water clusters of a running PEFC. The 4D tomographic data-sets were furthermore used to validate different two-phase computational fluid dynamic models of research groups in Germany, Sweden and UK.

In the second part of the thesis, the limiting factors of the evaporation of liquid water in the GDL were identified by a combined experimental and imaging study. Dedicated *ex situ* experiments allowed to measure the evaporation rate of water at a broad range of boundary conditions. In collaboration with researchers at the EPFL in Lausanne that implemented heat and mass transport simulations on the pores scale, it was possible to correlate the macroscopic

evaporation rates to the features observed at the pore scale and the complex interplay of mass transport in the GDL and in the flow field channels for commercial GDL materials, but also for GDLs with dedicated separation of hydrophilic and hydrophobic zones. These insights are of particular interest for the improvement of the evaporation cooling principle that is being developed at PSI.

Efficient Carbon Dioxide Conversion in Bipolar and Bipolar-like Electrolyser Systems

Bernhard Pribyl-Kranewitter

Ph.D. Thesis No. 27481, ETH Zürich, March 2021.

Examiners:
Prof. Dr. T.J. Schmidt, PSI Villigen/ETH Zürich
Prof. Dr. Christophe Copéret, ETH Zürich
Dr. Alexandra Beard, PSI Villigen

Abstract

The consequences of climate change are forecast to worsen if the CO_2-emissions causing the greenhouse effect are not drastically reduced. One of the means to attain this is by electrochemically reducing captured CO_2 into value-added chemicals.

Carbon monoxide (CO) features the highest economic potential among the possible products of CO_2-electroreduction, and can be produced in all-alkaline electrochemical systems exhibiting promising current densities and CO-selectivities (≤ 600 mA/cm^2 and $\approx 95\,\%$, respectively). However, these all-alkaline cells suffer from a cathode-to-anode CO_2-transport that decreases their net CO_2-consumption and efficiency, and renders the systems more complex. This CO_2-transport could be prevented by substituting the alkaline electrolytes discussed above with bipolar membranes that have barely been investigated for cell-level CO_2-electroreduction. Thus, the goal of this thesis was to develop and optimize a CO_2-to-CO electrolysis cell based on the use of such bipolar membranes.

The work started by developing a procedure for the online detection and quantification of the CO_2-reduction species produced in the cell. This product analysis was conducted using mass spectrometry, and required a calibration method to deconvolute the contributions of CO_2, CO and N_2 to the measured signals. This was followed by showing for the first time that the CO_2-transport observed in all-alkaline cells is suppressed with bipolar membranes. The subsequent optimization of this bipolar cell led to a maximum CO partial current density of 127 mA/cm^2 with a selectivity of 93 % at an ohmically-corrected cell voltage of 2.9 V.

Finally, a techno-economic analysis was performed to assess the economic and environmental potential of the developed technology and compare it to other currently employed

CO_2-electrolyser configurations. This analysis showed that the largest barriers to the commercialization of this technology are the high cost of membranes and catalysts, along with the system complexity derived from the transport of CO_2 in all-alkaline systems, which can be circumvented with the bipolar membrane approach presented in this work.

In summary, this doctoral thesis has led to an improved understanding of low-temperature CO_2-electrolysis by providing an analytical background for online measurements of gas mixtures, mitigating CO_2-transport by adopting a bipolar membrane, and shedding light on the impact of cell design on the process' economic and environmental potential.

Towards High Efficiency Water Electrolysis with Thin Membranes and Elevated Temperature Operation

Steffen Garbe

Ph.D. Thesis No. 27643, ETH Zürich, May 2021.

Examiners:
Prof. Dr. T.J. Schmidt, PSI Villigen/ETH Zürich
Prof. Dr. M. Lukatskaya, ETH Zürich
PD Dr. L. Gubler, PSI Villigen

Abstract

Decarbonization of the energy system across different sectors (Power-to-X) relies heavily on the availability of low-cost hydrogen produced from renewable power by water electrolysis. The increase in conversion efficiency of H_2 production in a polymer electrolyte water electrolysis (PEWE) cell is imperative to lower the cost of green hydrogen to be competitive to blue and grey hydrogen in industrial applications and mobility.

In this thesis project, it is demonstrated that the efficiency of H_2 production on the cell-level can be increased by 14 % at a current density of 3 A/cm^2 using a thin (50 µm) standard membrane material (Nafion™) and an elevated operating temperature of 120°C. This, however, comes at the cost of an increase in gas permeability across the membrane, in particular H_2 crossover to the anode side. This can lead to the formation of an explosive gas mixture. The increased gas crossover can be successfully mitigated through the incorporation of a Pt gas recombination catalyst. It allows the dissolved H_2 and O_2 in the membrane to react to water.

A novel method for the doping of the membrane with Pt-nanoparticles was developed: the membrane is partially ion-exchanged with Pt-ions, followed by a reduction step using H_2 gas, which leads to a homogeneous deposition of Pt-particles throughout the membrane. A patent application describing this invention has been filed. With a recombination catalyst loading of 0.06 mg(Pt)/cm^2, a significant reduction of H_2 accumulation in the O_2 product stream could be achieved, which enhances the operating range of the electrolyzer.

Elevated temperature operation, however, also comes at the cost of an increased rate of membrane degradation. Cell operation at 100°C for 300 h showed a notable degradation of the membrane as well as the anode catalyst. The

activation energy for membrane degradation was measured at ~60 kJ/mol, which leads to a reduction of the projected lifetime at 100°C of a factor of 10 compared to 60°C, which is today's standard operating temperature of an electrolyzer. This calls for the further development of membrane materials for water electrolysis applications, which are currently underway in the Laboratory.

Understanding Degradation Mechanisms of Sulfide-based All-solid-state Lithium-ion Batteries at Multiple Length Scales

Laura Höltschi

Ph.D. Thesis No. 27931, ETH Zürich, September 2021.

Examiners:
Prof. Dr. Petr Novák, PSI Villigen/ETH Zürich
Prof. Dr. M. Kovalenko, ETH Zürich
Dr. Mario El Kazzi, PSI Villigen

Abstract

All-solid-state Li-ion batteries are currently receiving more attention due to their non flammable solid components and promising electrochemical performance compared to conventional batteries with liquid organic electrolytes. Nevertheless, several fundamental and engineering challenges are still limiting their commercialization. Among them are

1) the narrow electrochemical stability window,
2) the reactivity of the solid electrolyte with electrode materials,
3) the difficult preparation of the composite electrode, especially the inhomogeneous distribution and the poor contact between the solid electrolyte and active materials, and
4) the mechanical degradation caused by the volume changes during cycling.

In this thesis, both graphite and commercial cathode materials $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$ (NMC111) and $LiCoO_2$ (LCO) were investigated in combination with sulfide-based solid electrolytes (LPS).

In the first part, the excellent cycling stability of graphite is demonstrated. A suite of bulk and surface analytical techniques was applied to shed light on the factors limiting fast cycling. Among those techniques were *operando* X-ray diffraction, X-ray tomography, Raman microscopy, *operando* X-ray photoelectron spectroscopy (XPS) and X-ray absorption spectroscopy (XAS).

In the second part, the oxidation of solid electrolyte and the formation of degradation products at the interface between the LPS and NCM111 were analyzed by combining measurements with tender and soft X-rays. It was possible to follow the electrochemical decomposition of the LPS electrolyte by probing the S and P K-edges XAS spectra, while monitoring the reactions at

the surface of the NCM111 active material was performed by acquiring the L-edges of Ni, Co, and Mn.

From Stochastic to Deterministic Transport – Advanced Gas Diffusion Layer Materials for Polymer Electrolyte Fuel Cells

Christoph Csoklich

Ph.D. Thesis No. 28099, ETH Zürich, December 2021.

Examiners:
Prof. Dr. T.J. Schmidt, PSI Villigen/ETH Zürich
Prof. Dr. T. Lippert, PSI Villigen/ETH Zürich
Prof. Dr. G. Jeschke, PSI Villigen/ETH Zürich
Dr. F.N. Büchi, PSI Villigen

Abstract

In a polymer electrolyte fuel cell, hydrogen and oxygen are supplied through gas channels from both sides of the polymer electrolyte membrane at the center. The gas diffusion layers (GDLs) distribute the feed gases homogeneously across the catalyst layers located at the membrane interfaces. When product water accumulates in the porous gas diffusion layers, the oxygen transport resistance to the cathode catalyst increases and thus prevents current PEFC technology from reaching higher current and power densities. This thesis investigated water and gas transport in conventional and advanced GDL materials and highlights the benefits of deterministic structures, as compared to today's stochastic materials.

The first part investigated the potentials and limits of modifying conventional carbon fiber GDLs by laser perforation. This technique introduces deterministic, straight channels that locally direct liquid water. A wide variety of patterns was studied to evaluate the influence of the pattern geometry on fluid transport through the porous materials. With this approach, a suitable descriptor was found that accurately relates the optimum perforation density with fuel cell performance. The competing effects of improved water transport (and thus reduced oxygen transport resistance) and increased membrane drying (by increasing the effective diffusivity) limit the maximum power density increase to 20 %.

Furthermore, the water transport through the laser-structured GDL of one specific perforation pattern was investigated with sub-second *operando* XTM. The comparison with a conventional GDL showed that the total water saturation is similar for both gas diffusion layers. However, the perforations distribute product water more evenly, reducing the water saturation close to the catalyst layer, leading to increased current densities for the perforated GDL. These studies

show that the introduced deterministic structure can direct and improve water transport in conventional, stochastic GDLs and thereby increase fuel cell power density.

The second part followed a different approach and investigates the potential of novel, fully deterministic GDLs. Woven and gold-coated PET fabrics offer unique tuning properties, enabling a rational, bottom-up design of transport optimized gas diffusion layers. In a first study, these woven materials are successfully used for the first time in a PEFC, and their superior performance under cold and saturated operating conditions was analyzed. The water transport, investigated again by *operando* XTM, is fundamentally different. Water moves much faster through the structure, as it needs to pass only one pore throat before reaching the gas channel. The last study focuses on the thermal conductivity of GDLs, as heat removal is another essential transport duty. Different thickness of gold coatings enable changing the thermal conductivity independently of the pore structure. The strongly reduced thermal conductivity is unexpectedly not a performance limiting factor. It leads to significant thermal gradients across the GDL and thus drives a heat pipe through water evaporation. Thus, more water is transported in the vapor phase, and the GDL structure stays dry for improved oxygen transport. In comparison with a conventional GDL it was shown that the structure is the key to the improved performance and that the thermal conductivity, different by orders of magnitude, causes just smaller differences between the materials.

Both approaches emphasize the benefits of designed, deterministic GDL structures that locally reduce liquid water saturation and facilitate oxygen transport. Future GDL development can therefore focus on optimizing the structure and tuning it to the locally heterogeneous operating conditions present in a fuel cell to further increase PEFC power density.

THE ELECTROCHEMISTRY LABORATORY

FACTS & FIGURES

STRUCTURE 2021

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Dr. M. El Kazzi

Battery Electrodes and Cells

Dr. S. Trabesinger

Membranes and Electrochemical Cells

PD Dr. L. Gubler

Fuel Cell Systems and Diagnostics

Dr. F.N. Büchi i.P.

Electrocatalysis and Interfaces

Prof. Dr. T.J. Schmidt i.P.

Neutron Radiography of Electrochemical Systems

Dr. P. Boillat

PERSONNEL 2021

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PAUL SCHERRER INSTITUT

Members of the Electrochemistry Laboratory in 2021.

AWARDS 2021

Dr. Tobias Schuler

PSI Impuls Award

Towards a Generic Understanding of Porous Transport Layers in Polymer Electrolyte Water Electrolysis.

Event: Impuls Award Ceremony 2021
November 25, 2021
Paul Scherrer Institut, Villigen, Switzerland

BACHELOR AND MASTER STUDENTS

Matteo Lecchi Politecnico di Milano, Milano, Italy

Modification of proton exchange membranes with antioxidant and/or performance boosting additives for hydrogen fuel cell membranes

September 2020 – March 2021 (Membranes and Electrochemical Cells)

Moritz Bohn Technical University Munich, Germany

Gas-phase surface modification of solid electrolyte materials

September 2020 – January 2021 (Battery Materials and Diagnostics)

Gian Müller ETH Zürich, Switzerland

Development of an operando X-ray imaging cell for polymer electrolyte water electrolysis

September 2020 – January 2021 (Fuel Cell Systems and Diagnostics)

Emil Skoglund University of Mälardalen, Västerås, Sweden

A numerical analysis of heat transfer and species transport in polymer electrolyte fuel cells

December 2020 – June 2021 (Fuel Cell Systems and Diagnostics)

Michail Ntalis Aristotle University of Thessaloniki, Greece

GDLs modification by radiation-grafting for improved freeze-start capability in PEFC

March – July 2021 (Neutron Radiography of Electrochemical Systems)

Linfeng Xu ETH Zürich, Switzerland

3D structural reconstruction of PEFC catalyst layer and microporous layer morphology from SAXS data

March – September 2021 (Fuel Cell Systems and Diagnostics)

Quinhao Chen Technical University Munich, Germany

Palladium-based catalysts for CO_2-and NO_2-electroreduction

March – July 2021 (Electrocatalysis and Interfaces)

Joel Handschin ETH Zürich, Switzerland

Investigation on $NaNbO_3$ as a coating material for P2-$Na_{0.67}Mn_{0.6}Fe_{0.25}Al_{0.15}O_2$ cathode material for Na-ion batteries to improve cycling performance

April – June 2021 (Battery Materials and Diagnostics)

Bagdaulet Shugay Nazarbayev University, Nur-Sultan, Kazakhstan

Preventing ice propagation in fuel cells by segmented gas diffusion layers

September – November 2021 (Neutron Radiography of Electrochemical Systems)

Gian Müller ETH Zürich, Switzerland

ECSA estimation and CO_2 reduction on Au_3Cu and AuCu aerogels

September 2021 – January 2022 (Electrocatalysis and Interfaces)

Laura King

University of Edinburgh, United Kingdom

Enabling anode-free lithium metal cells for high energy density batteries

September 2021 – May 2022 (Battery Electrodes and Cells)

Bowen Li

ETH Zürich, Switzerland

Surface coatings for layered cathode materials for Na-ion batteries and Li-ion batteries in order to improve the stability towards electrolytes

October 2021 – January 2022 (Battery Materials and Diagnostics)

Michael Neuwirt

ETH Zürich, Switzerland

Influence of process parameters on the properties of PBI membranes for use in redox flow batteries

November 2021 – January 2022 (Membranes and Electrochemical Cells)

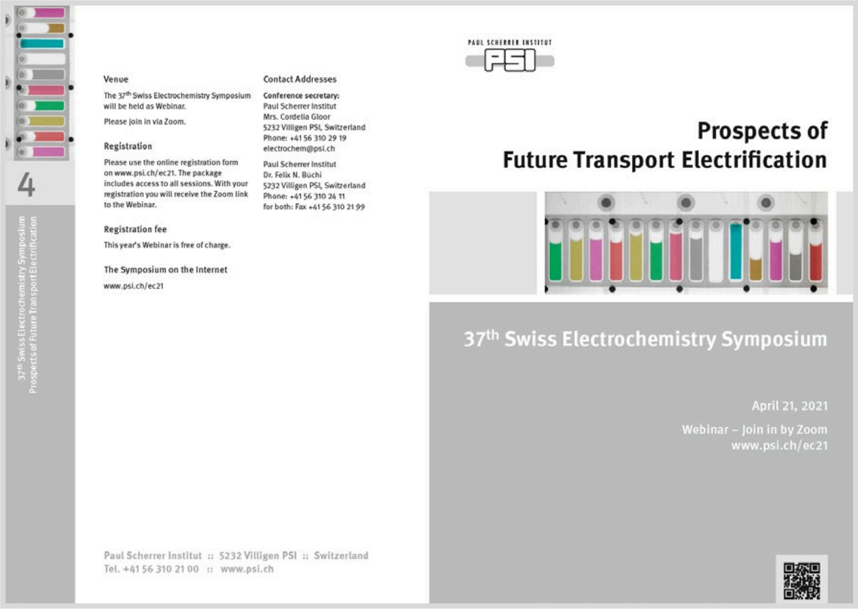
CONFERENCES – SYMPOSIA

37th Swiss Electrochemistry Symposium

April 21, 2021
Webinar via Zoom

Prospects of Future Transport Electrification

Organizers	Felix N. Büchi, Lorenz Gubler, Sigita Trabesinger, Cordelia Gloor (Electrochemistry Laboratory)
Contributions from (in order of appearance)	Konstantinos Boulouchos, ETH Zürich, Switzerland Hubert Gasteiger, Technical University of Munich, Garching, Germany Patrick Stadelmann, H_2 Energy AG, Glattpark-Opfikon, Switzerland Ralph-Uwe Dietrich, German Aerospace Center (DLR), Stuttgart, Germany Jürgen Janek, Justus-Liebig-Universität, Giessen, Germany Andrea Vezzini, Bern University of Applied Sciences, Biel, Switzerland Tobias Schmidt, ETH Zürich, Switzerland



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37th Swiss Electrochemistry Symposium
Prospects of Future Transport Electrification

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Prospects of Future Transport Electrification

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The Electrochemistry Laboratory had the pleasure to collaborate with the following industrial partners during the year 2021:

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Novel catalysts for CO_2 Hydrogenation and Electro-reduction

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Actions

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Teaching Activities

Teaching

Dr. E. Fabbri, PD Dr. L. Gubler, Dr. J. Herranz	*Electrochemical Energy Conversion & Storage Technologies* ETH Zürich, FS 2021.
Dr. E. Fabbri, PD Dr. L. Gubler, Dr. J. Herranz, Dr. S. Trabesinger	*Hands-on Electrochemistry for Energy Storage and Conversion Applications* ETH Zürich, HS 2021.
PD Dr. L. Gubler	*Electrochemistry: Fundamentals, Cells & Applications* ETH Zürich, HS 2021.
Prof. Dr. T.J. Schmidt	*Physical Electrochemistry & Electrocatalysis* ETH Zürich, FS 2021.

Contributions to Courses

PD Dr. L. Gubler	*Energy Storage Systems* Lucerne University of Applied Sciences and Arts, March 31, 2021.
Dr. J. Herranz, Dr. S. Trabesinger	*CAS ETH Zürich Mobilität der Zukunft* ETH Zürich, HS 2021.

Publications

Peer Reviewed Papers

C. Baeumer, J. Li, Q. Lu, A.Y.-L. Liang, L. Jin, H.P. Martins, T. Duchoň, M. Glöß, S.M. Gericke, M.A. Wohlgemuth, M. Giesen, E.E. Penn, R. Dittmann, F. Gunkel, R. Waser, M. Bajdich, S. Nemšák, J.T. Mefford, W.C. Chueh	*Tuning electrochemically driven surface transformation in atomically flat LaNiO$_3$ thin films for enhanced water electrolysis* Nature Materials **20**, 674–682 (2021). DOI: 10.1038/s41563-020-00877-1
D. Baster, Ł. Kondracki, E. Oveisi, S. Trabesinger, H.H. Girault	*Prussian Blue Analogue—Sodium–Vanadium Hexacyanoferrate as a Cathode Material for Na-Ion Batteries* ACS Applied Energy Materials **4**, 9758–9765 (2021). DOI: 10.1021/acsaem.1c01832
C.E. Beall, E. Fabbri, T.J. Schmidt	*Perovskite Oxide Based Electrodes for the Oxygen Reduction and Evolution Reactions: The Underlying Mechanism* ACS Catalysis **11**, 3094–3114 (2021). DOI: 10.1021/acscatal.0c04473
A. Boucly, L. Artiglia, E. Fabbri, D. Palagin, D. Aegerter, D. Pergolesi, Z. Novotny, N. Comini, J.T. Diulus, T. Huthwelker, M. Ammann, T.J. Schmidt	*Direct evidence of cobalt oxyhydroxide formation on a La$_{0.2}$Sr$_{0.8}$CoO$_3$ perovskite water splitting catalyst* Journal of Materials Chemistry A (2021). DOI: 10.1039/D1TA04957G
M. Bührer, H. Xu, A.A. Hendriksen, F.N. Büchi, J. Eller, M. Stampanoni, F. Marone	*Deep learning based classification of dynamic processes in time-resolved X-ray tomographic microscopy* Scientific Reports **11**, 24174 (2021). DOI: 10.1038/s41598-021-03546-8

T.A. Butcher, L. Prendeville, A. Rafferty, P. Trtik, P. Boillat, J.M.D. Coey	*Neutron Imaging of Paramagnetic Ions: Electrosorption by Carbon Aerogels and Macroscopic Magnetic Forces* The Journal of Physical Chemistry C **125**, 21831–21839 (2021). DOI: 10.1021/acs.jpcc.1c06031
Y.-C. Chen, A. Berger, S. De Angelis, T. Schuler, M. Bozzetti, J. Eller, V. Tileli, T.J. Schmidt, F.N. Büchi	*A Method for Spatial Quantification of Water in Microporous Layers of Polymer Electrolyte Fuel Cells by X-ray Tomographic Microscopy* ACS Applied Materials & Interfaces **13**, 16227–16237 (2021). DOI: 10.1021/acsami.0c22358
C. Csoklich, R. Steim, F. Marone, T.J. Schmidt, F.N. Büchi	*Gas Diffusion Layers with Deterministic Structure for High Performance Polymer Electrolyte Fuel Cells* ACS Applied Materials & Interfaces **13**, 9908–9918 (2021). DOI: 10.1021/acsami.0c20896
C. Csoklich, H. Xu, F. Marone, T.J. Schmidt, F.N. Büchi	*Laser Structured Gas Diffusion Layers for Improved Water Transport and Fuel Cell Performance* ACS Applied Energy Materials **4**, 12808–12818 (2021). DOI: 10.1021/acsaem.1c02454
S. De Angelis, H.P. Nguyen, S. Nagao, K. Kishita, F. Marone, F.N. Büchi	*In-situ observation of water evaporation in exhaust gas catalyst via sub-micron and sub-second X-ray tomography* Chemical Engineering Science: X, **10**, 100091 (2021). DOI: 10.1016/j.cesx.2021.100091
S. De Angelis, T. Schuler, M.A. Charalambous, F. Marone, T.J. Schmidt, F.N. Büchi	*Unraveling two-phase transport in porous transport layer materials for polymer electrolyte water electrolysis* J. Mater. Chem. A, **9**, 22102–22113 (2021). DOI: 10.1039/D1TA03379D
T. de Wild, T. Németh, T.M. Nolte, T.J. Schmidt, T. Nauser, L. Gubler	*Possible Repair Mechanism for Hydrocarbon-Based Ionomers Following Damage by Radical Attack* Journal of The Electrochemical Society **168**, 054514 (2021). DOI: 10.1149/1945-7111/abf9be
P. Desai, J. Huang, H. Hijazi, L. Zhang, S. Mariyappan, J.-M. Tarascon	*Deciphering Interfacial Reactions via Optical Sensing to Tune the Interphase Chemistry for Optimized Na-Ion Electrolyte Formulation* Advanced Energy Materials **11**, 2101490 (2021). DOI: 10.1002/aenm.202101490

J.S. Diercks, B. Pribyl-Kranewitter, J. Herranz, P. Chauhan, A. Faisnel, T.J. Schmidt	*An Online Gas Chromatography Cell Setup for Accurate CO_2-Electroreduction Product Quantification* Journal of The Electrochemical Society **168**, 064504 (2021). DOI: 10.1149/1945-7111/ac0363
F. Du, J.A. Hirschfeld, X. Huang, K. Joswiak, T.A. Dao, A. Bauer, T.J. Schmidt, A. Oranidi	*Simulative Investigation on Local Hydrogen Starvation in PEMFCs: Influence of Water Transport and Humidity Conditions* Journal of The Electrochemical Society **168**, 074504 (2021). DOI: 10.1149/1945-7111/ac148e
J.C. Duburg, K. Azizi, S. Primdahl, H.A. Hjuler, E. Zanzola, T.J. Schmidt, L. Gubler	*Composite Polybenzimidazole Membrane with High Capacity Retention for Vanadium Redox Flow Batteries* Molecules **26**, 1679 (2021). DOI: 10.3390/molecules26061679
K. Ebner, L. Ni, V.A. Saveleva, B.P. Le Monnier, A.H. Clark, F. Krumeich, M. Nachtegaal, J. Luterbacher, U.I. Kramm, T.J. Schmidt, J. Herranz	*^{57}Fe-Enrichment effect on the composition and performance of Fe-based O_2-reduction electrocatalysts* Physical Chemistry Chemical Physics **23**, 9147–9157 (2021). DOI: 10.1039/D1CP00707F
K. Elbouazzaoui, M. Srout, I. Saadoune, L. Bih, H.B.E.N. Youcef, M. Dahbi, M. Mansori	*NASICON-type $Li_{0.5}M_{0.5}Ti_{1.5}Fe_{0.5}(PO_4)_3$ (M = Mn, Co, Mg) phosphates as electrode materials for lithium-ion batteries* Electrochimica Acta **399**, 139438 (2021). DOI: 10.1016/j.electacta.2021.139438
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S. Garbe, J. Futter, T.J. Schmidt, L. Gubler	*Insight into Elevated Temperature and Thin Membrane Application for High Efficiency in Polymer Electrolyte Water Electrolysis* Electrochimica Acta **377**, 138046 (2021). DOI: 10.1016/j.electacta.2021.138046
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A. Heinritz, T. Binninger, A. Patru, T.J. Schmidt	*Asymmetric Butler–Volmer Kinetics of the Electrochemical Ce (III)/Ce (IV) Redox Couple on Polycrystalline Au Electrodes in Sulfuric Acid and the Dissociation Field Effect* ACS Catalysis **11**, 8140–8154 (2021). DOI: 10.1021/acscatal.0c04587
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K. Iwase, K. Ebner, J.S. Diercks, V.A. Saveleva, S. Ünsal, F. Krumeich, T. Harada, I. Honma, S. Nakanishi, K. Kamiya, T.J. Schmidt, J. Herranz	*Effect of Cobalt Speciation and the Graphitization of the Carbon Matrix on the CO_2 Electroreduction Activity of Co/N-Doped Carbon Materials* ACS Applied Materials & Interfaces **13**, 15122–15131 (2021). DOI: 10.1021/acsami.0c21920
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ACS Applied Materials & Interfaces **13**, 2547–2557 (2021).
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A. Mularczyk, A. Michalski, M. Striednig, R. Herrendörfer, T.J. Schmidt, F.N. Büchi, J. Eller	*Mass Transport Limitations of Water Evaporation in Polymer Electrolyte Fuel Cell Gas Diffusion Layers* Energies **14**, 2967 (2021). DOI: https://doi.org/10.3390/en14102967
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H.Q. Pham, M.T. Nguyen, M. Tarik, M. El Kazzi, S. Trabesinger	*Cross-Talk-Suppressing Electrolyte Additive Enabling High Voltage Performance of Ni-Rich Layered Oxides in Li-Ion Batteries* ChemSusChem **14**, 2461–2474 (2021). DOI: 10.1002/cssc.202100511
R.K. Pittkowski, D.F. Abbott, R. Nebel, S. Divanis, E. Fabbri, I.E. Castelli, T.J. Schmidt, J. Rossmeisl, P. Krtil	*Synergistic effects in oxygen evolution activity of mixed iridium-ruthenium pyrochlores* Electrochimica Acta **366**, 137327 (2021). DOI:10.1016/j.electacta.2020.137327
B. Pribyl-Kranewitter, A. Patru, T. Schuler, N. Diklic, T.J. Schmidt	*Investigation and Optimization of Operating Conditions for Low-Temperature CO_2 Reduction to CO in a Forward-Bias Bipolar-Membrane Electrolyzer* Journal of The Electrochemical Society **168**, 043506 (2021). DOI: 10.1149/1945-7111/abf063

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Unraveling the Voltage-Dependent Oxidation Mechanisms of Poly(Ethylene Oxide)-Based Solid Electrolytes for Solid-State Batteries
Advanced Materials Interfaces, 2100704 (2021).
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Nature Materials **20**, 353–361 (2021).
DOI: 10.1038/s41563-020-00870-8

E. Winter, T.J. Schmidt, S. Trabesinger
Identifying Pitfalls in Lithium Metal Battery Characterization
Batteries & Supercaps **4**, 1–14 (2021).
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X. Wu, M. Mirolo, C.A.F. Vaz, P. Novák, M. El Kazzi	*Reactivity and Potential Profile across the Electrochemical $LiCoO_2$–Li_3PS_4 Interface Probed by Operando X-ray Photoelectron Spectroscopy* ACS Applied Materials & Interfaces **13**, 42670–42681 (2021). DOI: 10.1021/acsami.1c096057
H. Xu, M. Bührer, F. Marone, T.J. Schmidt, F.N. Büchi, J. Eller	*Effects of Gas Diffusion Layer Substrates on PEFC Water Management: Part I. Operando Liquid Water Saturation and Gas Diffusion Properties* Journal of The Electrochemical Society **168**, 074505 (2021). DOI: 10.1149/1945-7111/ac1035
H. Xu, S. Nagashima, H.P. Nguyen, K. Kishita, F. Marone, F.N. Büchi, J. Eller	*Temperature dependent water transport mechanism in gas diffusion layers revealed by subsecond operando X-ray tomographic microscopy* Journal of Power Sources **490**, 229492 (2021). DOI: 10.1016/j.jpowsour.2021.229492
L. Zhang, C. Tsolakidou, S. Mariyappan, J.-M. Tarascon, S. Trabesinger	*Unraveling gas evolution in sodium batteries by online electrochemical mass spectrometry* Energy Storage Materials **42**, 12–21 (2021). DOI: 10.1016/j.ensm.2021.07.005
T. Zhou, Y. Zhao, M. El Kazzi, J.W. Choi, A. Coskun	*Stable Solid Electrolyte Interphase Formation Induced by Monoquat-Based Anchoring in Lithium Metal Batteries* ACS Energy Letters **6**, 1711–1718 (2021). DOI: 10.1021/acsenergylett.1c00274
M. Zlobinski, T. Schuler, F.N. Büchi, T.J. Schmidt, P. Boillat	*Elucidation of Fluid Streamlining in Multi-Layered Porous Transport Layers for Polymer Electrolyte Water Electrolyzers by Operando Neutron Radiography* Journal of The Electrochemical Society **168**, 014505 (2021). DOI: 10.1149/1945-7111/abcf19

Talks

Invited Talks

F.N. Büchi	*Wasserstoff und Brennstoffzellen Fahrzeuge* European Power Network, EPN Tagung, Olten, Virtual, April 29, 2021.
F.N. Büchi	*Tomographic analysis of porous transport layers in PEM fuel cells and electrolyzers* Departmental Seminar, Department of Chemical and Biomolecular Engineering, University of California at Irvine, Virtual, June 4, 2021.
F.N. Büchi	*Towards understanding of the key properties of porous transport layers* Workshop PRETZEL and NEPTUNE FCH2 projects, Virtual, June 17, 2021.
F.N. Büchi	*Material-Entwicklung für PEM Elektrolyse* Deutsch-Schweizer H_2-Forum, Konstanz, Germany, September 28, 2021.
F.N. Büchi	*Tomographic analysis of water in PEFC porous Materials* Materials Research Meeting 2021 (MRM2021), Yokohama, Japan, Virtual, December 14, 2021.
M. El Kazzi	*Recent Advances in Operando X-Ray Photoelectron/Absorption Spectroscopy to Probe the Electrolyte/Electrode Interfaces in All-Solid-State Batteries* ACS Fall 2021, Atlanta, USA, Virtual, August 22–26, 2021.
M. El Kazzi	*(Electro-)Chemical and Electronic Properties of $LiCoO_2$-Li_3PS_4 Interface Probed by Operando X-Ray Photoelectron Spectroscopy* 72nd Annual Meeting of the International Society of Electrochemistry, Jeju Island, Korea, Virtual, August 29 – September 3, 2021.
E. Fabbri	*Towards the understanding of water splitting catalysts under operando conditions* Italian Virtual Workshop on Fuel Cells (IVWFC 2021), March 19, 2021.

E. Fabbri	*Development of catalyst materials for the electrochemical splitting of water via in-situ/operando synchrotron techniques* University of Zurich, Department of Chemistry, Web seminar, April 27, 2021.
E. Fabbri	*Materials developments for energy storage through water electrolysis: advanced characterization methods and design principles for oxide electrocatalysts* University of Basel, Department of Chemistry, Web seminar, April 30, 2021.
E. Fabbri	*Development of catalyst materials for the electrochemical splitting of water via in-situ/operando synchrotron techniques* Nanyang Technological University, Web seminar, May 6, 2021.
L. Gubler	*Membrane Requirements and Opportunities for Next-Generation Water Electrolyzers* 3rd International Conference on Electrolysis (ICE), Virtual, June 18, 2021.
L. Gubler	*Technische Optionen der Produktion von grünem Wasserstoff in der Schweiz* Swiss Association of Energy Economics (SAEE), Webinar, July 1, 2021.
L. Gubler	*Fuel Cells & Electrolyzers: Some Insights from a Materials Science Point of View* ETH Materials Colloquium, Webinar, September 1, 2021.
L. Gubler	*Electrochemical Technologies in the Context of the Energy Transition* University of Science and Technology (UST), Korea, Global Mentoring Conference, Online, October 1, 2021.
L. Gubler	*Membrane Design & Engineering Principles for Next-Generation Water Electrolysers* 2nd Edition Next Generation Electrolysers, Online, December 7–8, 2021.
T.J.Schmidt	*Sustainable Industrial Processes – Novel Pathways in CO_2 Electrochemistry* BASF Star Community Week, Online, March 16, 2021.

T.J.Schmidt	*Gasversorgung und Klimaneutralität: Unmöglich oder unentbehrlich?* Stadtwerkekongress, Online, March 26, 2021.
T.J.Schmidt	*Electrochemical CO_2 Reduction to CO: From Materials to Cell Level Development* Solcat21 (Photo-) Electrocatalysis: From the Atomistic to System Scale, Online, October 18–21, 2021.
S. Trabesinger	*Battery Electrodes and Cells Group at PSI* iBAT Battery Tuesdays, Virtual, May 4, 2021.
S. Trabesinger	*Keynote: Tracking Gas Evolution in Na-ion Battery by Online Electrochemical Mass Spectrometry* ISE-2021 72nd Annual Meeting of the International Society of Electrochemistry, Jeju Island, Korea, Virtual, August 29 – September 3, 2021.
S. Trabesinger	*Performance and Interfaces of Graphite Anodes with Capacity-Enhancing Electrode Additives* Batteries Sweden Workshop «Battery chemistries – from Generation 3 to 5», Virtual, November 17, 2021.

Contributed Talks

D. Aegerter, E. Fabbri, S. Yüzbasi, T. Graule, T.J. Schmidt	*Flame Spray Synthesized Co-based Electrocatalysts for Oxygen Evolution Reaction in Alkaline Media* EFCF-2021 European Fuel Cell Forum, Online, June 29 – July 2, 2021.
K. Aliyah, L. Gubler, J. Eller	*Validation of Small Angle X-ray Scattering Technique for Catalyst Layer Saturation Determination* ModVal-2021 17th Symposium on Modeling and Experimental Validation of Electrochemical Energy Technologies, Online, April 21–22, 2021.
K. Aliyah, L. Gubler, J. Eller	Progress towards SAXS-based PEFC catalyst layer saturation determination InterPore2021 13th Annual Meeting, Online, May 31 – June 4, 2021.

K. Aliyah, L. Gubler, J. Eller	*Mechanistic Insight of PEFC Catalyst Layer Saturation via Small Angle X-ray Scattering* EFCF-2021 European Fuel Cell Forum, Online, June 29 – July 2, 2021.
K. Aliyah, L. Gubler, J. Eller	*PEFC Catalyst Layer Saturation Determination Using Representative Structure Modeling via Small Angle X-Ray Scattering* ECS-2021 240th The Electrochemical Society Meeting, Online, October 10–14, 2021.
E.R. Carreon-Ruiz, G. Burca, M.S. Cochet, P.M. Kadletz, J. Lee, A.S. Tremsin, E. Winter, R. Woracek, M. Zlobinski, L. Gubler, P. Boillat	*Study of Phase Changes in Lithium-Ion Battery Electrolytes via Spectroscopic Neutron Imaging* ECS-2021 240th The Electrochemical Society Meeting, Online, October 10–14, 2021.
Y.-C. Chen, Ch. Csoklich, A. Berger, T.J. Schmidt, F.N. Büchi	*Operando Fuel Cell Liquid Water Distribution in the Microporous Layer* EFCF-2021 European Fuel Cell Forum, Online, June 30, 2021.
J.S. Diercks, J. Herranz, T.J. Schmidt	*A Combined Spectroscopic and Electrochemical Approach to Investigate the Selectivity Determining Factors for CO_2 Electroreduction on Pd-Nanoparticles* EFCF-2021 European Fuel Cell Forum, Online, June 29 – July 2, 2021.
J.S. Diercks, M. Georgi, J. Herranz, N. Diklic, P. Chauhan, A. Eychmüller, T.J. Schmidt	*CO_2 Electroreductionon Unsupported PdPtAerogels: Effects of Alloying and Surface Composition* ECS-2021 240th The Electrochemical Society Meeting, Online, October 10–14, 2021.
N. Diklić, A. Beard, A. Heinriz, T. Cen, J. Herranz, T.J. Schmidt	*Evaluation of commercial catalysts' OER performance by means of RDE* EFCF-2021 European Fuel Cell Forum, Online, June 29 – July 2, 2021.
N. Diklić, A. Beard, A. Heinriz, T. Cen, J. Herranz, S. Garbe, T.J. Schmidt	*Breaking down the activity loss during O_2-evolution stability tests on commercial IrO_2-based catalysts* 4th International Meeting on Materials Science for Energy Related Applications, Digital meeting, September 22–23, 2021.

N. Diklić, A.H. Clark, J. Herranz, J. Diercks, D. Aegerter, T.J. Schmidt	*X-ray absorption spectroscopy studies of Ir-oxide based, oxygen evolution catalysts revisited* ECS-2021 240th The Electrochemical Society Meeting, Online, October 10–14, 2021.
J.C. Duburg, K. Azizi, S. Primdahl, H.A. Hjuler, E. Zanzola, T.J. Schmidt, L. Gubler	*Preparation and performance of a polybenzimidazole based composite membrane for VRFBs* Next Generation – Flow Battery Conference and Networking Event, Online, March 11, 2021.
M. Fikry, M. Georgi, N. Weiss, J. Herranz, A. Eychmuller, T.J. Schmidt	*Impact of ink formulation and processing on the PEFC performance of self-supported Pt-Ni aerogels* EFCF-2021 European Fuel Cell Forum, Online, June 30, 2021.
M. Fikry, P. Khavlyuk, J. Herranz, A. Eychmuller, T.J. Schmidt	*Low temperature PEFC performance of un-supported Pt-Ni aerogel cathode catalyst layers* ECS-2021 240th The Electrochemical Society Meeting, Online, October 10–14, 2021.
S. Garbe, T.J. Schmidt, L. Gubler	*Towards Thin Membrane Use in Polymer Electrolyte Water Electrolysis Using Pt Recombination Catalyst* ECS-2021 240th The Electrochemical Society Meeting, Online, October 10–14, 2021.
L. Gubler, D. Vonlanthen, A. Schneider, F.J. Oldenburg	*Composite Membrane for the VRFB: Bilayer of a Porous Separator and a Polybenzimidazole 'Skin'* International Flow Battery Forum (IFBF), Online, January 19–21, 2021.
L. Gubler, T. de Wild, T. Nemeth, T.J. Schmidt, T. Nauser	*Hydrocarbon Fuel Cell Membranes: Can Radical Induced Damage be Repaired ?* ModVal-2021 17th Symposium on Modeling and Experimental Validation of Electrochemical Energy Technologies, Online, April 20–22, 2021.
Ł. Kondracki, J. Stropp, S. Steiner, S. Trabesinger	*Lithium-substituted Cobalt-free Materials for Na-ion Batteries* Green Batteries Conference 2021, Online, October 26, 2021.

J. Lee, E.R. Carreon-Ruiz, M. Siegwart, P. Boillat	*Segmentation for Preventing Ice Propagation in Operating Fuel Cells* ECS-2021 240th The Electrochemical Society Meeting, Online, October 10–14, 2021.
W. Liu, V.M. Orezzoli, T.J. Schmidt, P. Boillat	*Effects of Hydrophobic Coatings on Non-Isothermal Ice Crystallization in PEMFC GDMS* ECS-2021 240th The Electrochemical Society Meeting, Online, October 10–14, 2021.
A. Mularczyk, Q. Lin, D. Niblett, J. Eller	*Insights into Capillary Pressure in Operating Polymer Electrolyte Fuel Cells* EChemCONSTORE 2021, January 28–29, 2021.
A. Mularczyk, Q. Lin, D. Niblett, J. Eller	*Insights into Capillary Pressure in the GDL of Operating Polymer Electrolyte Fuel Cells* InterPore2021 13th Annual Meeting, Digital conference, May 31 – June 4, 2021.
A. Mularczyk, Q. Lin, D. Niblett, J. Eller	*Operando Determination of the Capillary Pressure in the GDL of Polymer Electrolyte Fuel Cells* EFCF-2021 European Fuel Cell Forum, Online, June 29 – July 2, 2021.
A. Mularczyk, Q. Lin, D. Niblett, A.-P. Vasile, M.J. Blunt, V. Niasar, F. Marone, T.J. Schmidt, F.N. Büchi, J. Eller	*Capillary Pressure Evolution in Operating Polymer Electrolyte Fuel Cells* ECS-2021 240th The Electrochemical Society Meeting, Online, October 10–14, 2021.
H.Q. Pham, Ł. Kondracki, S. Trabesinger	*Exploring the Structural-Gas-Interface Evolution of Co-free Li-rich Layered Oxide Cathode* PSI Swiss Battery Days 2021, Online, February 15–17, 2021.
A. Schuller, T.J. Schmidt, J. Eller	*Noninvasive Determination of local Membrane Conductivity in Polymer Electrolyte Fuel Cells based on a Finite Element Model* ModVal-2021 17th Symposium on Modeling and Experimental Validation of Electrochemical Energy Technologies, Online, April 22, 2021.

A. Schuller, T.J. Schmidt, J. Eller	*Non-invasive Measurement of Humidity Distribution in Polymer Electrolyte Fuel Cells* EFCF-2021 European Fuel Cell Forum, Online, June 30, 2021.
A. Schuller, T.J. Schmidt, J. Eller	*Non-invasive Measurement of Conductivity Distribution in Polymer Electrolyte Fuel Cells (PEFCs)* ECS-2021 240th The Electrochemical Society Meeting, Online, October 10–14, 2021.
M. Srout, S. Trabesinger	*Towards Better Understanding of the Electrochemical Impedance of Lithium Metal Anode* Green Batteries Conference 2021, Online, October 5, 2021.
A. Štefančič, C. A. F. Vaz, M. El Kazzi	*Gas-Phase Surface Fluorination of High-Voltage Layered Transition Metal Oxide ($LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ – NCA): A Convenient Path to Improve the Cycling Stability of Li-Ion Batteries.* PSI Swiss Battery Days, Online, February 15–17, 2021.
M. Striednig, M. Cochet, P. Boillat, T.J. Schmidt, F.N. Büchi	*Evaporative Cooling of Polymer Electrolyte Fuel Cells – A System Level Analysis* ModVal-2021 17th Symposium on Modeling and Experimental Validation of Electrochemical Energy Technologies, Online, April 22, 2021.
M. Striednig, M. Cochet, P. Boillat, T.J. Schmidt, F.N. Büchi	*Evaporative Cooling of Polymer Electrolyte Fuel Cells – A System Level Analysis* ECS-2021 239th The Electrochemical Society Meeting, Online, May 30 – June 3, 2021.
M. Striednig, M. Cochet, P. Boillat, T.J. Schmidt, F.N. Büchi	*A System Level Analysis of Evaporative Cooling for Polymer Electrolyte Fuel Cells* EFCF-2021 European Fuel Cell Forum, Online, June 29 – July 2, 2021.
M. Striednig, A. Michalski, A. Mularczyk, W. Liu, M. Cochet, D. Scheuble, P. Boillat, T.J. Schmidt, F.N. Büchi	*A Combined Numerical and Experimental Study of Evaporative Cooling for Polymer Electrolyte Fuel Cells* ECS-2021 240th The Electrochemical Society Meeting, Online, October 10–14, 2021.

D. Tim, S. Mayank, E. Jens	*Insights into the Stability and Formation of Water Droplets Using Operando X-Ray Tomographic Microscopy* ECS-2021 240th The Electrochemical Society Meeting, Online, October 10–14, 2021.
S. Ünsal, T.J. Schmidt, J. Herranz	*The Effect of the Precursor Agglomerate Size on the Morphology and Performance of a Non-Noble Metal Oxygen Reduction Catalyst* EFCF-2021 European Fuel Cell Forum, Online, June 29 – July 2, 2021.
S. Ünsal, S. Gialamoidou, T.J. Schmidt, J. Herranz	*Agglomerate Size Effect on the PEMFC Performance of a Non-Noble Metal Oxygen Reduction Catalyst* ECS-2021 240th The Electrochemical Society Meeting, Online, November 11, 2021.
E. Zanzola, J.C. Duburg, K. Azizi, S. Primdahl, H.A. Hjuler, T.J. Schmidt, L. Gubler	*Polybenzimidazole membrane for vanadium redox flow batteries with high capacity retention* EFCF-2021 European Fuel Cell Forum, Online, June 29 – July 2, 2021.

Posters

J.B. Falqueto, A.H. Clark, N. Casati, C.A.F. Vaz, A. Štefančič, N. Bocchi, M. El Kazzi	*Understanding the Role of Al, Co or Ni Doping in $LiMn_2O_4$ Nanoparticles for Improving the Structural Stability in Li-Ion Battery* Swiss Battery Days PSI, Virtual conference, February 15–17, 2021.
S.D. Lacey, E. Gilardi, E. Müller, C. Merckling, G. Saint-Girons, C. Botella, R. Bachelet, D. Pergolesi, M. El Kazzi	*Integration of $Li_4Ti_5O_{12}$ Thin Films on Silicon towards High-Rate Performance Microbatteries* Swiss Battery Days PSI, Virtual conference February 15–17, 2021.
T. de Wild, T. Nemeth, T. Nauser, T.J. Schmidt, L. Gubler	*Proposing a Repair Mechanism for Hydrocarbon-based Polymer Electrolyte Membrane to Enhance Chemical Durability* EMEA-2021 Workshop on Ion Exchange Membranes for Energy Applications, virtual conference, June 22–24, 2021.

T. de Wild, T. Nemeth, T. Nauser, T.J. Schmidt, L. Gubler	*Possible Repair Mechanism for Aromatic Hydrocarbon-based Polymer Electrolyte Membranes used in Fuel Cells* EFCF-2021 European Fuel Cell Forum, online conference, June 29 – July 2, 2021.
J.C. Duburg, K. Azizi, S. Primdahl, H.A. Hjuler, E. Zanzola, T.J. Schmidt, L. Gubler	*Composite Polybenzimidazole Membrane with High Capacity Retention for Vanadium Redox Flow Batteries* EMEA-2021 Workshop on Ion Exchange Membranes for Energy Applications, virtual conference, June 22–24, 2021.
L. Höltschi, M. Mirolo, C. Jordy, V. Pelé, C. Borca, T. Huthwelker, C.A.F. Vaz, P. Novák, M. El Kazzi	*Revealing Chemical and Electrochemical Reactions via Operando X-Ray Spectroscopy: an Interface Study Between NCM111 and a Sulfide Electrolyte* Swiss Battery Days PSI, Virtual conference, February 15–17, 2021.

Conference Organizations

F.N. Büchi, C. Gloor, L. Gubler, S. Trabesinger	*37th Swiss Electrochemistry Symposium: Prospects of Future Transport Electrification* Virtual, Paul Scherrer Institut, Villigen, Switzerland, April 21, 2021.
F.N. Büchi, (Co-Organizer)	*Symposium Polymer Electrolyte Fuel Cells and Electrolyzers 21 (PEFC&E 21)* Virtual, 240th ECS Fall Meeting, October 10–14, 2021.
T.J. Schmidt, E. Fabbri (Chair)	*2021 European Fuel Cell Forum (EFCF 2021)* Virtual, July 5–8, 2021.
S. Trabesinger (Organizer)	*Swiss Battery Days 2020-2021* Virtual, Paul Scherrer Institut, Villigen, Switzerland, February 15–17, 2021.

Editorial Work

F.N. Buechi, A. Kusoglu, B.S. Pivovar, C.A. Rice, H. Uchida, K.E. Ayers, D.R. Dekel, J.M. Fenton, H. Jia, D.J. Jones, Y.T. Kim, E. Kjeang, U.I. Kramm, B. Lakshmanan, R.A. Mantz, S. Mitsushima, W. Mustain, P.N. Pintauro, P. Strasser, K. Swider-Lyons, A.Z. Weber, H. Xu, I.V. Zenyuk (Editors)	*Polymer Electrolyte Fuel Cells and Electrolyzers 21 (PEFC&E 21),* *ECS Transaction* **104** *(8)* The Electrochemical Society, Pennington, NJ (2021).
T.J. Schmidt (Associate Editor)	Journal of the Electrochemical Society.
T.J. Schmidt (Co-Author)	*Technology Outlook 2021* *(https://www.satw.ch/de/technology-outlook-2021),* Schweizer Akademie der Technischen Wissenschaften.

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