PAUL SCHERRER INSTITUT

Annual Report 2020

Electrochemistry Laboratory

Cover

Neutron Imaging at ICON beamline. Detection of partial solidification in commercial Li-based electrolytes and organic solvents to study degrading effects.

PAUL SCHERRER INSTITUT

Annual Report 2020

Electrochemistry Laboratory

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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, 2020 has also been a special year for the Electrochemistry Laboratory. Still we were able to have a strong scientific output. In this Annual Report 2020, 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 2020 are given, including a full list of our 65 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 36th Swiss Electrochemistry Symposium «Seasonal Energy Storage: The Role of Electrochemistry» was hold successfully in September as online event with world-class scientists and engineers as speakers, attracting an audience of more than 120 participants. Details of this event you will find also in this annual report on page 53.

2020 was also a successful year for four 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

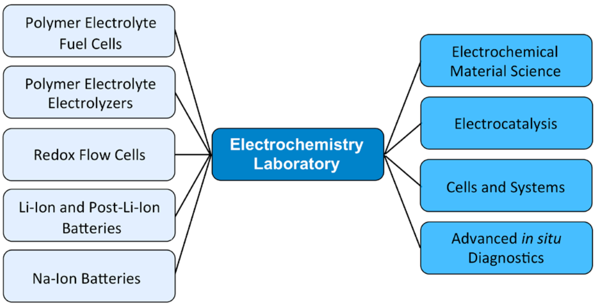
Our Mission
Advancing electrochemical energy storage and conversion by
- **developing novel electrochemical materials, cells and devices;**
- **using novel, advanced in situ and operando methods;**
- **performing fundamental, applied and industry-oriented research.**

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 of electrochemical energy storage and conversion specifically in the context of a sustainable energy system, where renewable energy is required to be stored in secondary batteries or chemicals such as hydrogen and (re-)converted into electricity. The Laboratory's R & D, is hence focused on secondary batteries – specifically Li- and Na-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, e.g., fuel cell systems.

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.

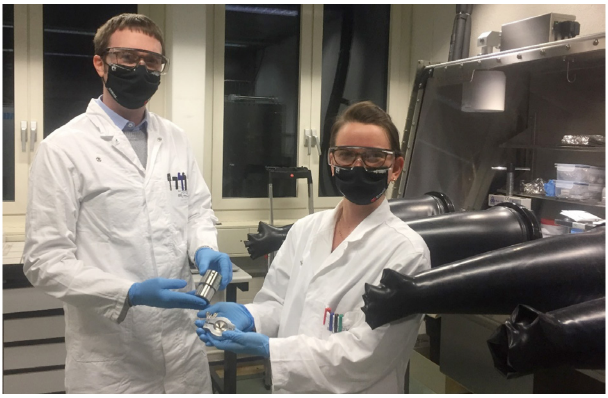


Electrochemical Energy Storage

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.



Juliana Bruneli Falqueto (right), PhD student from Federal University of Sao Carlos, Brazil, and Aleš Štefančič (left), postdoc at Paul Scherrer Institut with the operando cell developed internally at PSI and used at the SuperXAS beamline at Swiss Light Source (SLS).

Electrochemical Energy Conversion

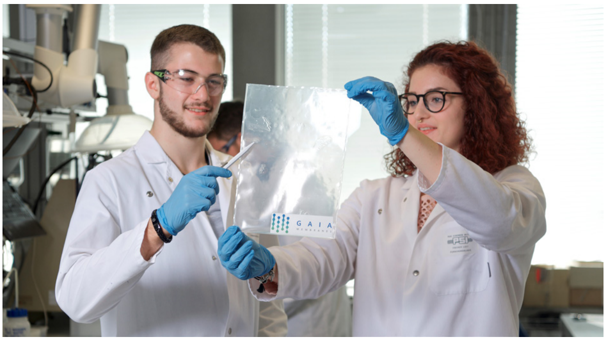
The Electrochemical Energy Conversion focuses on the development and in-depth understanding of materials, processes and devices for the conversion of renewable power to hydrogen (or syngas through the electrochemical reduction of carbon dioxide) and back to power. Especially in the context of a sustainable 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 elecgtrocatalysts and improved understanding of intrinsically limiting factors;
- development of new porous materials for PEFC and PEWE;
- development of new membrane architectures for fuel cells, electrolyzers and redox flow cells based on PSI's own radiation-grafting technology;
- 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.



Team members of the PSI Spin-Off Gaia Membranes with amphoteric ion exchange membrane for vanadium redox flow batteries.

CURRENT SCIENTIFIC TOPICS

SCIENTIFIC HIGHLIGHTS

Scientific
Highlight

1

Hierarchically Structured Porous Transport Layers for Polymer Electrolyte Water Electrolysis

The high operational and capital costs of polymer electrolyte water electrolysis technology originate from limited catalyst utilization and the use of thick membrane electrolytes. PSI researchers have developed novel multi-layer porous transport materials, which provide superior electrochemical performance in comparison to conventional single-layer structures.

Conventionally, in polymer electrolyte water electrolyzers, thick membrane electrolytes are used which leads to high operational cost because of high power consumption and the catalyst utilization is limited leading to increased capital cost because of the required high noble metal catalyst loadings. This is due to the coarse surface structure of the state-of-the-art titanium porous transport layer materials used. Therefore, a series of materials with three different microporous layers (MPLs) with advanced interface properties are fabricated and characterized.

It is shown that these sintered multilayer structures, made from economically viable titanium powders, have improved interface properties with low surface roughness, as characterized by X-ray laboratory and synchrotron-based tomographic microscopy. The transport layer materials provide superior electrochemical performance in comparison to conventional single-layer structures, with up to three times higher catalyst layer utilization and a ≈ 60 mV decrease in (anodic) mass transport overpotential at 2 A cm^{-2}. The MPLs combine preferential surface properties with high open porosity and low tortuosity of sinter materials, enabling for the first time the use of thin membranes, in combination with anodic titanium transport layers. The fundamental mechanism of the MPL effect is elucidated and shown to be based on a homogeneous contact pressure distribution, resulting in high catalyst utilization and low mass transport losses.

Figure 1
Left: 3D-rendered two-layer porous transport layer with smooth layer on top facing the catalyst layer;
right: mechanistic illustration of the double layer structure leading to increased catalyst layer utilization and reduced membrane deformation.

Acknowledgements

Swiss Federal Office of Energy (SFOE, Grant No. SI/501331-01) for funding of this project and Umicore for providing catalyst material for this study.

Publications

Hierarchically Structured Porous Transport Layers for Polymer Electrolyte Water Electrolysis.
Tobias Schuler, Joseph M. Ciccone,
Bernd Krentscher, Federica Marone,
Christian Peter, Thomas J. Schmidt, and
Felix N. Büchi.
Advanced Energy Materials 10(2), 2020.
DOI: 10.1002/aenm.201903216

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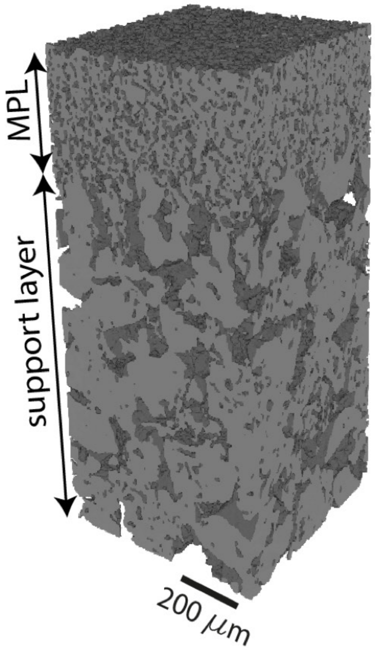

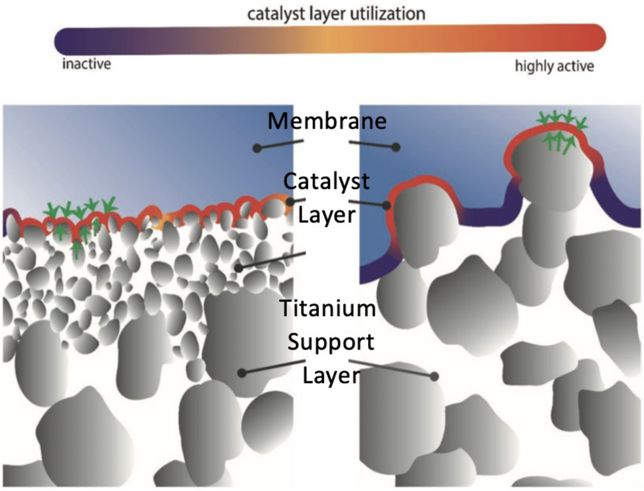

Membrane Lifetime Estimation in a Vanadium Redox Flow Battery using an Accelerated Stress Test

A vanadium redox flow battery (VRFB) is a grid-scale energy storage device. Its energy conversion unit consists of a cell stack that comprises ion-exchange membranes to separate positive and negative electrodes. The projected lifetime of a VRFB is 20 years and 7'000 charge-discharge cycles. Lifetime tests of membranes under application relevant conditions are therefore impractical, and the development of an accelerated stress test (AST) to assess the chemical stability of membranes is crucial.

In the VRFB, vanadium-ions of different oxidation state are used to build a rechargeable electrochemical energy storage device (battery). Vanadium-ions of valence +II and +III are dissolved in an aqueous sulfuric acid in the negative electrolyte, and vanadium-ions of valence +IV and +V in the positive electrolyte. The vanadium +V, V(V), can cause oxidative membrane degradation. In our AST, membranes of various composition are immersed in a solution containing V(V) in 2 M sulfuric acid. The oxidative membrane degradation was accelerated by increasing the temperature of the solution and its oxidative strength. Therefore, we replaced V(V) by cerium(+IV), Ce(IV), which is a stronger oxidant than V(V). The degradation caused by Ce(IV) follows a similar reaction pathway but is faster by a factor of ~4 compared to V(V). The acceleration factor of the degradation reaction in Ce(IV) solution at 80 °C is ~200 compared to the test in the VRFB single cell.

The validity of this AST method was assessed by comparing tests in the cell with accelerated tests *ex situ* in the AST solution. Based on an *in situ* charge-discharge cell test over 1'500 cycles and post-test analysis of the state of health of the membrane, the lifetime of the membrane was projected to be 4'600 ± 900 h. The lifetime of the membrane estimated based on the *ex situ* AST was 3'300 h, which is within the limits of the projection. However, the uncertainty of the estimation is rather high (2'000 h), which is a result of limited number of statistically sound AST data points. Statistics of estimates are improved with a larger body of AST data.

For the application of the AST method to other membrane materials and polymer classes for vanadium and other flow battery types, the assumption of a consistent degradation mechanism and transferable composition-property relations to the conditions in the cell needs to be established and validated. Thus, the developed AST method can be a valuable tool for membrane and flow battery developers, together with other membrane characterization techniques, to screen and down-select candidate materials for development. It provides an «engineering test» or «index test» and can serve as a basis for predictive modeling of the membrane lifetime in VRFBs. With an acceleration factor of 200, tests over 500 h can be used to mimic membrane aging over 10 years under flow cell application conditions.

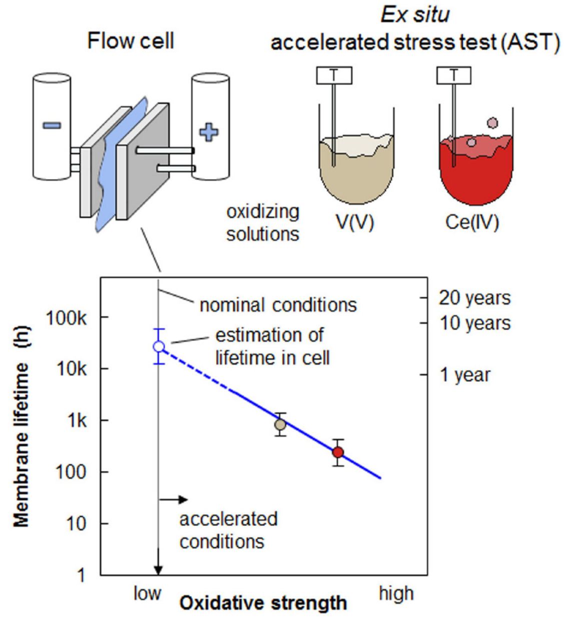

Figure 2
The membrane in a vanadium redox flow battery suffers from oxidative aging. It needs to be stable for > 5 years in the battery. Chemical stability of membrane materials can be assessed in much shorter time (10s to 100s of hours) by immersing them in test solutions with higher oxidative strength. Knowing the functional relationship between oxidative strength of the environment and the rate of chemical aging allows to estimate membrane lifetime under nominal flow cell operating conditions.

Publications

Accelerated Stress Test Method for the Assessment of Membrane Lifetime in Vanadium Redox Flow Batteries.
Fabio J. Oldenburg, Ayoub Ouarga,
Thomas J. Schmidt, Lorenz Gubler.
ACS Applied Materials & Interfaces, 11(51), 2019.
DOI: 10.1021/acsami.9b15736

Acknowledgements

The Swiss Federal Office of Energy (grant no. SI/501421-01) is kindly acknowledged for funding of the project. T.J. Schmidt thanks Innosuisse and the Swiss Competence Center for Energy Research (SCCER) Heat and Electricity Storage. A. Ouarga thanks the Materials Science and Nanoengineering Department (MSN) of Mohammed VI Polytechnic University (UM6P) for financially supporting the research stay at PSI.

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Scientific Highlight

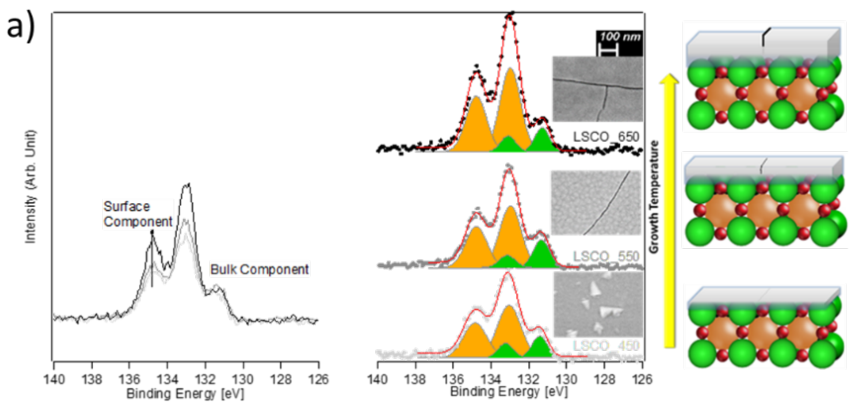
3

Surface Segregation Acts as Surface Engineering for the Oxygen Evolution Reaction on Perovskite Oxides in Alkaline Media

Electrolyzers show great promise for compensating the variable energy production from renewable resources by producing the versatile energy vector hydrogen; however, the development of efficient, robust and economically viable electrolyzers is still facing important scientific and technological challenges. Oxygen electrocatalysis plays a pivotal role in the development of electrolyzers since the anodic reaction, the oxygen evolution reaction (OER), represents one of the main sources of overpotential. Perovskite oxides have emerged as promising anodic electrodes for alkaline water electrolyzers, and an increasing number of studies have been recently published about perovskite OER catalysts. Particularly, many studies were dedicated to finding «activity descriptors» which means the key perovskite properties governing the OER activity.

While the first studies on perovskite catalysts have been mostly focused on the correlation of bulk perovskite properties with the OER activity, recent research efforts have been more and more directed towards the understanding of the electrode/electrolyte interface. The growing attention towards interface properties has fostered the development of surface sensitive techniques, particularly *in situ* and under *operando* conditions, such as near ambient pressure X-ray photoelectron spectroscopy (NAP-XPS).

PSI researchers have studied the influence of surface segregation on the OER activity for the, $La_{0.2}Sr_{0.8}CoO_{3-d}$ (LSCO) perovskite, one of the most active perovskites towards the OER in alkaline electrolyte. LSCO has been produced in the form of thin films by means of pulsed la-



ser deposition (PLD) using different deposition temperatures (between 350 and 650 °C). The morphology of the films has been characterized by means of SEM and atomic force microscopy (AFM) while the crystalline structures have been checked with X-ray diffraction (XRD). NAP-XPS has been used to check the surface composition of perovskite films synthesized at different temperatures, showing that high temperature favors strontium segregation in the topmost layers. However, strontium forms surface compounds that are efficiently removed when the catalyst surface is in contact with water. Indeed, NAP-XPS measurements revealed that after immersion in water LSCO shows a cobalt rich surface, which is very active for the OER.

More generally, higher deposition temperature in the 450–550 °C range translates into higher OER activity for the LSCO catalysts. Indeed, the higher the perovskite synthesis temperature the more strontium segregation occurs on the surface. However, being the segregated strontium compounds soluble in water, they are easily removed when the surface of the electrode is in contact with the electrolyte, leading to the exposure of cobalt enriched layers which are very active for OER.

Publications

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

Acknowledgements

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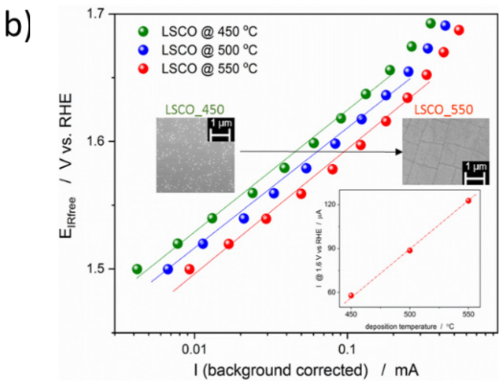

Figure 3
a) Sr 3d spectrum showing the increased surface strontium peak with deposition temperature for samples prepared at 450 °C, 550 °C and 650 °C. Sr 3d peak deconvolution: surface strontium (orange), bulk strontium (green), and the envelope fitting (red).
Next to the XPS spectra the SEM images are shown.
A schematic indication of the increased segregated strontium islands layer with growth temperature is shown on the right.
b) Tafel plot in 0.1 M KOH of samples prepared at 450 °C (green), 500 °C (blue) and 550 °C (red). Lower right corner: the evolution of the current measured at 1.6 VRHE for the three preparation temperatures.

Operando X-Ray Photoelectron Spectroscopy to Monitor the Chemical and Electronic Interface Evolution in All-Solid-State Batteries

Degradation of the solid-electrolyte interface occurring during cycling is currently one of the most challenging issues for the development of all-solid-state batteries. Here we designed a unique electrochemical custom made cell for *operando* X-ray photoelectron spectroscopy (XPS) capable of maintaining high mechanical pressure with reliable electrochemistry and able to monitor in real-time the surface (electro-)chemical reactivity at the interfaces between the different composites.

The (electro-)chemical reactivity and electronic properties across electrified solid-solid interfaces critically determine the functionality of solid-state electrochemical devices. To date, questions still arise in the scientific community due to the limitations of surface-sensitive characterization techniques, especially in post-mortem mode. Particularly, parameters such as the electrochemical stability of the solid electrolyte (SE) as well as the ionic and electronic percolation within the composite electrode are essential in order to push all-solid-state lithium-ion batteries (SSBs) towards practical applications.

In this work, we highlight *operando* X-ray photoelectron spectroscopy (XPS) as a straightforward method to monitor the SE reduction within a composite electrode consisting of $Li_4Ti_5O_{12}$ (LTO) as the electrode active material, $(Li_2S)_3$-P_2S_5 (LPS) as the SE and vapor-grown carbon fiber as the conductive additive under real working conditions. The results clarify previous discrepancies between experimental and theoretical results regarding the anodic stability limit of LPS and also reveal the limitations of post-mortem XPS measurements that can potentially lead to misinterpretations. We identify the reduction process of LPS which starts at 1.7 V vs. Li^+/Li (1.1 V vs. $InLi_x$), resolved as a two-step process accompanied by the formation of an electronically insulator Li_2S and electronically conductive Li_xP species. Furthermore, we propose in our study a fundamental physical model to underline the effect of the external cell voltage on the core levels binding energy

Figure 4

Operando XPS enables monitoring the chemical and electronic properties of the interface solid electrolyte/active material in all-solid state battery. When cycling $Li_4Ti_5O_{12}$ (LTO) with Li_3PS_4 (LPS) a strong irreversibility is observed in the first cycles. Thanks to the operando XPS on the S2p and P2p core levels this behavior is identified to be related to the formation of an electronically insulator Li_2S and conductive Li_xP species as byproducts from the LPS reduction at the interface with LTO.

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shifts, which provides a direct contactless measurement of the local surface potentials of the different particles. Thanks to that, the *operando* methodology resolves the LTO redox activity and the distribution of conductive and non-conductive LTO species as a function of lithiation. This allows a deeper understanding of both the lithium-ion transport through the composite electrode and the characteristic metal-insulator transition associated with LTO.

Finally, we discuss how the interfacial reactivity between LPS and LTO impacts the electrochemical performance of SSBs. Our novel *operando* XPS development paves the way for an accurate interpretation of the core levels peaks and to the assignment of the SE byproducts leading to a better description of the interfacial reaction mechanism in SLiB.

Publications

Insights into the chemical and electronic interface evolution of $Li_4Ti_5O_{12}$ cycled in $Li_2S–P_2S_5$ enabled by operando X-ray photoelectron spectroscopy.
Xiaohan Wu, Claire Villevieille, Petr Novák, Mario El Kazzi.
J. Mater. Chem. A, 8, 2020.
DOI: 10.1039/C9TA14147B

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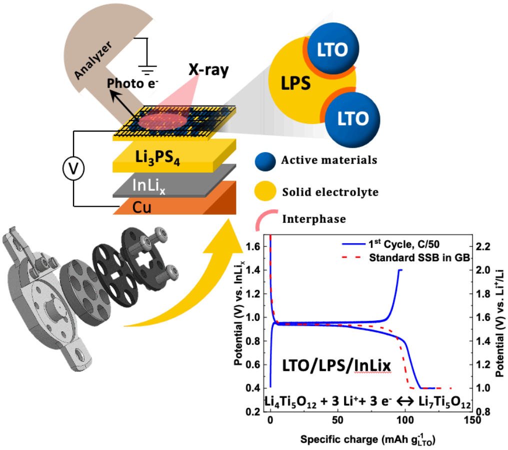
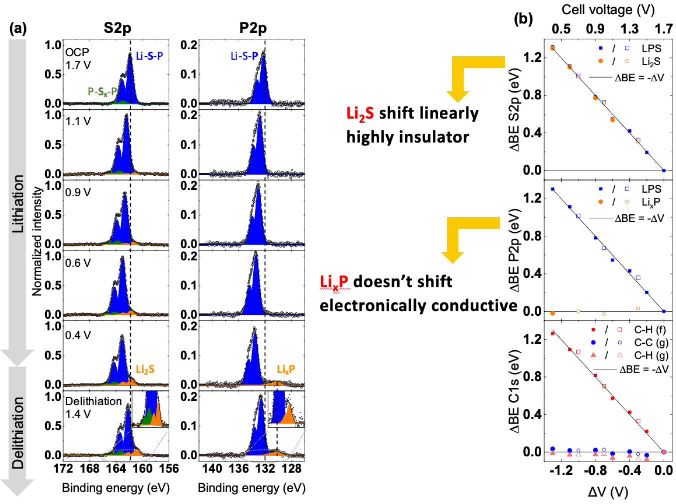
***Operando* XPS cell**



Li_3PS_4/Li_4TiO_{512} interface chemical and electronic properties

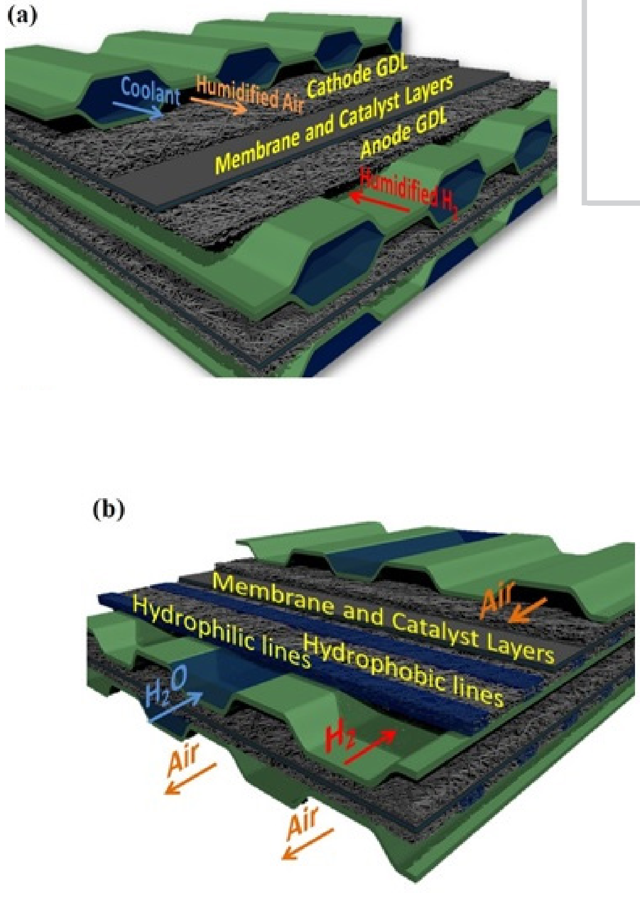


How to Increase the Power Density of Polymer Electrolyte Fuel Cells with Evaporative Cooling

A system of evaporative cooling for Polymer Electrolyte Fuel Cells (PEFCs) has been developed at PSI, based entirely on one simple, yet effective change of one of the fuel cell components. Our team at the Electrochemistry Laboratory has demonstrated how this single change allows to operate a cell without the need of bulky and costly external humidifiers. Additionally, the proposed design has the potential to increase the power density of a PEFC stack by up to 35% due to the sparing of the space usually dedicated to the convective coolant circulation.

Polymer Electrolyte Fuel Cells (PEFCs) are considered an alternative to the carbon-based combustion engines, as they generate electricity out of hydrogen and oxygen, and release nothing but water. In a fuel cell, the reactant gases are brought in through gas channels on two opposite sides, one for the hydrogen (anode) and one for the air (cathode). Chemical reactions take place on the so-called catalyst layers (CL) to break up the reactant molecules on one hand and create water and electricity on the other. The two catalyst layers are very thin layers separated by a polymer membrane, which blocks electrons and only allows the passage of protons. The electrons are forced through an external circuit, which allows electrical energy to be harvested. Membranes need to be well humidified to conduct protons and this is usually achieved by humidifying the reactant gases upstream from the cell using costly external humidifiers.

To reach the CL, the reactant gases in the channels have to diffuse through very thin porous media made of intertwined carbon fibers, and called gas diffusion layer (GDL). The GDL also helps transport the water produced by the chemical reaction from the CL to the gas channels, where it can be removed by the gas



stream. To avoid accumulating water droplets in the GDL's small pores, which would block the pathways of the gases to the CL, the GDL is coated with a highly hydrophobic solution (very often, PTFE).

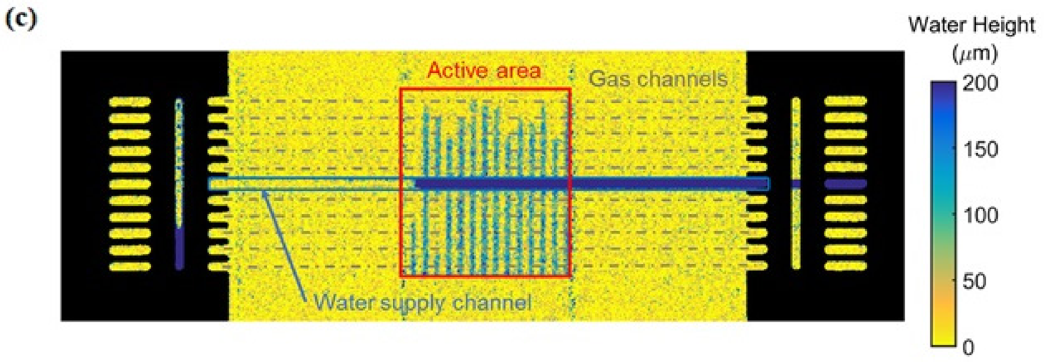
Last, the electrochemical reactions are on balance exothermic, and the heat produced must be removed to avoid overheating the cell. Usually this is achieved by flowing a coolant in an internal volume placed between two adjacent cells, at the expense of the cell's compactness.

PEFC still face many challenges to reduce their size and cost before they can reach commercialization on a large scale. One of these challenges is to humidify the reactant gases without expensive external humidifiers. Another one is to remove the waste heat of the reactions without dedicating a consequent part of the PEFC volume to cooling only.

At PSI, a process has been developed to add a hydrophilic polymer on selected surfaces of the hydrophobic coating and transform the GDL into a succession of hydrophobic and hydrophilic zones. These zones of opposite wettability allow the separate transport of gases and water. We can use these advanced GDLs to build a fuel cell with evaporative cooling, where humidification and cooling are managed simultaneously, in a way that entirely removes the need for internal cooling circuits and for external humidifiers.

The modified GDL is placed on the anode side, with the hydrophilic/hydrophobic lines perpendicular to the gas channels. A small number of

Figure 5
a) PEFC with convective cooling. Humidified gases diffuse from the gas channels to the catalyst layers through the GDL. The coolant flows in a dedicated circuit between two adjacent cells.
b) PSI evaporative cooling concept. Water, supplied by some anode channels, is wicked into the hydrophilic lines of the GDL, spreads above the gas channels, and evaporates in contact with the gas flow.
c) Neutron image of test laboratory scale PEFC with evaporative cooling. The hydrophilic lines are perpendicular to the gas channels.



these channels are transformed into liquid water supply channels. The water brought into these channels is wicked directly by the hydrophilic lines, and spreads above several gas channels. Once in contact with the hydrogen flow, the water in the hydrophilic line evaporates, which provides the humidification and the cooling effect needed. Thanks to the constant supply of water, the hydrophilic lines are always filled, and high evaporation rates can be sustained as long as needed for different operating conditions. Meanwhile, the (very) hydrophobic lines remain dry pathways for the reactant gases and ensure a good operation of the cell.We have demonstrated, using a 4 cm^2 laboratory cell equipped with thermal management instrumentation, that our concept can indeed provide the levels of humidification and cooling power needed for the good operation of a fuel cell at 80 °C, and potentially at higher temperatures. The concept is robust, provided the pressure of the water is well controlled, and the performance of the cell with evaporative cooling is similar to that of the cell without evaporative cooling, albeit with a small loss of efficiency at very high current densities. Fuel cell stacks equipped with this concept could be up to 33% more compact, which would mean, after accounting for the small loss in efficiency, that an increase of the global power density of up to 35% of the cell would be possible. In addition the removal of the external humidifiers, and the simplification of the entire system would make a fuel cell equipped with this evaporative cooling concept an attractive option for an automotive powertrain.

Publications

Enabling High Power Density Fuel Cells by Evaporative Cooling with Advanced Porous Media.
M. Cochet, A. Forner-Cuenca, V. Manzi-Orezzoli, M. Siegwart, D. Scheuble, P. Boillat.
J. Electrochem. Soc. 167, 084518 (2020).
DOI: 10.1149/1945-7111/ab8e82

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Graphite Anodes with Si as Capacity-Enhancing Electrode Additive

Silicon is a long-standing candidate for replacing graphite as the active material in negative electrodes for Li-ion batteries, due to its significantly higher specific capacity. However, Si suffers from rapid capacity loss, as a result of the large volume expansion and contraction during lithation and de-lithiation. As an alternative to pure Si electrodes, Si could be used as a capacity-enhancing additive to graphite electrodes.

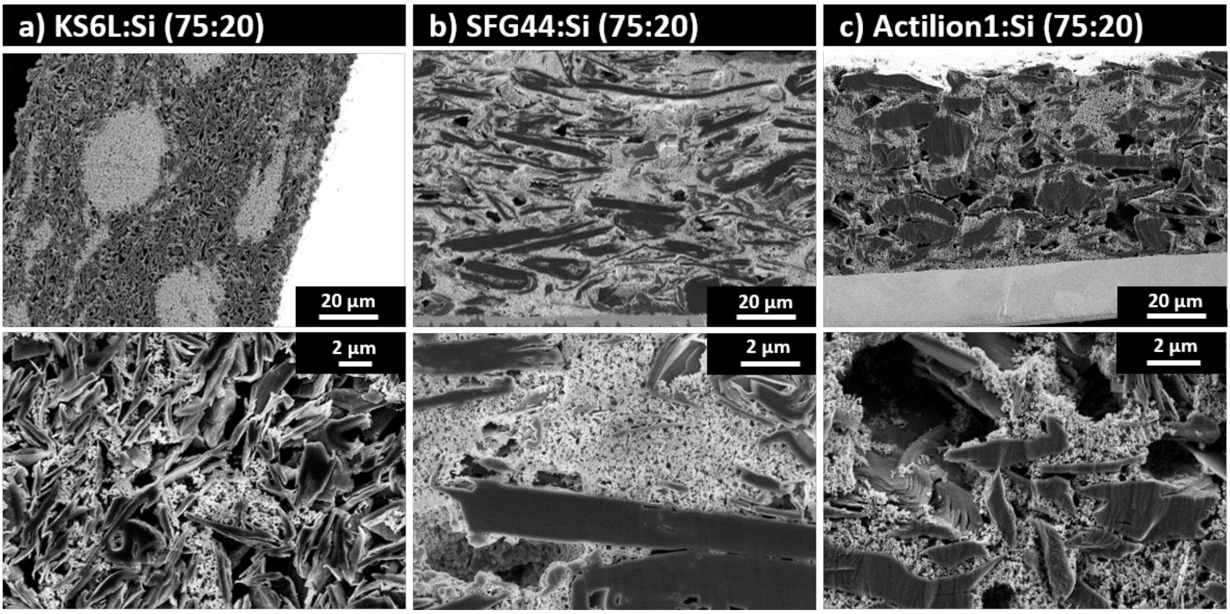
Such graphite–Si blend-electrodes have lower irreversible-charge losses during the formation of the solid electrolyte interphase (SEI) and maintain a better electronic contact within electrodes and with current collectors.

In this context we studied how the choice of graphite matrix can alter the cell performance. Varying the type of graphite and the Si content (5 or 20 wt%) results in different electrode morphologies and performance upon long-term cycling. In electrode blends comprising graphite and Si, graphite takes on the role as both active material and conductive additive. We have looked into several graphite properties, such as particle-size, particle-size distribution, conductivity, crystallinity and surface area. Our results demonstrate that, despite unfavorable electrode morphologies, such as large void spaces and poor active-material distribution, certain types of graphites with large particle sizes were found to be competitive with graphite–Si blends, containing smaller graphite particles. A broader particle-size-distribution within a specific graphite helps to achieve equally good capacity retention with an overall larger graphite particle-size, due to the ability of the fine fraction of the graphite to penetrate into the agglomerated Si domains, filling part of the void space created by the larger particles. In the presence of only 5 wt% Si, large Si domains are found between the large graphite particles, which are potential spots of increased stress and strain during the expansion of Si upon lithiation. A more homogeneous material intermixing was achieved only when graphites with smaller particle sizes were added to the electrodes. When the Si content is raised to 20 wt% and the graphite dimensions are in the range of tens of µm, the electrodes show an «inverse morphology», where the Si-fraction forms the electrode matrix with embedded graphite particles (graphite in silicon), see image. This is in stark contrast to electrodes containing smaller graphite particles, such as KS6L, where the main matrix is still provided by the graphite fraction (silicon in graphite), but which does not prevent Si agglomeration.

Rate capability is always coming into the question when new materials are characterized; however, this question also arises when blends of different electroactive materials are used. As delithiated Si is considered a poor electronic conductor, one would assume that the presence of Si would induce additional internal resistance that hampers fast charging of this type of electrodes. We demonstrated, using current-interruption measurements that the internal resistance of graphite–Si blends indeed increases proportionally with the amount of Si in the electrode. And indeed, the additional resistance induced by the Si fraction in the active material blend impedes fast charging. Two strategies have been tested to mitigate the poor charging behavior at high rates:

1) the densification of the electrode coating and
2) higher contents of conductive additive.

However, our results have shown that additional cell resistances, resulting in poor charging capability, do not solely originate from the presence of Si, because even the Si-free, solely graphite electrodes are also lagging behind at the technologically-feasible rates, despite the small potential gaps measured during the current-interruption experiments. It is therefore likely that besides limitations of the active materials and the electrode architecture, intrinsic limitations of our cell setup are approached at high rates. Nonetheless, the experiments allowed a comprehensive comparison of fast charging limitations in graphite–Si electrodes and an evaluation of the two common approaches to mitigate poor charge capability



Publications

Graphite Particle-Size Induced Morphological and Performance Changes of Graphite–Silicon Electrodes.
Fabian Jeschull, Yuri Surace, Simone Zürcher, Giacomo Lari, Michael E. Spahr, Petr Novák, Sigita Trabesinger.
J. Electrochem. Soc. 167, 100535 (2020).
DOI: 10.1149/1945-7111/ab9b9a

Fast-Charge Limitations for Graphite Anodes with Si as Capacity-Enhancing Additive.
Fabian Jeschull, Sigita Trabesinger.
Batteries & Supercaps, 4(1), 131–139 (2021).
DOI: 10.1002/batt.202000177

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Figure 6
SEM cross-section images of graphite–Si electrodes (wgraphite/wSi = 75:20), containing graphites KS6L (a), SFG44 (b) and Actilion1 (c). The electrodes with smaller graphite (a) are prone to Si segregation at higher Si contents, while graphites with larger particle size becomes embedded into the Si matrix (b, c).

Scientific Highlight

7

Bilayer Composite Membrane for the Vanadium Redox Flow Battery

The vanadium redox flow battery (VRFB) is designed for grid-scale energy storage applications. Ion-exchange membranes are performance and cost relevant components of redox flow batteries. Currently used materials are largely «borrowed» from other applications that have different functional requirements. For next generation VRFBs, it would be desirable to develop membrane materials based on low-cost porous separators with low resistance and high transport selectivity to minimize vanadium-ion and electrolyte crossover.

We present a composite membrane for the VRFB consisting of a porous propylene (PP) separator of 30 µm thickness laminated with a thin meta-polybenzimidazole (mPBI) layer. PBI films are prepared by solution casting to obtain thicknesses in the range of 0.2 to 10 µm. Upon immersion of PBI in the electrolyte solution containing 1.6 M vanadium in 2 M sulfuric acid, the benzimidazole units of PBI are protonated, hence creating an ion conducting polymer. Ohmic resistance of PBI equilibrated in the electrolyte was measured at ~30 mΩ·cm^2 per micron of film thickness. This offers the prospect of designing a composite membrane-separator with low ohmic resistance.

PP-PBI composite membranes with PBI layer thickness ranging from 0.2 to 10 µm were tested in single laboratory cells and compared to a cell with Nafion 212 membrane. For all PBI thicknesses, an improved coulombic efficiency was measured. The voltage efficiency was similar to that with Nafion 212 for the membrane with the thinnest PBI layer (0.2 µm), thicker PBI layers induced increasing ohmic losses. The overall charge-discharge efficiency was superior to that with Nafion 212 for the membranes with 0.2 and 1 µm PBI thickness up to a current density of 0.2 A/cm^2. These results demonstrate that with a careful choice of constituents and architecture, it is possible to prepare membranes with high performance, high efficiency and low cost for next generation redox flow battery systems.

Figure 7

Charge/discharge cycling efficiency at room temperature of a single cell with 25 cm^2 active area as a function of current density with different composite membranes based on a dense PBI layer on a porous PP substrate, compared to Nafion® 212 (thickness: 50 µm).

Electrodes: Toyobo carbon felt, type AAF304ZS (compression: 28%); electrolyte: 1.6 M V in 2 M H_2SO_4 and 0.5 M H_3PO_4.

Publications

Composite Membranes Containing a Porous Separator and a Polybenzimidazole Thin Film for Vanadium Redox Flow Batteries.
Lorenz Gubler, David Vonlanthen, Aaron Schneider, Fabio J. Oldenburg.
Journal of The Electrochemical Society, 167, 2020.
DOI: 10.1149/1945-7111/ab945f

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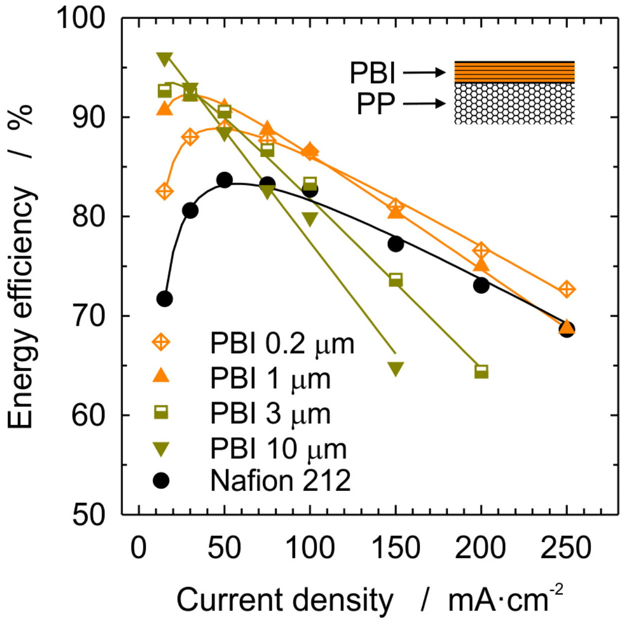

Understanding of the Oxygen Evolution Reaction Kinetics in Acidic Environment

The high operational expenditure of polymer electrolyte water electrolysis (PEWE) technology, dominated by kinetic losses from the sluggish oxygen evolution reaction (OER), inhibits large-scale market penetration. PSI researchers have developed a novel methodology to probe underlying reaction mechanism of the OER. For the first time the reaction order for water has been determined. Advanced benchmarking of catalysts in technical environment also supports the development of novel, highly efficient catalyst materials.

For rational design of highly active OER catalyst materials, the reaction mechanism (RM) and related reaction intermediates are of main interest. Over the last decades, several RM have been proposed but experimental evaluation has still been missing. In particular, the reaction order with respect to water is essential for the identification of valid RM. Furthermore, benchmarking of OER catalyst with conventionally used characterization techniques based on liquid electrolytes is shown to be sensitive towards gas passivation effects. Accumulation of gas bubbles disturb the kinetic measurements and lead to undesired mass transport effects.

We have determined the RM based on the development of a novel characterization technique providing the essential degree of freedom: variation of the water activity. This vapor feed based methodology provided not only for the first time access to characterization of all kinetic parameters, the isolation of valid RM and rate determining steps but also ensures benchmarking in absence of gas passivation effects. Subsequently, the deduction of a generic relation for gas as well as liquid OER kinetics in polymer electrolyte water electrolysis was obtained. The cited paper gives the reaction order with respect to water (2.07 ± 0.10) and other electro-kinetic parameters for the state-of-the-art OER catalyst IrO_2/TiO_2 (iridium oxide supported on titanium oxide).

Figure 8
Measurement of the reaction order of water for the oxygen evolution reaction in acidic environment for TiO_2 supported IrO_2 catalyst using vapor fed polymer electrolyte electrolysis cell set-up, images adapted from Schuler et al. (DOI: 10.1039/D0EE00673D).

Publications

Towards a Generic Understanding of Oxygen Evolution Reaction Kinetics in Polymer Electrolyte Water Electrolysis.
Tobias Schuler, Taro Kimura, Thomas J. Schmidt, Felix N. Büchi.
Energy Environ. Sci. 13, 2153 (2020).
DOI: 10.1039/d0ee00673d

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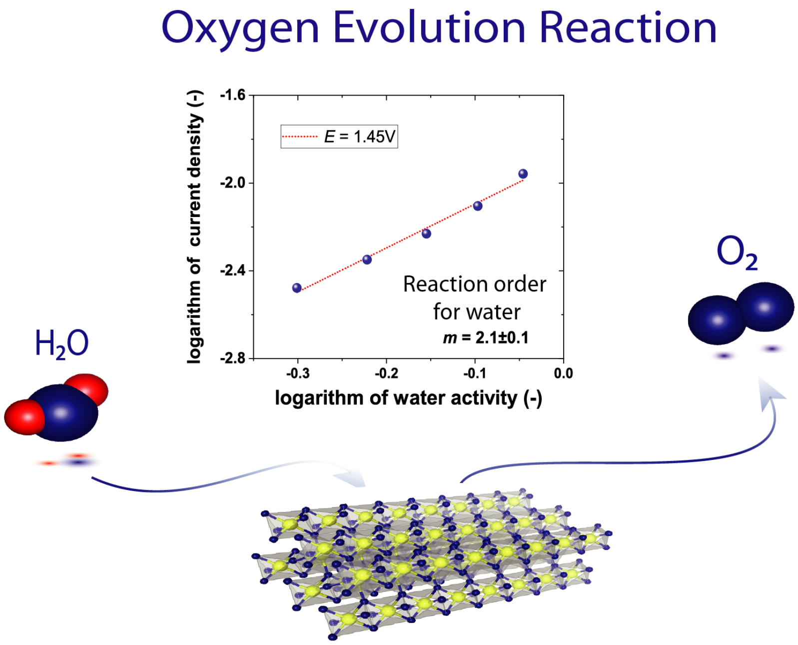

Scientific
Highlight

9

Improved Interfacial Stability of Ni-rich Oxide Lithium-Ion Battery Full Cells

We have identified a novel electrolyte additive, allowing an extended voltage range of Ni-rich oxide battery cells, while keeping excellent performance. The instability of the cathode–electrolyte interface causes the structural degradation of cathode active material and the electrolyte consumption, resulting in a rapid capacity fading and shortening battery life-time. The PSI-identified additive helps to alleviate these problems and extend battery life-time.

Ni-rich layer oxides $\mathrm{Li[Ni_xCoyMn_{1-x-y}]O_2}$ ($x \geq 0.8$, NCMs) are promising advanced cathode materials for high-energy density Li-ion batteries (LIBs) because of their high specific capacity (≥ 200 $\mathrm{mAh\,g^{-1}}$), as compared to the commercialized cathode materials. The even higher energy density of these materials can be achieved by increasing the upper cut-off voltage of the cell, as the energy density depends not only on capacity but also on the voltage of the cell. However, the instability of the cathode–electrolyte interface, especially under increased voltage range conditions, causes the structural degradation of cathode active material as well as the electrolyte consumption and gas evolution due to oxidative decomposition of electrolyte, resulting in a rapid capacity fading. Thus, improvement of the cathode–electrolyte interphase stability is a key requirement to inhibit Ni-rich active-material structural degradation and enhance its electrochemical performance. Therefore, interfacial stabilization through a novel multifunctional electrolyte additive is of high importance not only for the scientific community but also for industry and consumers.

Density-functional-theory calculations, combined with linear sweep voltammetry and cyclic voltammetry suggest that our multifunctional electrolyte additive is oxidized and reduced prior to the solvents, contributing to stable protective surface films on either of the electrodes. Cycling tests confirm that the addition of as little as 1 wt% of multifunctional electrolyte additive significantly reduces the cell polarization and improves the long-term cycling of the cell, which delivers a maximum capacity of 198 $\mathrm{mAh\,g^{-1}}$, with excellent capacity retention of 84% after 100 cycles at a cycling rate C/5, in

Figure 9
With the use of the novel multifunctional electrolyte additive, a significantly improved cycling performance is observed, in contrast to a rapid performance fade of the full-cell with conventional electrolyte. The development and propagation of cracks with cycling is reduced through the interfacial stabilization of the cathode with the PSI-identified multifunctional electrolyte additive.

contrast to a rapid performance fade of the cell with conventional electrolyte.

The greatly improved electrochemical performance is attributed to the participation of the additive in forming a stable surface protective film at the Ni-rich cathode surface, effectively reducing crack formation, metal-dissolution, and structural degradation of the cathode upon long-term cycling in an elevated voltage window. Moreover, the additive forms a stable solid electrolyte interphase (SEI) layer on the graphite anode, improving cycling performance of the full-cell.

In addition, the identification of this electrolyte additive gives an insight into the design principles of electrolyte additives for high energy-density LIBs and allows to realize stable electrode–electrolyte interfaces.

Publications

Multifunctional Electrolyte Additive for Improved Interfacial Stability in Ni-rich Layered Oxide Full-Cells.
Hieu Quang Pham, Marta Mirolo, Mohamed Tarik, Mario El Kazzi, Sigita Trabesinger.
Energy Storage Materials 33, 216–229 (2020).
DOI: 10.1016/j.ensm.2020.08.026

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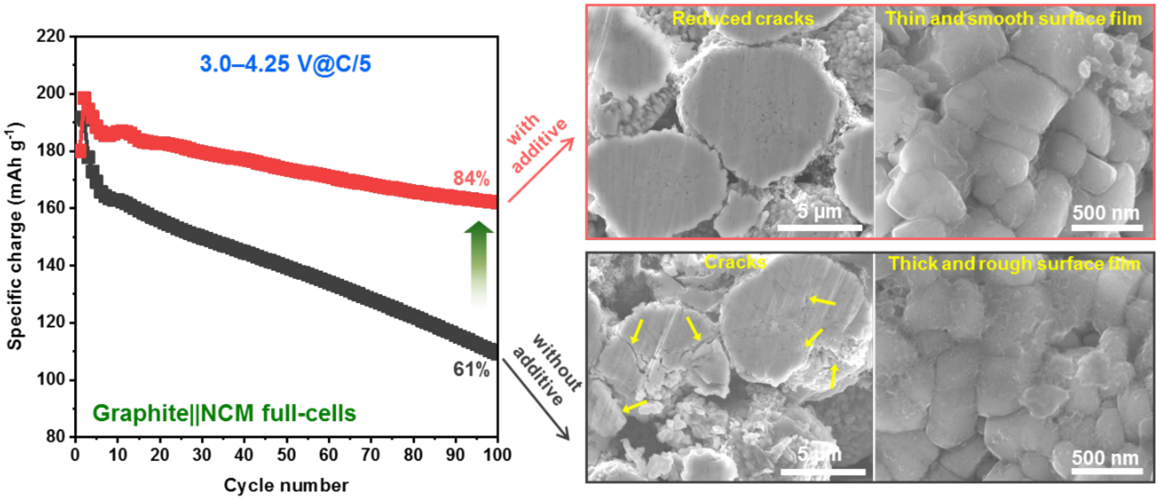


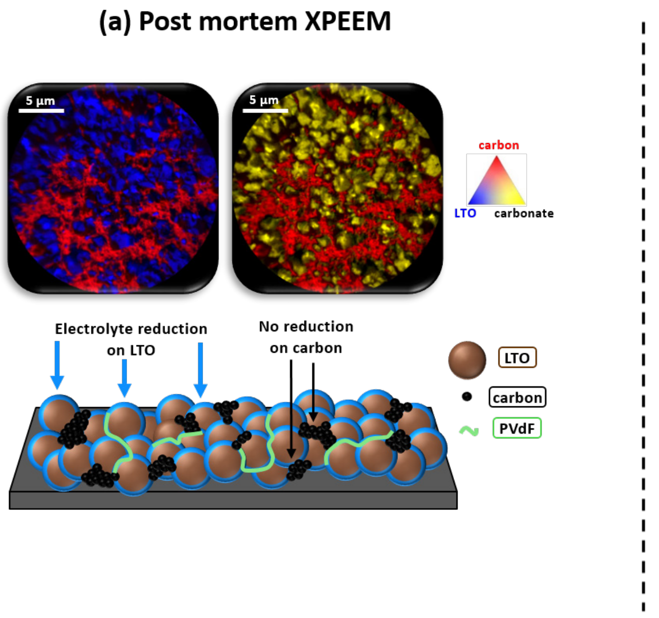
Scientific Highlight

10

Post Mortem/Operando XPEEM for Studying the Surface of Single Particles in Li-Ion Battery Electrodes

X-ray photoemission electron microscopy (XPEEM) with its excellent spatial resolution is a well-suited technique to elucidate the complex electrode-electrolyte interface reactions in Li-ion batteries. It provides element-specific contrast images and enables the acquisition of local X-ray absorption spectra on single particles. Here we demonstrate the strength of post mortem measurements and we show the first electrochemical cell dedicated for *operando* experiments in all-solid-state batteries.

The continuous need for increasing the electrochemical performance and safety of Li-ion batteries requires an incessant development of characterization techniques capable of providing better insights into the physics and chemistry of the various parts of the battery, in particular, the composite electrodes. Both, the bulk and the surface structural modifications of the electrodes, need to be investigated upon cycling, since they both have a direct impact on the battery performance. Specifically, understanding the different reactions taking place at the interface between the electrolyte and the various particles of the electrode remains crucial



for the development of stable, efficient, and safe batteries. However, due to the inherent difficulty of probing the confined interfacial regions, a full picture of the processes remains elusive, despite being the target of extensive investigation in the last three decades.

In this context, we explored the potential of synchrotron radiation X-ray photoemission electron microscopy (XPEEM) to study single particles in composite electrodes of Li-ion batteries. We have shown that the simultaneous investigation of the transition metal oxidation states in the lithium oxide particles, the stability of non-active components (i.e., conductive carbon and binder), and of the electrolyte upon cycling can be carried out using XPEEM, at both the microscopic and spectroscopic levels. By varying the energy of the incoming photons, local elemental maps of the sample can be obtained with a spatial resolution better than 70 nm. The limited escape depth of electrons from materials, in the range of 2–3 nm for low energy electrons, makes this technique very surface sensitive and particularly well suited for the study of surfaces and interfaces. Another intrinsic advantage of XPEEM resides in its non-destructive nature: in our case a 13 mm disc of a commercial-like electrode can be transferred to the microscope without prior sample thinning or grinding or

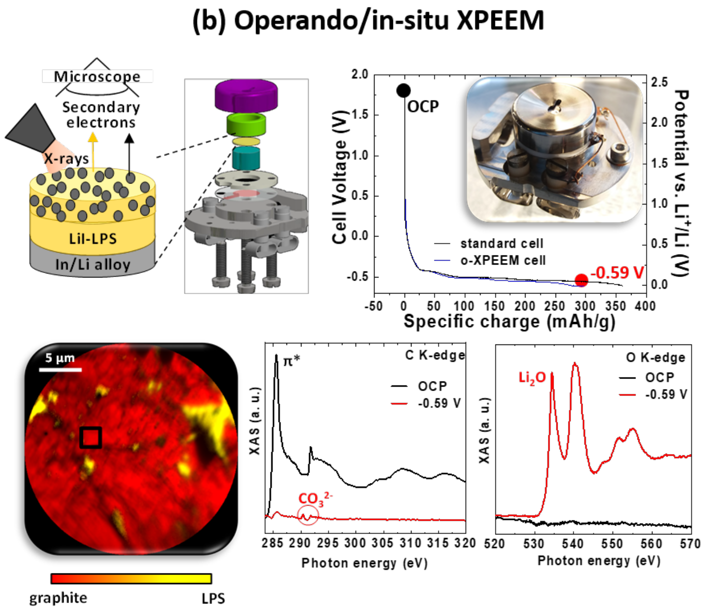


Figure 10

(a) Elemental map of a lithiated LTO electrode in carbonate-based electrolyte, showing the complementary position of LTO and carbon particles. The carbonate is preferentially covering the LTO surface.

(b) Discharge profile of the operando XPEEM cell hosting the all-solid-state stack graphite/LiI-LPS/InLi compared to the standard cell. Elemental mapping of the graphite electrode at OCP state. XAS spectra acquired on the graphite area at OCP and at the lithiated state for the C and O K-edges.

changing the working environment of the particles in general, as required for other microscopy techniques.

In this study, we discuss in detail the technical aspects that need to be considered in order to use XPEEM for studying Li-ion battery electrodes and our strategy to solve such challenges. First, we outline how we solved several technical problems, such as surface roughness, spectral normalization, and beam damage. Then, several case studies are presented to highlight the usefulness of combining lateral resolution and surface sensitivity with chemical information. Finally, we present a first XPEEM electrochemical cell dedicated for *operando/in situ* experiments in all-solid-state batteries. Measurements were carried out on a graphite electrode cycled with LiI-incorporated sulfide-based electrolyte. This measurements demonstrate the strong thermodynamic competitive reactions between the lithiated graphite surface and the Li_2O formation caused by the reaction of the intercalated lithium with the residual oxygen and water in the ultra-high vacuum environment. Moreover, we show the versatility of the *operando* XPEEM cell to investigate other active materials, e.g. $Li_4Ti_5O_{12}$.

Publications

Post mortem and operando XPEEM: a surface-sensitive tool for studying single particles in Li-ion battery composite electrodes.
Marta Mirolo, Daniela Leanza, Laura Höltschi, Christian Jordy, Vincent Pele, Petr Novák, Mario El Kazzi, Carlos A. F. Vaz.
Anal. Chem. 92, 3023–3031 (2020).
DOI: 10.1021/acs.analchem.9b04124

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Analysis of Cation Contamination of Polymer Electrolyte Water Electrolysers (PEWEs)

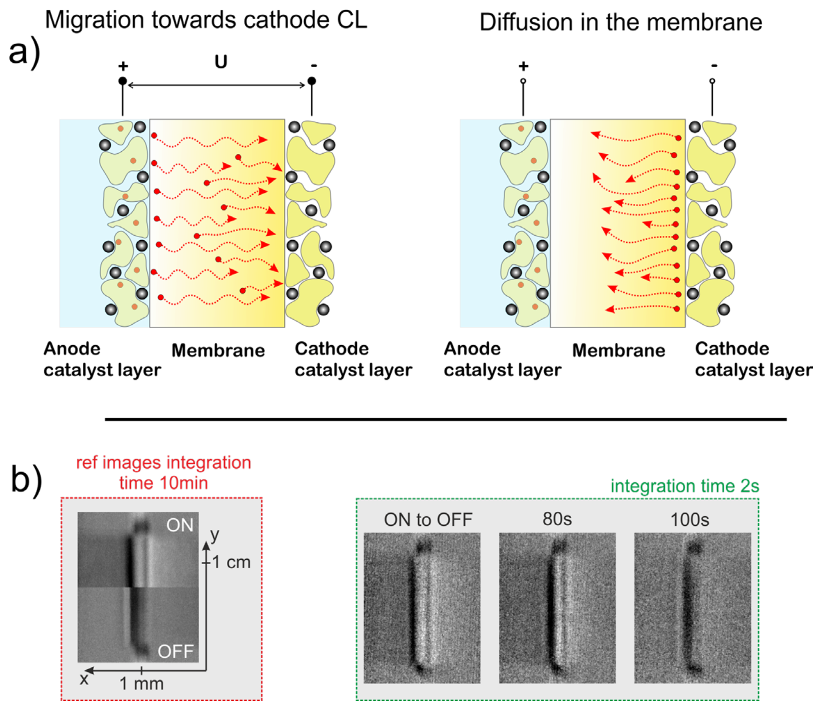
With the help of in situ PEWE regeneration methods, we can potentially enable the treatment of degraded cells without the necessity of stack disassembly, saving operation costs of the plant. In this context, we observed the movement of cations in a PEWE cell using neutron imaging and compared it with a model. This model is expected to be useful for the early detection of cation contamination problems in PEWEs, and the monitoring of *in situ* regeneration.

We are experiencing rapid changes in the energy sector towards green sources, and hydrogen technologies are on the verge of a breakthrough. The hydrogen, however, has to be produced using emission-free devices such as polymer electrolyte water electrolyzers (PEWE) if we are planning to become truly environment friendly. For large scale use, this clean technology has to be economically viable, which is why it is necessary to bring operation and capital costs further down.

One of the ways to reduce the operating costs is to prolong the lifetime of the PEWE stack by mitigating the deterioration of the performance. Among many degradation factors, the contamination caused by cationic impurities in feed water is a major problem in PEWE. The good news is that this type of damage is reversible and we believe that our efforts can enable easy diagnostics and the regeneration without the need to disassemble the electrolyzer stack. Using neutron radiography and electrochemical characterization techniques, we found that the contaminants have a different impact on the cell depending on their position across the catalyst-coated membrane (CCM). To obtain a fundamental understanding of why and how this happens we performed multiple neutron imaging experiments using Gd^{3+} as a model contaminant due to its high neutron cross-section and developed a model, which was tuned using experimental data.

It is common knowledge that two main forces govern the dynamics of ionic movement and positioning in the eletrolyser membrane. Diffusion, which is driven by the ion concentration gradient and migration, which is driven by the electric field acting on charged particles during the cell operation. The cation distribution in the CCM depends on the equilibrium between these two forces. We performed steady-state measurements to measure the absolute amount of contaminants in the membrane as well as to observe the final ionic distribution during operation and standby of the electrolyzer. In the next step, we measured transients to obtain time constants between the steady states, which allowed to calculate the transport properties of the gadolinium ion in the membrane. Coupling these measurements with electrochemical analysis, we obtained conclusive results. We found that upon startup of the contaminated PEWE cell, which was on standby for an extended period one could expect an exceptionally high voltage peak, which quickly decays. The voltage and membrane conductivity quickly recovers as the cations migrate to the neighborhood of the cathodic catalyst layer (CL). It takes around 10 seconds for the cations to accumulate in the cathode CL and over 10 minutes to diffuse back to the bulk of the membrane after shutdown. Using data from quantified neutron images and electrochemical overpotential breakdown analysis, we found that even very small amounts of cations can severely impair cell performance. The fraction of occupied exchange groups (2.5%) does not match the membrane resistance increase (10%). The reason behind this higher than expected impact most likely stems from additional yet unknown mechanisms (e.g. polymer morphology alteration). Since the membrane resistance is affected only by the cations remaining in the bulk of the membrane, we performed cyclic current density ramp up and down measurements to confirm that the amount of the cations in the membrane will follow the membrane resistance values. We observed a very good agreement between the two and concluded that performing polarization curve measurements cyclically can be a good way to check for ionic presence in the membrane – identified as a hysteresis between the up and down current sweeps.

As the next step, we developed a model, which predicts the distribution and movement of these



ions. We tuned and validated the model using our neutron imaging data. The model was later set-up to simulate the cell contaminated with Fe^{3+} to simulate the cyclic polarization curve measurement, which was measured using a different PEWE cell. Once again, we found good agreement between the simulation and experiments confirming the method's effectiveness. The article also discusses the transport properties of ions in the membrane and provides values for diffusion and migration coefficients of Gd^{3+} ions in Nafion based membranes. The full article gives more in-depth information on cationic contamination of PEWE systems.

Figure 11
Movement of the cations in the membrane under the effect of the electric field.
A) Schematic indicating the direction and behavior of cationic impurities during operation and standby of the PEWE.
B) Neutron radiographs of reference conditions representing steady states and transient back diffusion process after a long operating period.

Publications

Dynamic Neutron Imaging and Modeling of Cationic Impurities in Polymer Electrolyte Water Electrolyzer.
M. Zlobinski, U. Babic, M. Fikry, L. Gubler,
T.J. Schmidt, P. Boillat.
J. Electrochem. Soc. 167, 144509 (2020).
DOI: 10.1149/1945-7111/abc83b

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Financial support from the Swiss Federal Office of Energy (SFOE), grant ELY-DEG SI/501198-01 is gratefully acknowledged. T.J. Schmidt thanks Innosuisse and the Swiss Competence Center for Energy Research (SCCER) Heat & Electricity Storage. The authors kindly acknowledge Oxford Instruments for providing the Andor camera, which was used to record the neutron images.

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Scientific Highlight

12

Oxygen Evolution Reaction Activity and Underlying Mechanism of Perovskite Electrocatalysts at Different pH Values

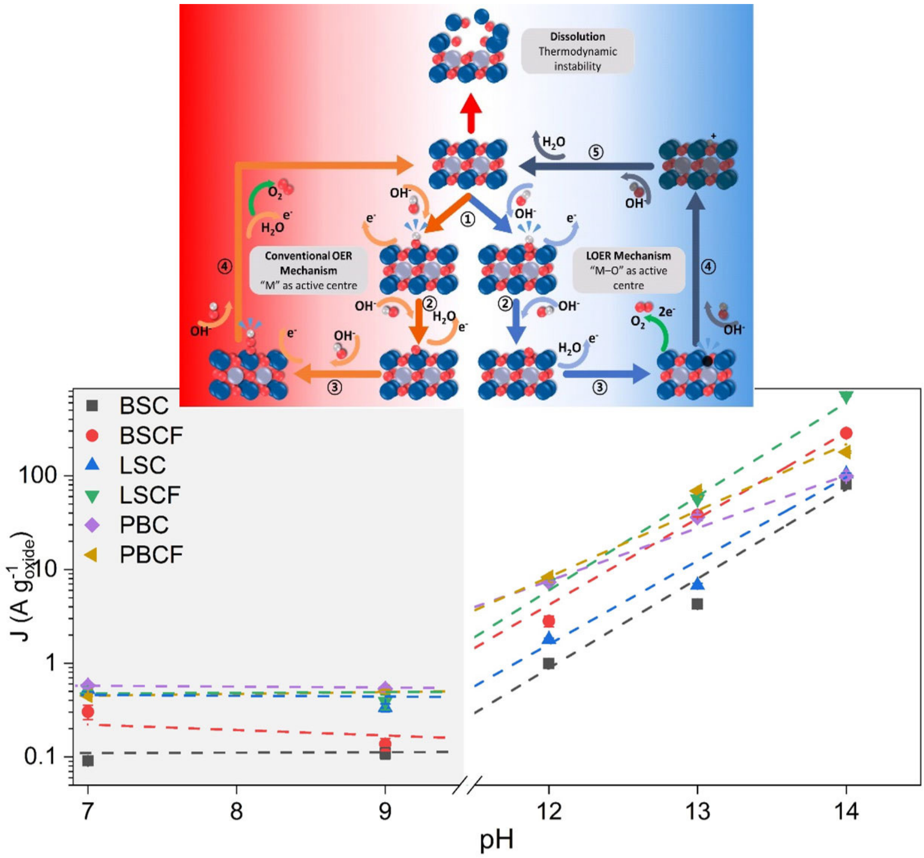
We have studied how the electrolyte pH values influence the oxygen evolution reaction (OER) activity and stability of different promising perovskite oxide catalysts for application as anodic electrodes in alkaline water electrolyzers. The OER activity and stability decreased lowering the electrolyte pH values. By combining electrochemical studies and *operando* X-ray absorption spectroscopy measurements, it has been suggested that different reaction mechanisms dominate in alkaline and near-neutral electrolyte pH region.

Energy conversion and storage technologies are drawing growing attention for mediating inconsistent power generation from renewable energy sources. Water electrolysis utilizes hydrogen as an energy vector to store large amounts of energy, and typically operates either in acidic or alkaline environment. Particularly, operation under alkaline conditions allows exploring abundant and low-cost non-noble metals as electrocatalysts, such as perovskite oxides. However, KOH solutions as electrolyte present vulnerability to carbonation during operation, which shifts the pH level towards near-neutral values and decreases the electrolyte conductivity.

In the present study the oxygen evolution reaction (OER) activity and stability versus different electrolyte pH levels have been investigated for a series of perovskite oxides prepared via flame spray synthesis, namely $Ba_{0.5}Sr_{0.5}CoO_{3-\delta}$ (BSC), $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (BSCF), $La_{0.5}Sr_{0.5}CoO_{3-\delta}$ (LSC), $La_{0.2}Sr_{0.8}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (LSCF), $PrBaCo_2O_{6-\delta}$ (PBC), and $PrBaCo_{1.6}Fe_{0.4}O_{6-\delta}$ (PBCF). The studied perovskites showed a first-order relationship between OER activity and electrolyte pH in the high alkaline region while revealing zeroth-order relationship in the near-neutral pH region. Differences in the OER activity versus pH, suggest that different mechanisms are predominant in the different pH regions. From *operando* X-ray absorption spectroscopy (XAS) measurements of the catalysts during the OER process, we could observe the development of Co/Fe-oxy(hydroxide) species only alkaline environment. The formation of an oxyhydroxide layer on perovskite surface is generally associated to the occurrence of a different OER mechanism than the conventional one, which involves oxidation of lattice oxygen and it is named lattice oxygen evolution reaction (LOER) mechanism. Differently, in the near neutral region the development of a Co/Fe-oxy(hydroxide) surface layer was not visible. This suggests that the studied perovskite catalysts are more inclined to evolve oxygen through the LOER mechanism in a highly alkaline condition, while in near-neutral environment the convetional OER mechanism dominates. However, a catalyst would not perform oxygen evolution through only one of the two mechanisms discussed in here, but it would rather preferentially and proportionally take both paths (conventional and LOER) as they are thermodynamically entangled.

Figure 12

On the bottom: Tafel plots are constructed from series of chronoamperometry measurements comparing OER activities at different pH levels for each prepared catalyst. Markers represent the current densities (J expressed as current density as A/g_{oxide}) at 1.58 VRHE of the studied perovskites in logarithmic scale with respect to pH depicting the order of activity. The investigated perovskites are: $Ba_{0.5}Sr_{0.5}CoO_{3-\delta}$ (BSC), $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (BSCF), $La_{0.5}Sr_{0.5}CoO_{3-\delta}$ (LSC), $La_{0.2}Sr_{0.8}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (LSCF), $PrBaCo_2O_{6-\delta}$ (PBC), and $PrBaCo_{1.6}Fe_{0.4}O_{6-\delta}$ (PBCF).
On the top: sketch of the conventional OER vs. LOER mechanism.



Publications

Oxygen Evolution Reaction Activity and Underlying Mechanism of Perovskite Electrocatalysts at Different pH.
Bae-Jung Kim, Emiliana Fabbri, Mario Borlaf, Daniel F. Abbott, Ivano E. Castelli, Maarten Nachtegaal, Thomas Graule, Thomas J. Schmidt.
Mater. Adv., 2, 345–355 (2021).
DOI: 10.1039/D0MA00661K

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DISSERTATIONS 2020

Fe/N/C-type Oxygen Reduction Reaction Catalysts – From Synthesis to Advanced *In Situ* Spectroscopic Characterization

Kathrin Ebner

Ph.D. Thesis No. 27058, ETH Zürich, September 2020.

Examiners:
Prof. Dr. T.J. Schmidt, PSI Villigen/ETH Zürich
Prof. Dr. M. Kovalenko, ETH Zürich
Dr. J. Herranz, PSI Villigen

Abstract

The platinum-based catalysts customarily used in polymer electrolyte fuel cells (PEFCs) substantially contribute to the excessive cost of these energy conversion devices. Thus, an approach to foster the commercialization of this technology is to substitute these costly materials with inexpensive counterparts, especially for the more catalytically demanding oxygen reduction reaction (ORR).

The most promising, Pt-free ORR-catalysts identified so far are Fe-based materials on an N-doped carbon matrix (sometimes referred to as Fe/N/C-catalysts). The last years have witnessed significant improvements in these materials' performance, but often at the expense of using expensive precursors to synthesize them, thus compromising their initial low-cost aim.

With this motivation, the first part of Kathrin's PhD work focused on the development of a novel Fe/N/C-catalyst synthesis approach using inexpensive precursors that led to materials with initial ORR-activities comparable to those of the best catalysts of this class. Furthermore, upon attempting to characterize these materials with ^{57}Fe-sensitive Mössbauer spectroscopy, Kathrin found that the use of ^{57}Fe-enriched precursors in Fe/N/C-synthesis impacted the catalysts' iron speciation and ORR-activity. These results put into question the common practice to extend conclusions derived from samples synthesized with ^{57}Fe-enriched precursors to their non-enriched analogues, and highlight the importance of verifying the comparability of the catalysts' characteristics prior to doing so.

Then, in the second part of her thesis, Kathrin's work shifted towards the understanding of the nature of the active sites and the catalytic process in these materials using advanced, *in situ* spectroscopic techniques. Specifically, she applied *in situ* modulation-excitation X-ray absorption spectroscopy to shed light onto the

potential-induced changes in the coordination environment of the N-coordinated Fe-ions that are believed to constitute the ORR-active centers in these materials. This was complemented by *in situ* X-ray emission spectroscopy measurements that showed that these conformational changes are accompanied by a change of the sites' spin state.

Thus, these results provide unprecedented insight on the potential-induced changes undergone by the ORR-active sites in these catalysts, and pave the road for the development of better performing materials that could significantly decrease the costs of PEFCs.

Multiple scales *post mortem* and *operando* XPS, XAS and XPEEM investigations of reactivity at interfaces in Li-ion batteries

Marta Mirolo

Ph.D. Thesis No. 27215, ETH Zürich, November 2020.

Examiners:
Prof. Dr. Petr Novák, PSI Villigen/ETH Zürich
Prof. Dr. Markus Niederberger, ETH Zürich
Dr. Mario El Kazzi, PSI Villigen
Dr. Carlos A.F. Vaz, PSI Villigen

Abstract

The understanding of the electrode-electrolyte interface in general and of the degradation reaction mechanisms at the surface of the active materials in particular remain crucial topics in the search for high energy density and longevity next generation of Li-ion batteries. For both liquid and solid-state electrolytes, the interface stability challenges associated with the electrolytes and the surface structure of the active materials arise when the cell operates at high working voltages.

In this thesis, Marta highlights how the combination of X-ray photoelectron spectroscopy (XPS), X-ray photoemission electron microscopy (XPEEM) and X-ray absorption spectroscopy (XAS) can provide a reliable platform to investigate the electrode-electrolyte interface and the surface modifications of the active materials. The complementarity of these techniques allows an in-depth analysis from the surface (few nm) to the bulk (few hundreds of nm) of the active material particles and with different lateral resolutions, thus probing from single particles to the overall electrode.

The first part of the thesis is dedicated to the combination of post mortem measurements to investigate electrodes cycled in liquid electrolytes. The $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ (NCA) cathode is thoroughly investigated at different states of charge, clarifying the role during cycling of the adventitious carbonate on the pristine NCA powder and the charge compensation mechanisms occurring during (de-)lithiation. In particular, at the high potential of 4.9 V vs. Li^+/Li, oxygen redox activity is observed on the NCA surface, accompanied with partial dissolution of the transition metals, which are subsequently detected after long-term cycling on the $Li_4Ti_5O_{12}$ (LTO) counter electrode.

In the second part of the thesis, Marta discusses the development of electrochemical cells for

operando and *in situ* analysis, using a solid-state electrolyte to adhere to the vacuum requirement of XPS, XAS and XPEEM techniques. The combination of *in situ* XAS and *operando* XPS analysis during (de-)lithiation of SnO_2 conversion-alloy anode material allows to identify with remarkable precision the potential at which the different redox reactions take place. The versatility of the electrochemical cell for operando XPS measurements also allows the *in situ* XAS monitoring of the transition metals oxidation state in the case of the cathode $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ (NCM111). Finally, a newly developed electrochemical cell for in situ XPEEM is presented, permitting to localise with high lateral resolution the reactions occurring on the surface of the active materials and the solid-electrolyte.

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

Tobias Schuler

Ph.D. Thesis No. 26835, ETH Zürich, September 2020.

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

Abstract

The social demand of green, CO_2 free product upgrading and balancing of renewable energy requires technologies such as polymer electrolyte water electrolysis (PEWE) for production of green hydrogen and converting electrical energy into storable chemical energy.

The scope of the thesis aims at the fundamental understanding of the role of porous transport layers (PTLs) in polymer electrolyte water electrolysis (PEWE). The missing-link between the microstructure of PTL and cell efficiency, governed by the so-called overpotentials associated to kinetics, ionic and electric transport as well as mass transport is elucidated. A combination of X-ray tomographic microscopy (XTM) and in-depth electrochemical analysis is employed. Ultimately, the deduction of an innovative, efficient PTL concept is pursued.

Morphological properties such as porosity, pore and particle size distributions, transport parameters as well as surface properties, e.g. surface roughness, were characterized by XTM. A systematically designed matrix based of six fibre Ti PTLs allowed for controlled analysis of bulk as well as surface properties. Mechanical interference between the ductile membrane, catalyst layer and the PTL were investigated by *post operando* XTM deformation analysis. A plastic membrane deformation of up to 25 % of the entire catalyst coated membrane thickness was observed. A novel concept based on combination of membrane deformation and PTL surface characterization provided access to interfacial contact area between catalyst layer and transport layer. In-depth electrochemical characterization of the PTL matrix revealed that the catalyst layer is only partially utilized. The utilization is scaling with the interfacial contact area. Deconvolution of the mass transport loss into its sub-resistances shows the governing role of the catalyst layer resistance for the partial catalyst utilization. Thus, the conventional

correlation between mass transport losses and PTL bulk properties is proved questionable. The transport resistance shows a local resistance behavior as previously observed in the field of proton exchange membrane fuel cells.

For the first time, all losses are systematically correlated to bulk and surface properties of PTLs revealing that the interfacial contact area impacts all loss categories. The novel insights enabled the deduction of generic design guidelines for PTL in the field of PEWE. Theses novel insights were then employed for designing novel, efficient PTL structures.

In the third part, a novel class of hierarchically structured, multi-layer PTL (ML-PTL) was introduced being comprised of a coarse support layer for mechanical integrity and a fine structured microporous top-layer for high interfacial contact area with low surface roughness. The impact of the particle size of the fine layer on performance was evaluated. Synchrotron and laboratory based XTM showed the superior surface properties of ML-PLTs versus single layer only materials, reducing membrane deformation by a factor of two and surface roughness up to a factor of three. The in-depth electrochemical analysis revealed an increase of catalyst utilization by a factor of 2.8 and the reduction of mass transport losses by ~40 %. Established stack technology, commercially available Ti sintered PTLs and surface treated commercial Ti sintered PTLs were outperformed.

For the first time, thin membranes of ~20 µm thickness were employed in a technical cell configuration with Ti PTLs resulting in loss reduction of more than 200 mV at 2 A cm^{-2} versus state-of-the-art single layer sintered PTLs and 125 µm membranes. The improvement in performance (MPL effect) is identified to be due to increasing catalyst utilization. The underlying mechanism of partial catalyst utilization is related to microstructural changes in the catalyst layer when exposed to liquid water environment affecting the electrical percolation network and reducing in plane conductivity.

Investigation of Water and Gas Distribution in Proton Exchange Membrane Water Electrolyzer

Mateusz Zlobinski

Ph.D. Thesis No. 27132, ETH Zürich, September 2020.

Examiners:
Prof. Dr. T.J. Schmidt, PSI Villigen/ETH Zürich
Prof. Dr. T. Lippert, PSI Villigen/ETH Zürich
Dr. P. Boillat, PSI Villigen

Abstract

The content of the thesis on polymer electrolyte water electrolyzers (PEWEs) can be divided into two subjects: water and gas distribution in porous transport layers (PTLs) and cationic contamination of the catalyst coated membranes (CCM). Both projects rely on neutron imaging results to tackle separate issues. The motivation behind both of these research directions was to underline the sources of losses within the proton exchange membrane (PEM) on the fundamental level and identify possible cell performance and durability improvement areas.

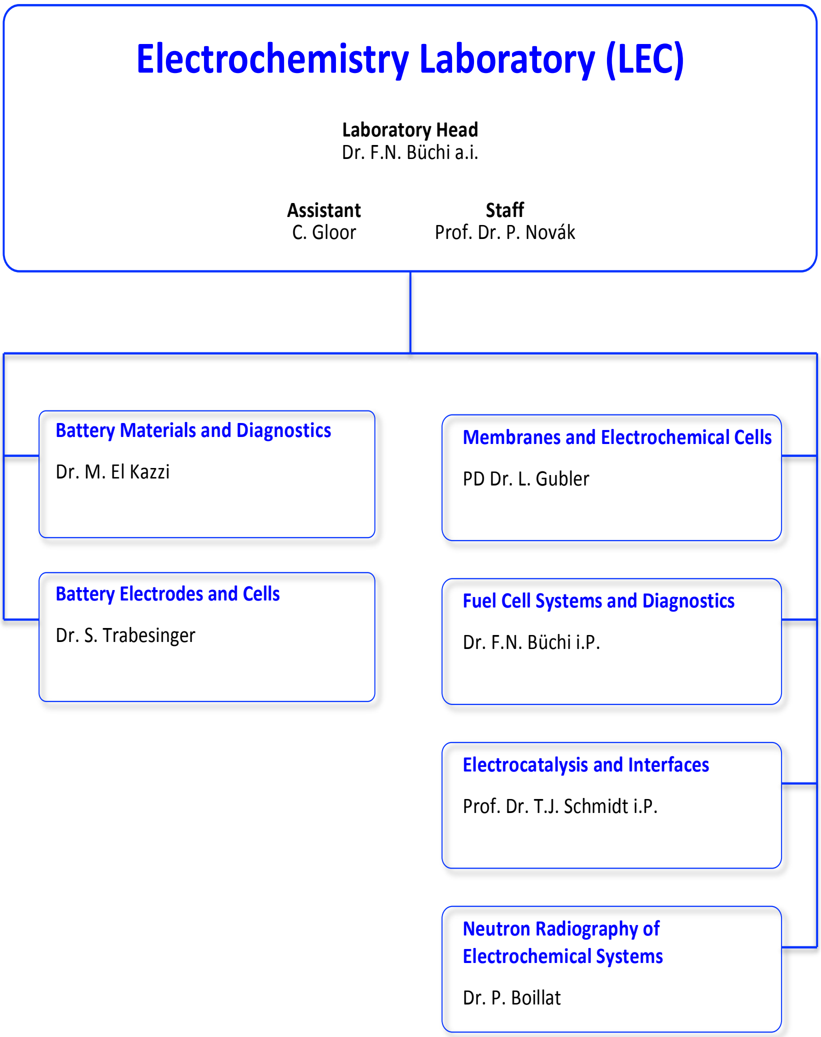
The first part of the thesis describes transient and steady-state 2-phase flow mechanisms in the PTLs. The flow character, gas pathway establishment and re-imbibition of these pathways with water are discussed in detail. Followed by an investigation with the introduction of the microporous layer on top of the PTL and its influence on water management in the PEWE cells. Later on water and gas distribution is analyzed in the frame of fundamental understanding of 2 phase flow in porous media and its implications related to the potential impact on the durability and performance.

The second part of the dissertation presents the analysis of the distribution of cationic contaminants in the CCM based on neutron images. Using Gd^{3+} ions, it was possible to obtain enough contrast and to trace the accumulation of positively charged ions in the CCM while the cell was operating. Thanks to the results, a novel approach of *operando* regeneration of the PEWE cell was developed and led to the presented proof of concept. The study also focuses on transient phenomena and electro diffusive motion of the cationic species in the contaminated CCM. The simulation designed using the data input from neutron imaging was successfully correlated with the results from previous experiments potentially identifying a new diagnostic tool for PEWE affecting the electrical percolation network and reducing in plane conductivity.

THE ELECTROCHEMISTRY LABORATORY

FACTS & FIGURES

STRUCTURE 2020



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Membranes and Electrochemical Cells
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Fuel Cell Systems and Diagnostics
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Electrocatalysis and Interfaces
Prof. Dr. T.J. Schmidt i.P.

Neutron Radiography of Electrochemical Systems
Dr. P. Boillat

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Members of the Electrochemistry Laboratory in 2020.

AWARDS 2020

Prof. Dr.-Ing. Petr Nóvak

IBA Yeager Award

For Distinguished and Pioneering Carreer-Long Scientific and Technological Contributions to Advanced Characterization and Electrochemical Method Development for Secondary Batteries.

Event:	IBA-2020 Conference and Meeting, International Battery Materials Association (IBA), Bled, Slovenia, March 9–13, 2020

Dr. Muriel Siegwart

2020 Young Scientist Prize of the Swiss Neutron Science Society

Neutron Transmission Imaging of Fuel Cells using Time-of-Flight and Dark-Field Imaging.

Event:	General Assembly of the Swiss Neutron Science Society, Paul Scherrer Institut, Villigen, Switzerland, October 29, 2020

BACHELOR AND MASTER STUDENTS

Lea Marti ETH Zürich

Investigation of Low-Loading and Alternative Catalysts for Electrochemical CO_2 Separation

February – July 2020 (Membranes and Electrochemical Cells)

Andreas Michalski ETH Zürich

A Numerical Analysis of Water Vapor Transport in Polymer Electrolyte Fuel Cells

April – October 2020 (Fuel Cell Systems and Diagnostics)

Ambra Maria Van Liedekerke ETH Zürich

Correlation Between Crack Formation and Structural Evolutions in $Li[Ni_xCo_yMn_y]O_2$ Layered Oxide Cathode Materials during Charge and Discharge Process

September – December 2020 (Battery Electrodes and Cells)

Moritz Bohn Technical University Munich, Munich, Germany

Gas-phase surface modification of solid electrolyte materials

September 2020 – January 2021 (Battery Materials and Diagnostics)

Matteo Lecchi

Politecnico di Milano, Milano, Italy

Investigations on the effect of selected membrane additives on fuel cell performance and degradation

September 2020 – March 2021 (Membranes and Electrochemical Cells)

Gian Müller

ETH Zürich

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

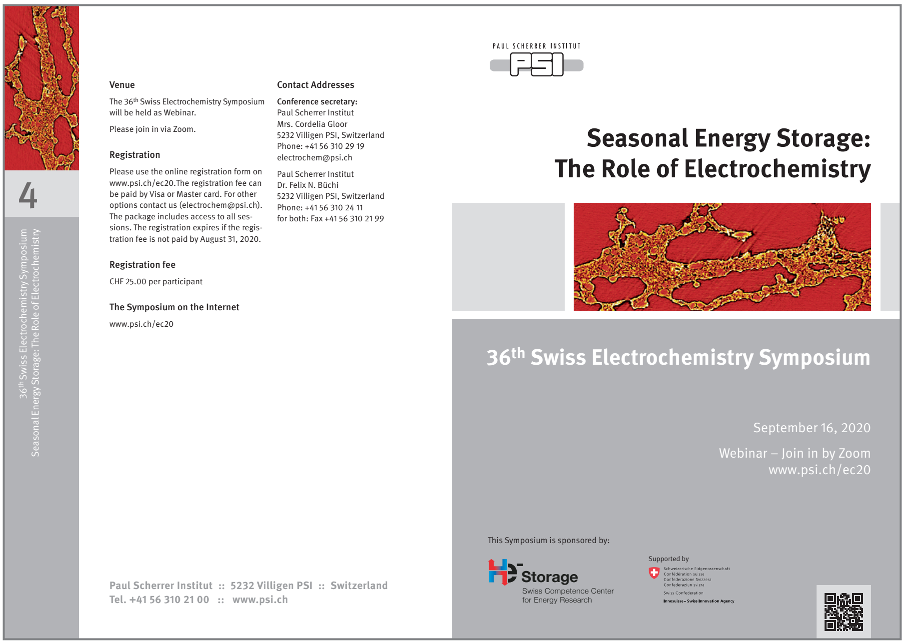
December 2020 – June 2021 (Fuel Cell Systems and Diagnostics)

36th Swiss Electrochemistry Symposium

September 16, 2020
Webinar via Zoom

Seasonal Energy Storage: The Role of Electrochemistry

Organizers	Felix N. Büchi, Lorenz Gubler, Cordelia Gloor, Electrochemistry Laboratory
Contributions from (in order of appearance)	Peter Strasser, Technical University Berlin, Germany Stefan Diethelm, SOLIDpower SA, Yverdon, Switzerland Dominik Härle, Fraunhofer-Institut IMWS, Halle, Germany Toshiki Shimizu, Panasonic Corporation, Tokyo, Japan Markus Borck, Innolith Science and Technology GmbH, Bruchsal, Germany David Parra Mendoza, University of Geneva, Switzerland Günter Schmid, Siemens Gas and Power GmbH & Co. KG, Erlangen, Germany



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36th Swiss Electrochemistry Symposium
Seasonal Energy Storage: The Role of Electrochemistry

Venue

The 36th Swiss Electrochemistry Symposium will be held as Webinar.

Please join in via Zoom.

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Please use the online registration form on www.psi.ch/ec20.The registration fee can be paid by Visa or Master card. For other options contact us (electrochem@psi.ch). The package includes access to all sessions. The registration expires if the registration fee is not paid by August 31, 2020.

Registration fee

CHF 25.00 per participant

The Symposium on the Internet

www.psi.ch/ec20

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Seasonal Energy Storage:
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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 2020.
PD Dr. L. Gubler	*Electrochemistry: Fundamentals, Cells & Applications* ETH Zürich, HS 2020.
Prof. Dr. T.J. Schmidt	*Physical Electrochemistry & Electrocatalysis* ETH Zürich, FS 2020.

Contributions to Courses

PD Dr. L. Gubler	*Energy Storage Systems* Lucerne University of Applied Sciences and Arts, March 25, 2020.
Dr. S. Trabesinger	*Analytical Strategy* ETH Zürich, September 29, 2020.

Publications

Peer Reviewed Papers

D. Aegerter, M. Borlaf, E. Fabbri, A.H. Clark, M. Nachtegaal, T. Graule, T.J. Schmidt	*Tuning the Co Oxidation State in $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$ by Flame Spray Synthesis Towards High Oxygen Evolution Reaction Activity* Catalysts **10**, 984 (2020). DOI: 10.3390/catal10090984
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R. Asakura, C. Bolli, P. Novák, R. Robert	*Insights into the Charge Storage Mechanism of Li_3VO_4 Anode for Li-Ion Batteries* ChemElectroChem **7** (9), 2033–2041 (2020). DOI: 10.1002/celc.202000161
U. Babic, M. Tarik, T.J. Schmidt, L. Gubler	*Understanding the effects of material properties and operating conditions on component aging in polymer electrolyte water electrolyzers* Journal of Power Sources **451**, 227778 (2020). DOI: 10.1016/j.jpowsour.2020.227778
H. Ben youcef, D. Henkensmeier, S. Balog, G.G. Scherer, L. Gubler	*Copolymer synergistic coupling for chemical stability and improved gas barrier properties of a polymer electrolyte membrane for fuel cell applications* International Journal of Hydrogen Energy **45** (11), 7059–7068 (2020). DOI: 10.1016/j.ijhydene.2019.12.208Get

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M. Cochet, A. Forner-Cuenca, V. Manzi-Orezzoli, M. Siegwart, D. Scheuble, P. Boillat	*Enabling High Power Density Fuel Cells by Evaporative Cooling with Advanced Porous Media* Journal of The Electrochemical Society **167**, 084518 (2020). DOI: 10.1149/1945-7111/ab8e82
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G. Ferraresi, S. Uhlenbruck, C.-L. Tsai, P. Novák, C. Villevieille	*Engineering of Sn and Pre-lithiated Sn as Negative Electrode Materials Coupled to Garnet Ta-LLZO Solid Electrolyte for All-Solid-State Li Batteries* Batteries & Supercaps **3** (6),557–565 (2020). DOI: 10.1002/batt.201900173
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Journal of Electroanalytical Chemistry **859**, 113832 (2020).
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C. Lawley, M. Nachtegaal, J. Stahn, V. Roddatis, M. Döbeli, T.J. Schmidt, D. Pergolesi, T. Lippert	*Examining the surface evolution of* $LaTiO_xN_y$ *an oxynitride solar water splitting photocatalyst* Nature Communications **11**, 1728 (2020). DOI: 10.1038/s41467-020-15519-y
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A. Mularczyk, Q. Lin, M.J. Blunt, A. Lamibrac, F. Marone, T.J. Schmidt, F.N. Büchi, J. Eller	*Droplet and Percolation Network Interactions in a Fuel Cell Gas Diffusion Layer* Journal of The Electrochemical Society **167**, 084506 (2020). DOI: 10.1149/1945-7111/ab8c85
A.P. Muroyama, A. Pătru, L. Gubler	*Review — CO_2 Separation and Transport via Electrochemical Methods* Journal of The Electrochemical Society **167**, 133504 (2020). DOI: 10.1149/1945-7111/abbbb9
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M. Siegwart, F. Huang, M. Cochet, T.J. Schmidt, J. Zhang, P. Boillat	*Spatially Resolved Analysis of Freezing during Isothermal PEFC Cold Starts with Time-of-Flight Neutron Imaging* Journal of The Electrochemical Society **167**, 064510 (2020). DOI: 10.1149/1945-7111/ab7d91

M. Siegwart, V. Manzi-Orezzoli, J. Valsecchi, R.P. Harti, M. Kagias, M. Strobl, C. Grünzweig, T.J. Schmidt, P. Boillat	*Operando Visualization of Water Distribution in Gas Diffusion Media of PEFCs with an Optimized Neutron Grating Interferometer* Journal of The Electrochemical Society **167**, 064509 (2020). DOI: 10.1149/1945-7111/ab7d92
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A.O. Tezel, S. Sunde, J.L. Gómez-Cámer, P. Novák, A.M. Svensson	*Effect of a Boron Based Anion Receptor on Graphite/*$LiFePO_4$ *Electrodes* Journal of The Electrochemical Society **167** (2), 020525 (2020). DOI: 10.1149/1945-7111/ab68c5
M. Ting, M. Burigana, L. Zhang, Y.Z. Finfrock, S. Trabesinger, A. Jonderian, E. McCalla	*Impact of Nickel Substitution into Model Li-Rich Oxide Cathode Materials for Li-Ion Batteries* Chemistry of Materials **32** (2), 849–857 (2020). DOI: 10.1021/acs.chemmater.9b04446
T.G. Tranter, P. Boillat, A. Mularczyk, V. Manzi-Orezzoli, P.R. Shearing, D.J.L. Brett, J. Eller, J.T. Gostick. A. Forner-Cuenca	*Pore Network Modelling of Capillary Transport and Relative Diffusivity in Gas Diffusion Layers with Patterned Wettability* Journal of the Electrochemical Society **167** (11), 114512 (2020). DOI: 10.1149/1945-7111/ab9d61
O. Vopička, P. Číhal, M. Klepić, J. Crha, V. Hynek, K. Trtík, P. Boillat, P. Trtik	*One-pot neutron imaging of surface phenomena, swelling and diffusion during methane absorption in ethanol and n-decane under high pressure* PLOS ONE **15**, e0238470 (2020). DOI: 10.1371/journal.pone.0238470

X. Wu, C. Villevieille, P. Novák, M. El Kazzi	*Insights into the chemical and electronic interface evolution of* $Li_4Ti_5O_{12}$ *cycled in* $Li_2S–P_2S_5$ *enabled by operando X-ray photoelectron spectroscopy* Journal of Materials Chemistry A **8** (10), 5138–5146 (2020). DOI: 10.1039/C9TA14147B
H. Xu, M. Bührer, F. Marone, T.J. Schmidt, F.N. Büchi, J. Eller	*Optimal Image Denoising for In Situ X-ray Tomographic Microscopy of Liquid Water in Gas Diffusion Layers of Polymer Electrolyte Fuel Cells* Journal of The Electrochemical Society **167**, 104505 (2020). DOI: 10.1149/1945-7111/ab9820
W. Yin, A. Grimaud, G. Rousse, A.M. Abakumov, A. Senyshyn, L. Zhang, S. Trabesinger, A. Iadecola, D. Foix, D. Giaume, J.-M. Tarascon	*Structural Evolution at the Oxidative and Reductive Limits in the First Electrochemical Cycle of* $Li_{1.2}Ni_{0.13}Mn_{0.54}Co_{0.13}O_2$ Nature Communications **11** (1), 1252 (2020). DOI: 10.1038/s41467-020-14927-4
M. Zlobinski, U. Babic, M. Fikry, L. Gubler, T.J. Schmidt, P. Boillat	*Dynamic Neutron Imaging and Modeling of Cationic Impurities in Polymer Electrolyte Water Electrolyzer* Journal of The Electrochemical Society **167**, 144509 (2020). DOI: 10.1149/1945-7111/abc83b
M. Zlobinski, T. Schuler, F.N. Büchi, T.J. Schmidt, P. Boillat	*Transient and Steady State Two-Phase Flow in Anodic Porous Transport Layer of Proton Exchange Membrane Water Electrolyzer* Journal of The Electrochemical Society **167**, 084509 (2020). DOI: 10.1149/1945-7111/ab8c89

Talks

Invited Talks

F.N. Büchi	*Measurement Technique for two-phase flow in porous media (FC)* 6th Symposium on Fluid Dynamics, Bosch, Rennigen, Germany, February 12, 2020.
J. Eller	*Sub-Second X-Ray Tomographic Imaging of Liquid Water Dynamics in PEFC Gas Diffusion Layers* Shell Research Seminar, Virtual, December 8, 2020.
M. El Kazzi	*Operando XPS pour Elucider des Réactions Electrochimiques Complexe dans les Batteries Tous Solide* Journées des Spectroscopies d'électrons Paris-Jussieu, France, January 20, 2020.
M. El Kazzi	*Advanced Characterization Techniques for Li-Ion Batteries* PhD days at the Institute of Nanotechnology of Lyon (INL), Lyon, France, November 25–26, 2020.
E. Fabbri	*Insight into Perovskite Oxide Catalysts for the Electrochemical Splitting of Water* CHAINS, Virtual, December 9, 2020.
P. Novák	*In Situ and Operando Techniques for Characterization of Interfaces and Interphases in Lithium-Ion Batteries* Towards Futuristic Energy Storage; Paving Its Way through Super-capacitors, Li-Ion Batteries and Beyond, Tours, France, January 23, 2020.
P. Novák	*Materials and Interphases in Li-Ion Batteries* Seminar at Norwegian University of Science and Technology, Trondheim, Norway, February 7, 2020.
P. Novák	*Batterien: Wunschdenken und Realität* Invited presentation for the Parliament of the Canton Zug, Zug, Switzerland, September 15, 2020.

P. Novák	*Energiespeicherung in Batterien: Realität vs. Wunschdenken* Invited presentation at Physikalische Gesellschaft Zürich, Zurich, Switzerland, Virtual, November 5, 2020.
P. Novák	*Back to the Surface: 35 Years of Operando Characterization of Battery Electrodes* IBA 2020 – International Battery Association Meeting, Virtual, November 18, 2020.
T.J. Schmidt	*Stromspeicher – der Schlüssel zur Energiewende?* ETH Science City, October 25, 2020.
S. Trabesinger	*The Quest for High Energy Density Batteries* Presentation at Speicher-Roundtable, Forum Energiespeicher Schweiz, AEE Suisse, Virtual, October 28, 2020.
S. Trabesinger	*Morphological Peculiarities from Lithium Plating and Stripping* INESS-2020: The 8th International Conference on Nanomaterials and Advanced Energy Storage Systems, Virtual, August 6, 2020.
S. Trabesinger	*The Quest for High Energy Density Batteries: Research Highlights* Presentation at VR&S-Anlass, Virtual, October 29, 2020.

Contributed Talks

F. N. Büchi, T. Schuler, T.J. Schmidt	*Interface Engineering: On the Impact of Micro-Porous Layers on Polymer Electrolyte Water Electrolysis Performance* PRiME 2020: 238th Meeting of The Electrochemical Society (ECS), Online, October 4–9, 2020.
Ch. Csoklich, T.J. Schmidt, F.N. Büchi	*From Gas Diffusion to Gas Distribution Layers* PRiME 2020: 238th Meeting of The Electrochemical Society (ECS), Online, October 4–9, 2020.
S. De Angelis, F. Marone, F.N. Büchi	*Observing oxygen transport in polymer electrolyte water electrolysis via Operando X-ray tomography* PRiME 2020: 238th Meeting of The Electrochemical Society (ECS), Online, October 4–9, 2020.

S. Garbe, T.J. Schmidt, L. Gubler	*Elevated Temperature Proton Exchange Membrane Water Electrolysis for Reduced Cost of Green Hydrogen Production* PRiME 2020: 238th Meeting of The Electrochemical Society (ECS), Online, October 4–9, 2020.
A. Muroyama, A. Pătru, B. Pribyl-Kranewitter, L. Gubler	*Electrochemical Pumping Behavior of an Anion Exchange Membrane CO2 Separation Device* PRiME 2020: 238th Meeting of The Electrochemical Society (ECS), Online, October 4–9, 2020.
T. Schuler, F.N. Büchi, T.J. Schmidt	*Towards a GEneric Understanding of the Oxygen Evolution Reaction – Experimental Determination of REaction Mechanism and Kinetic PArameters* PRiME 2020: 238th Meeting of The Electrochemical Society (ECS), Online, October 4–9, 2020.
A. Schuller, J. Eller	*Finite Element Model Based Determination of Local Membrane Conductivity of Polymer Electrolyte Fuel Cells* PRiME 2020: 238th Meeting of The Electrochemical Society (ECS), Online, October 4–9, 2020.

Patents

L. Gubler, A. Schneider, J. Duburg, E. Zanzola	*Procedure for the lamination of a polymer electrolyte film onto a porous support layer for energy storage devices* EP20215599, filed December 18, 2020.
T. Schuler, F. Büchi	*Porous Transport Layer Based on Multiple Micro and Nano Sintered Porous Layers* PCT/EP2020/050805, filed August 18, 2020.
A. Stefancic, M. El Kazzi	*A Method for Preparing a Surface Fluorinated Battery Materials* 2020P17253EP, filed August 14, 2020.

Posters

E. Winter, T.J. Schmidt, S. Trabesinger	*Pitfalls in Lithium Metal Battery Characterisation* ISE-2020: 71st Annual Meeting of the International Society of Electrochemistry, Virtual, August 31–September 1, 2020.

Conferences Organizations

F.N. Büchi, C. Gloor	*36th Swiss Electrochemistry Symposium:* *Seasonal Energy Storage: The Role of Electrochemistry* Virtual, Paul Scherrer Institut, Villigen, Switzerland, September 16, 2020.
T.J. Schmidt, F.N. Büchi, (Co-Organizers)	*Symposium Polymer Electrolyte Fuel Cells and Electrolyzers 20 (PEFC&E 20)* Virtual, 238th ECS Fall Meeting PRIME, October 4–9, 2020.

Editorial Work

C.A.F. Vaz, A. Kleibert, M. El Kazzi	*Nanoscale XPEEM Spectromicroscopy* In: K.D. Sattler (Ed.), 21st century nanoscience – a handbook. Advanced analytic methods and instrumentation (Volume 3). Taylor & Francis, Boca Raton, USA (2020). DOI: 10.1201/9780429340420-17
T.J. Schmidt	*Handbuch Energiespeicher (2020)* DOI: 10.3929/ethz-b-000441253.

K. Swider-Lyons,
H. Uchida, P.N. Pintauro,
W. Mustain, F. Buechi,
B.S. Pivovar, C.A. Rice,
J.M. Fenton, P. Strasser,
K.E. Ayers, A.Z. Weber,
R.A. Mantz, H. Xu, S.
Mitsushima, E. Kjeang,
T.J. Schmidt,
B. Lakshmanan,
A. Kusoglu,
H. Jia, D.J. Jones,
D.H. Ha, S.K. Kim
(Editors)

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