

Data Driven Acceleration of Materials Discovery Through Integrated Correlative Spectroscopy, Synthesis, and Experimentation

相関分光法, 合成, および実験の統合によるデータ駆動型材料探索の加速化

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An acceleration of the transition towards renewable energy production and storage necessitates new breakthrough materials. Discovering such new and improved materials does, however, call for new paradigms that incorporate combinatorial materials science and high-throughput experimentation to fast-track materials scaling from discovery to product. Herein a brief example of materials science acceleration through integrating complex workflows is explored as implemented in the platform for accelerated electrochemical energy storage research (PLACES/R) located at the Helmholtz Institute Ulm. With the synthesis, characterization, and performance evaluation techniques, we seek to unravel the physicochemical relationships between composition, structure, processing, and performance to discover and upscale future materials for short- and long-term energy storage.

再生可能エネルギーの生産と貯蔵への移行を加速するためには、新しい画期的な材料が必要である。しかし、このような新しい材料や改良された材料を発見するためには、コンビナトリアル材料科学とハイスループット実験法を取り入れて、材料の発見から製品化までの段階を迅速に行うための新しいパラダイムが必要である。本稿では、Helmholtz Institute Ulm研究所(HIU)の電気化学エネルギー貯蔵研究加速プラットフォーム(PLACES/R)を用いた、複雑なワークフローの統合による材料科学加速の実施例を紹介する。合成、特性評価、性能評価技術を駆逐することで、組成、構造、加工、性能の物理化学的な関係を解明し、短期・長期のエネルギー貯蔵のための未来の材料を発見し、スケールアップすることを目指す。

Introduction

There is a critical need for new and improved materials for intermittent renewable energy storage^[1] in both stationary and mobile applications.^[2] Here it is however not just important to find the highest performing^[3] material for a given functional property,^[4] but it is also important to discover materials that are scalable. Scalable on the TWh scale, which is the energy storage need of countries and continents, means that a material may only be comprised of earth abundant constituents and made through low-temperature green-chemistry processes.^[2] Most of the currently available chemistries for Hydrogen production^[5] or for batteries^[6] do only fulfill this requirement in part leaving great opportunities for materials discovery and optimization.^[7]

The first wave of trying to accelerate materials science

was started in fact in the 1990s in Japan^[8] after some early experiments in the 1950s in Germany.^[9] Both times scientists tried to optimize expensive to manufacture materials like dental alloys^{[9],[10]} containing gold or silver. The method of choice back then, and still today, was the synthesis of thin films using various CVD or PVD techniques.^[11] In the 1990s there was a trend towards automation which is why there had been a trend for almost 30 years to deploy automation to materials science.^[12]

The advancements in the field of “combinatorial materials science” (CMS) offered bespoke combinatorial synthesis and high-throughput characterization (using high-throughput experimentation - HTE). Many of these had the idea of connecting these systems to data management but few implemented a thorough integration.^[13] In the late 2010s there was then the push for accelerated materials understanding in the so called “materials genome initia-

tive”.^{[14],[15]} There scientists sought to screen hundreds of thousands of materials using computational tools to then synthesize them using the methods available from CMS and HTE. The paradigm then was to perform so called inverse design such that one could query a functional property and receive from correlators and models a structure, composition, or processing route. Given however the vast breath and complexity of the chemical space it became evident that the descriptors and underlying physicochemical relationships were well beyond one- or two-dimensional descriptors and with the explosive rise of capabilities in data science the community shifted

towards autonomous materials science. In autonomous materials science there is the idea to build so called materials acceleration platforms (MAP)^{[12],[16]} in which research tasks such as synthesis, characterization, data-management etc. are not just accelerated through automation or use of databases but also integrated through data pipelines and robotic automation.^[6] Using the perspective of accelerated and integrated research tasks one can in fact measure the acceleration of materials science through the MAP paradigm. Early demonstrations have in fact shown accelerations^[4] of up to 20x but even greater accelerations should become available in the near future.

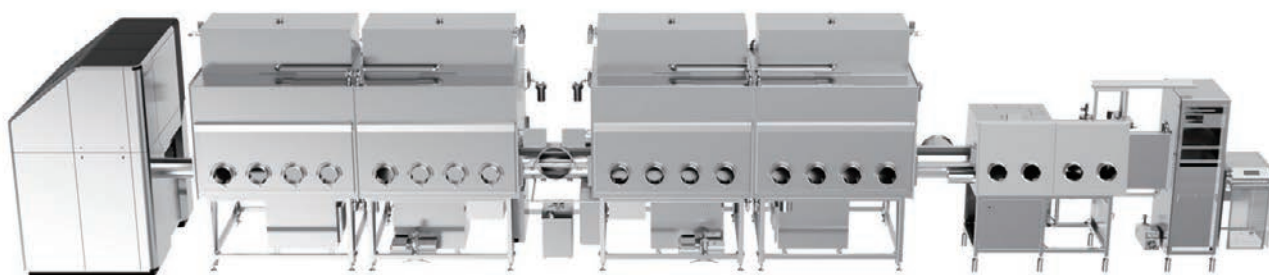


Figure 1 Rendering of the platform for accelerated electrochemical energy storage research (PLACES/R) that houses (from left to right) a near ambient pressure X-ray photoelectron spectrometer (NAP-XPS), high-throughput spectrometers and electrolyte formulation robots (left glove box), several scanning droplet cells and a coin cell assembly robot (right glovebox) as well as a sputtering system (right hand side).

Building a platform

In the following I will briefly describe an example set of measurements that highlight how the paradigm of a materials acceleration platform can be implemented. A platform is more than the sum of its parts as the overall added value is being created in the integration of its modules. Alone each setup and method can be accelerated but it is the integration that makes the difference over conventional high-throughput materials research.

Automation framework

The first and foremost important tool to build a platform that can acquire data on various of materials on different carriers using a multitude of techniques is the automation framework.^[7] Given the challenge of building a laboratory^[6] that utilizes instruments from a variety of vendors my current team, my coworkers from JCAP and I developed the hierarchical experimental laboratory automation and orchestration framework (HELAO). For this software we have written drivers for each and every device in our laboratory that expose them through a REST API - essentially a web server to which commands can be send. The driver functions are relatively low level i.e. turning a

motor by 12 revolutions at 5 revolutions/second. More complex features are grouped into something we call action servers i.e. moving a probe by 5 mm in x-direction and 2 mm in y-direction. A sequence of actions or sequence of events makes up an experiment. A list of experiments makes up a campaign or session. Because the communication with actions and drivers can be achieved through small JSON files (essentially nested dictionaries containing instructions) an experimental session becomes programmable and interchangeable^[17] between instruments (devices make up instruments).

Data management

Having all data acquisition and reporting done through code also enables fine grained data management that also entails all details of how data was acquired and processed. Any downstream processing,^{[18],[19]} if automated, can also be integrated and be used to expand the capabilities of autonomous workflows. Currently we have drivers for stages, robots, I/O boards, potentiostats and spectrometers including even NMR. We can thus build complex workflows.

Instruments

Second are the instruments themselves. In electrochemical energy storage research there are a lot of pertinent techniques, we do however currently focus on Raman and FTIR spectroscopy in a self-build integrated setup called HITS (High-throughput spectrometer), four scanning droplet cells to perform millimeter scale high-throughput half-cell battery research (MISCHBARES), a setup for Autonomous Synthesis and Analysis of Battery electrolytes (ASAB), an automatic battery assembly robot (AutoBASS),^[20] and various auxiliary and supporting techniques like high-throughput XRF and near ambient pressure XPS.

Examples

Due to the modularity of our platform,^[6] there is not a single common or even “standard” workflow but several emblematic “workflow paths” through which insights can be generated. The earliest example in our laboratory was in fact the corrosion of copper and subsequent analysis using Raman and FTIR as offered through our in-house build high-throughput spectrometer (HITS) that has two Raman probes (green and red lasers) and a FTIR probe from OEM manufacturers.^[21] A great benefit of high-throughput Raman spectroscopy is, that it is a relatively fast hyperspectral mapping technique that cannot just be used to fingerprint a crystal structure, but it also bears the possibility to pinpoint the location of a measurement area

with great accuracy. Figure 2 shows an emblematic mapping of a materials library where the false color spots demarcate areas at which electrochemical experiments were performed. Further correlation between the Raman signals and the electrochemical protocol employed can unravel corrosion behavior. Here, we used a simple copper foil, but the possibilities are virtually unlimited in terms of materials composition through combinatorial synthesis techniques such as reactive magnetron sputtering or coating of powders.

Coating of powders is however a tedious process that requires manual finetuning of process parameters. We have however recently employed screen printing of electrodes to manufacture materials libraries for use in the scanning droplet cell (SDC). These libraries offer the benefit of being rapidly deposited and then analyzed by various subsequent methods such as X-ray fluorescence (XRF), SDC and HITS prior and post to SDC. In Figure 3 a XRF intensity map over such a materials library containing 121 Lithium iron phosphate, a common cathode material in batteries, measurement areas are shown. These measurement areas can now be analyzed using all the available techniques in our lab and be also used in full cell setups using larger area coatings (different screen-printing mask) in the AutoBASS^[20] setup. The AutoBASS setup is shown in Figure 4. Here, a single battery can be assembled by a robot in under 3 minutes.

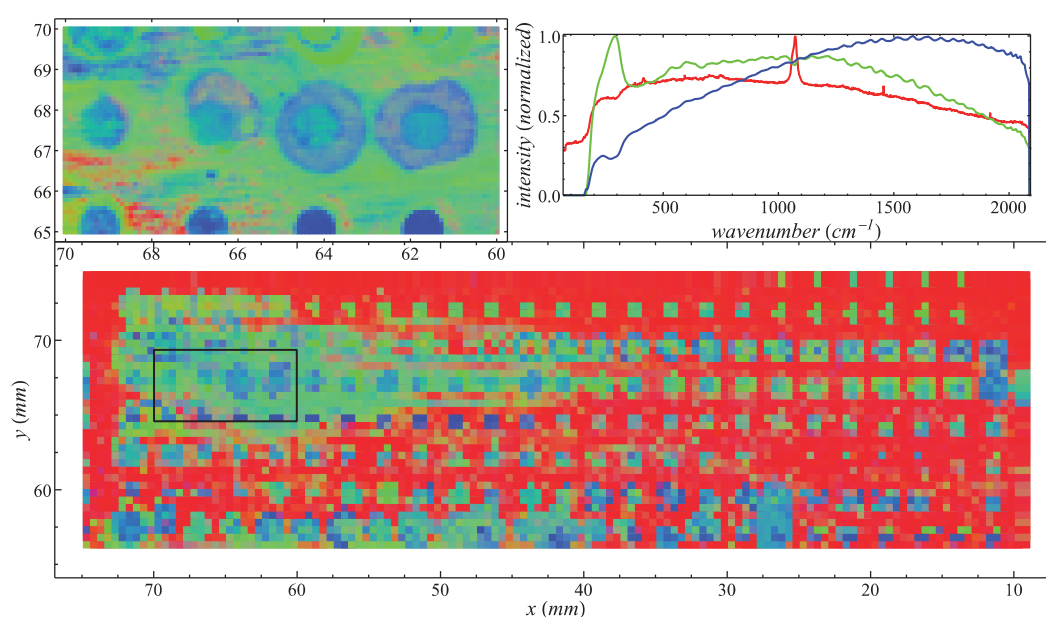


Figure 2 False color mappings using Raman spectroscopy and non-negative matrix factorization to create maps of three different phases corresponding to a background of metallic copper, and two corrosion phases. The top left image shows a zoomed in region, the top right shows the deconvoluted base spectra and the bottom part of the figure shows a large area overview of the materials library. The black box marks the area in which a high-resolution mapping was performed. The corrosion products were produced using scanning droplet cell electrochemistry.

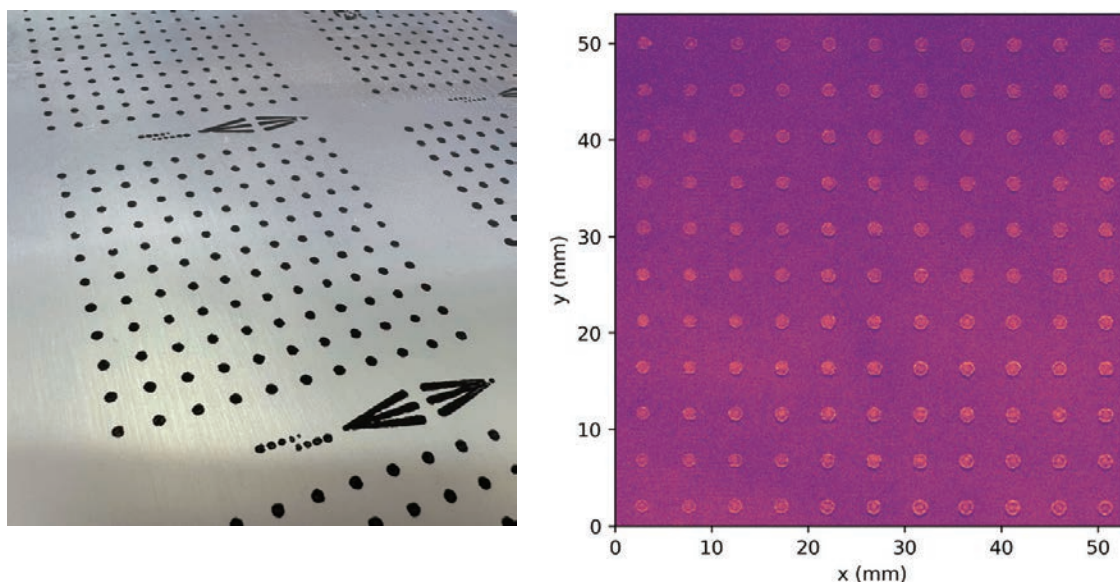


Figure 3 Left: Photograph of a screen printed materials library of Lithium iron phosphate (LFP) after drying in ambient air. Right: X-ray fluorescence mapping of a materials library coated through screen printing after drying. Brighter spots mark the areas where LFP was deposited through the screen printing mesh. The slight gradient from the top left to the bottom right is intended as the mask contained different effective mesh sizes to vary LFP loading over the materials library. The XRF measurements were performed using a HORIBA XGT-9000 with a polycapillary probe. Using the scanning droplet cell in a similar way as shown in Figure 2 it is possible to assess real battery materials without measurement area crosstalk.

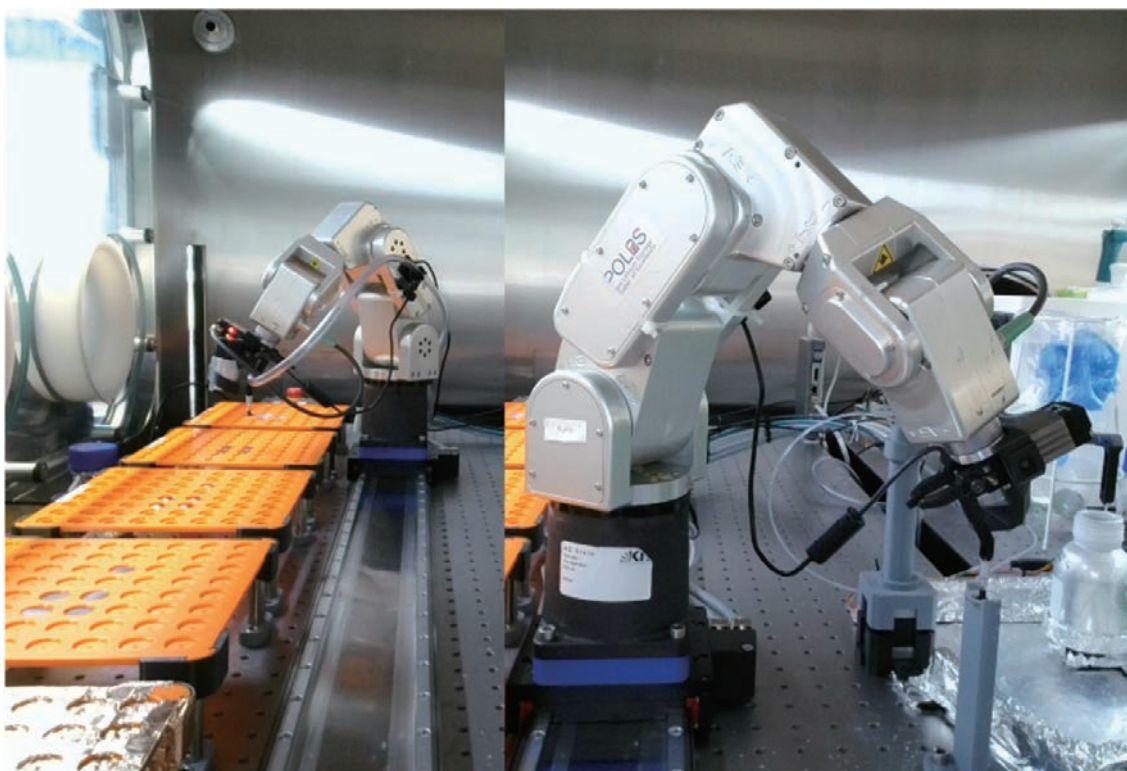


Figure 4 Photograph of the pick and placing actions of AutoBASS, a robot for the automatic assembly of coin cell batteries. On the left the robot is shown picking up cell components that are placed onto an assembly post on the right. On the assembly post additional steps such as electrolyte filling are performed. The entire process is housed inside a nitrogen filled glovebox to avoid moisture and oxygen impurities.

Conclusion

Using additional tools such as ASAB and the XPS we can thus cover the entire materials research value chain and integrate insights from all steps of battery research and even catalysis. Being able to acquire data from bulk, interface, and interphase processes in combination with system and long-term effects opens up new and interesting research directions. Through fine grained data management and automation control we also have the unique opportunity to deploy A.I. guidance of our robotic systems. With novel orchestration paradigms and software such as FINALES we can even integrate external data sources and measurement requests to build workflows across laboratories and modalities to create the lab of the future.

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