Key Requirements for Advancing Machine Learning Approaches in Single Entity Electrochemistry

Viacheslav SHKIRSKIY, Frédéric KANOUFI*

Université Paris Cité, ITODYS, CNRS, Paris, F-75013 France

**viacheslav.shkirskiy@cnrs.fr*

Abstract

Despite the noteworthy progress in Single Entity Electrochemistry (SEE) in the last decade, the field still must undergo further advancements to attain the requisite maturity for facilitating and propelling machine learning (ML)-based discoveries. This mini-review presents an analysis of the required developments in the domain, using the success of AlphaFold in biology as a benchmark for future progress. The first essential requirement is the creation and support of high-quality, centralized, and open-access databases on the electrochemical properties of single entities. This should be facilitated through the automation and standardization of experiments, promoting high-throughput output and facilitating comparison between datasets. Finally, the creation of a new type of interdisciplinary specialist, trained to pinpoint critical issues in SEE and implement solutions from applied informatics, is vital for ML approaches to flourish in the SEE field.

Keywords:, Electrochemistry, Single Entity, Machine Learning

Highlights

- Field of Single Entity Electrochemistry needs further development to enhance discoveries driven by Machine Learning
- High-quality and publicly accessible, extensive databases need to be established
- Automation and standardization of experiments should facilitate the creation of databases
- Cross-disciplinary collaborations need to be promoted

Introduction

In the past decade, few emerging technologies have garnered as much attention as those within the realm of machine learning (ML), owing to its groundbreaking successes in a wide array of applications. These include, but are not limited to, the development of large language models (e.g., ChatGPT[1]), the mastery of complex games such as Go and chess (exemplified by AlphaGo[2] and AlphaZero[3]), the advancement of autonomous driving systems,[4] to name a few. In spite of these advancements, a similar level of success remains relatively rare in natural sciences, with only a few notable examples such as AlphaFold[5] and the more recent GNoME[6] models. These models are capable of predicting protein and inorganic crystal structures with near-experimental accuracy. In this mini review, we critically analyze the reasons hindering breakthroughs in other fields, with a particular focus on single entity electrochemistry (SEE), reflecting the authors' primary research interest.

SEE is an emerging field centered on the electrochemical characterization of individual heterogeneities, or entities, within electrochemically active bulk materials.[7–9] These entities range from nanoparticles to individual grains, grain boundaries, and even single molecules or single enzymes. Unlike macroscale electrochemical measurements that average responses across a bulk material, SEE reveals each entity's unique contributions, proving indispensable in applications like electrocatalysis, corrosion prevention, and sensor development.[7–10] SEE has seen significant advancements in the past decade, driven by developments in nanoscale scanning methods such as scanning electrochemical cell microscopy (SECCM),[10,11] scanning ion conductance microscopy (SICM),[10,12] and various optical techniques,[13,14] overcoming the challenges of nanoscale measurement. Some experts foresee the next pivotal development in SEE being facilitated by recent ML advancements yet SEE must evolve further for this prediction to materialize.

Additionally, we note that ML has already been applied in adjacent areas of electrochemistry,[15] with reviews documenting its recent achievements and potential future applications in fields like voltammetry,[16,17] batteries,[18] electrocatalysis,[19] and corrosion.[20] Differing from this, our paper concentrates on the most examples within the SEE field and identifies the critical elements required for ML to excel, comparing these with the significant accomplishments of models like AlphaFold.

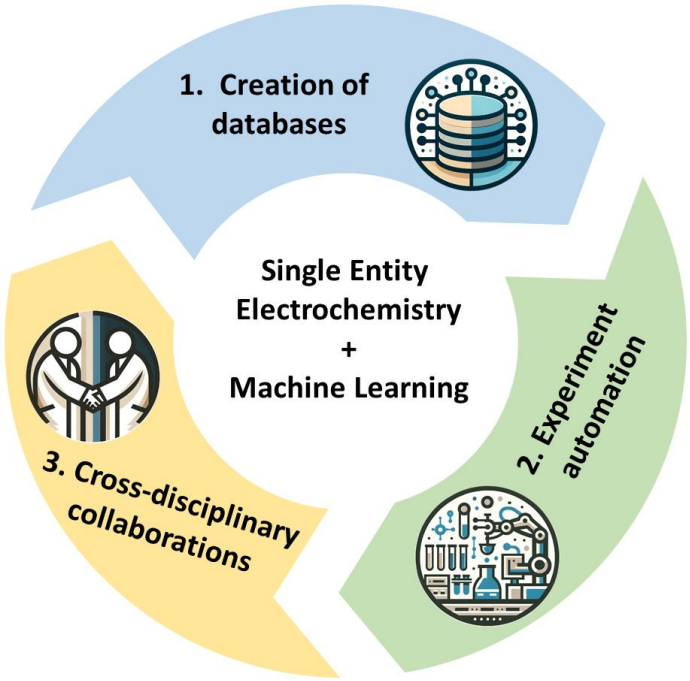

Figure 1. Schematic illustration of the essential requirements for the success of machine learning in the field of single entity electrochemistry

Creation of high-quality and publicly accessible databases

In essence, AlphaFold represents a significant advancement in predicting protein structures with near-experimental accuracy, relying solely on amino acid sequences.[5] Unlike traditional methods that focus on the calculations of physical interactions of molecular forces, AlphaFold employs deep learning and a protein structure database to establish correlations. Key to this innovation is the high-quality empirical database, a product of collaborative efforts by institutions like Worldwide PDB, RCSB in the US, and Europe's EMBL-EBI,[21] who have collectively fostered an environment of open data sharing essential for developments such as AlphaFold. In line with this, an equivalent effort must be directed toward building a large, comprehensive, standardized database for attaining an ML-driven breakthrough in SEE (Fig. 1).

The initial instances of such databases have begun to emerge within the realms of electrochemistry, encompassing corrosion, batteries, and electrocatalysis. Notably, efforts have been made to establish open databases containing crucial metrics, such as the efficiency of corrosion inhibitors,[22] degradation indicators for passive metals[23] and efficiency of electrocatalysts in catalytic reactions[24,25] etc. To speed up the creation of such databases, automated methods have been developed to efficiently extract electrochemically relevant data from extensive literature initially intended for human readers.[26,27] For example, the ChemDataExtractor toolkit processed 229,061 papers, converting data from standardized tests on battery materials into a comprehensive, auto-generated database detailing capacity, conductivity, chemical composition and other key properties.[28]

Unfortunately, as far as we are aware, there is no existing database in the field of SEE. Moreover, tools like ChemDataExtractor cannot be directly employed to create such a database from the current literature, owing to a variety of employed experimental methodologies with non-standardized protocols.[10] Take, for example, nanopipette methodologies: SICM operates under full immersion of the electrode, typically measuring ionic currents.[10,12] On the other hand, its closest analogue, SECCM, exposes only a small fraction of the electrode during measurement and typically extracts conventional electrochemical data, such as cyclic voltammetry data, chronoamperometric results, etc. [10,11] Direct comparison of electrode efficiency from SICM and SECCM data is challenging due to the different quantities measured under entirely different experimental conditions.

Among all methods, SECCM has been gaining popularity as a standardized technique due to its straightforward interpretation, which align with conventional electrochemistry.[29] Creating standardized SECCM databases with, for example, geometry of the single entities, their crystallographic and chemical composition vs the electrochemical activities (expressed via Tafel slopes) could be the first step toward establishing a unified database for reactivity at the nanoscale.[30,31] Then, steps should be taken to establish experimental configurations that enable reliable cross-correlation between SECCM and other methods, such as emerging optical techniques.[13,14,32–35] This approach is crucial for the development of a comprehensive database, especially in scenarios where SECCM is less effective, like with hydrophilic interfaces.[36,37]

Last but not least, a database of this nature should encompass not only positive results but also negative findings, which are indispensable for the effective training of any ML model.[38,39] Furthermore, the database must undergo thorough examination to identify redundancy and biases.[40,41] In the current landscape, only a fraction of positive results manages to find its way into final publications in SEE and other fields. Regrettably, original data sharing remains infrequent. To rectify this situation, proactive measures need to be implemented to promote open science practices and facilitate the widespread dissemination of negative results. Standardized protocols should be established to enhance machine readability and data accessibility.

Experiment automation and standardization

Experiment automation is increasingly recognized as a vital approach for generating databases in natural science that can be adopted in SEE (Fig. 1).[42,43] Advancements in this field could be propelled by either constructing highly autonomous robotic systems or automating specific critical points in current workflows.[42–45] In the first scenario, the literature demonstrates a marked trend toward completely eliminating human involvement by constructing an autonomous robotic system.[45,46] These systems are designed to search for and analyze literature from online resources independently, plan experiments, execute them using a robotic setup, and process and analyze data using ML. A prominent instance is the Coscientist platform, which integrates ChatGPT-4 for optimizing palladium-catalyzed cross-couplings.[46]

We believe that such full automation is currently premature in the field of SEE due to instrumental limitations and the limited independent reasoning capabilities of large language models like ChatGPT-4. However, SEE could still benefit from partial robotization, which would enhance high-throughput capabilities and aid in standardizing experimental procedures.

Among the possible robotization platforms, scanning droplet cells are a notable example,[47,48] similar to SECCM in their strategy of using a droplet to contain the electrochemical cell for measurements. The foremost difference is the decreased spatial resolution, typically ranging in the millimeter scale, in stark contrast to the nanoscale focus of SECCM. These droplet cells find significant application in electrocatalysis, where they facilitate the scanning of various solid electrocatalysts with diverse chemical compositions.[49,50] These electrocatalysts are typically prepared using a custom-programmed pipetting robot[49,50] or, alternatively, through a physical vapor deposition process.[51] Scanning droplet setups are also employed in corrosion research.[52] For instance, an advanced millifluidic platform has been developed to autonomously mix solutions and conduct corrosion-related tests using a droplet cell on Zn-Ni alloys.[53] Additionally, microarrays of droplets prove effective in autonomous screening the efficacy of corrosion inhibitors.[54] Collectively, these instances highlight the potential for partial automation that could be translated to SECCM approach.

Another example illustrating the trend toward partial automation in the SEE field is seen in targeted electrochemical measurements at the nanoscale.[34,55–58] Here, a SECCM probe can be externally guided to probe predefined locations of interest, often determined from coupled optical microscopy imaging, instead of raster scanning the entire interface.[34,58] Alternatively, it can be programmed to follow specific locations for targeted patterning in nanoscale additive manufacturing.[55–57] The most recent example comes from the field of battery research, where SECCM was coupled with in-situ optical microscopy for targeted analysis of Li^+ (de)intercalation dynamics in TiO_2 nanoparticles (Fig. 2). This approach enabled high-throughput electrochemical sensing and has the potential to find wide applications in the field. [34]

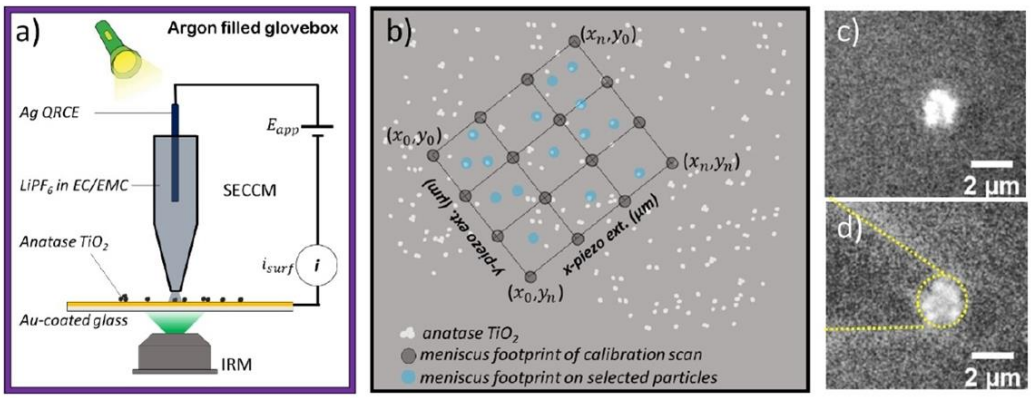

Figure 2. Example of a targeted SECCM analysis coupled with optical microscopy employed or single-entity Li-ion (de)intercalation measurements of TiO_2, adapted from [34]*. (a) Schematic of the setup. b) Smart scanning protocol to target selected TiO_2 nanoparticles for automated SECCM measurements at a series of specific position coordinates (not drawn to scale). c) Typical in situ optical image of a single TiO_2 nanoparticle cluster before SECCM tip approach, and d) after meniscus contact from the tip. The yellow dashed lines in (d) highlight the position of the pipet and meniscus.*

Stand-alone optical microscopy can also significantly benefit from automation in current workflows, particularly through image processing of high-throughput, data-rich, wide-field high-resolution images.[59–61] In our recent study, we proposed using object detection algorithms, enhanced with unsupervised machine learning, to automatically cluster reactivity patterns on Al alloys (Fig. 3).[62] The objective of this method is to accelerate data analysis while minimizing subjective human influence. In cases like this, such analysis becomes indispensable, as the raw, high-resolution, wide-field videos can exceed 100 gigabytes, making manual processing impractical.[60] Our research was inspired by advanced image processing methods prevalent in biology and bioinformatics,[59,60,63] highlighting a significant opportunity for development in the SEE field.

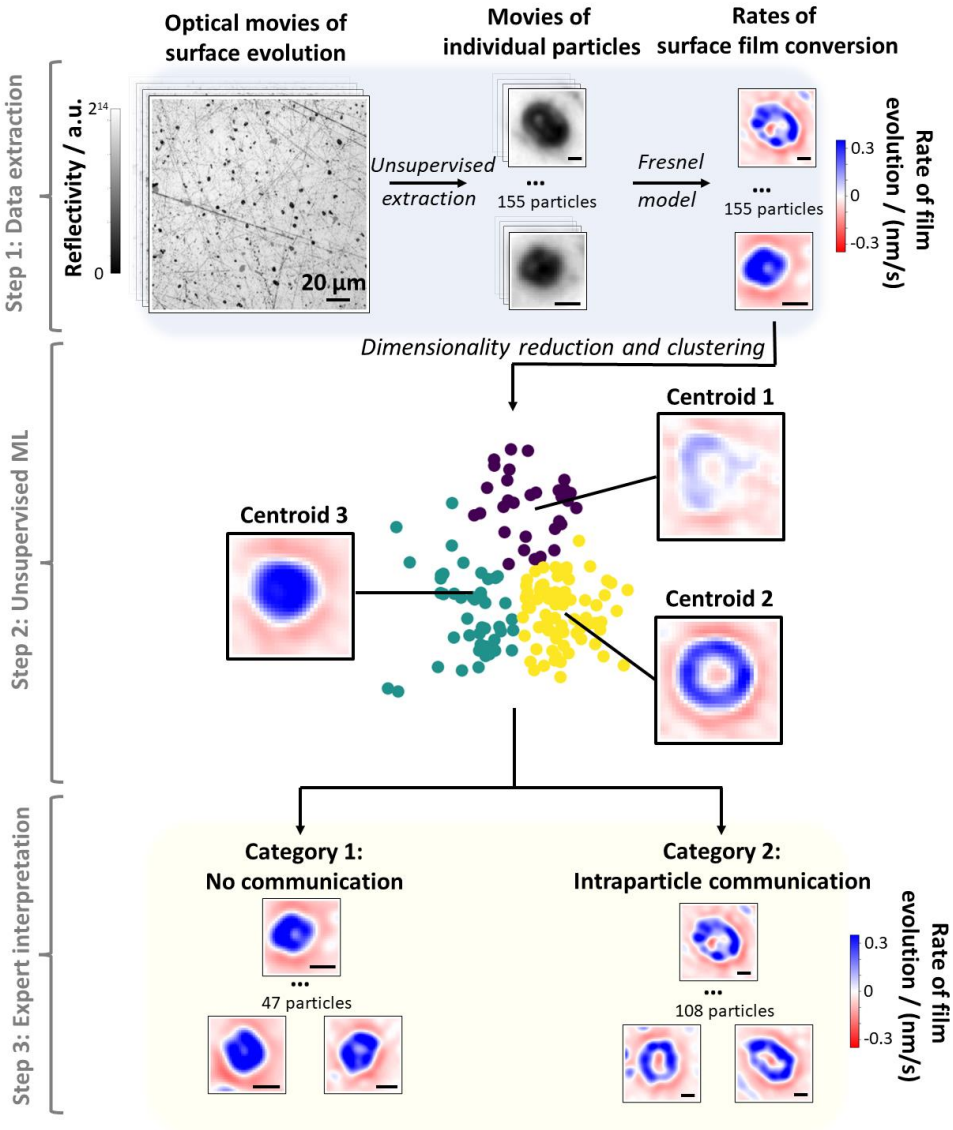

Figure 3. Example of an automated pipeline based on unsupervised ML for reactivity pattern recognition from optical reflectivity images of Al6061, adapted from [62]. *Step 1: The positions of all individual particles and converted into maps of rates of film evolution. Step 2: 2D projections of all maps of rates colored according to the results of clustering. The centroid image of each cluster is represented in the figure, and it was used to categorize the particle into a specific category in step 3. The scale bars of individual particles are 1 µm.*

Cross-disciplinary collaborations

In the final part of this review, it is crucial to step back and gain a broader perspective, recognizing the significance of the interdisciplinary field of bioinformatics. The very existence of this field that produces specialists trained to pinpoint critical issues in biology and implement solutions from applied informatics, has been vital in developing models like AlphaFold. Unfortunately, this is not yet the case in the field of SEE and electrochemistry at large. Traditionally rooted in chemistry, electrochemistry primarily draws students, PhD candidates, postdocs, and scientists with a non-mathematical background. In contrast, ML-

driven discoveries necessitate individuals with robust mathematical foundations, or at minimum, collaboration with interdisciplinary scientific teams. For SEE to thrive, it is essential to promote a diverse student body with robust mathematical skills, possibly through the creation of a separate interdisciplinary field at the intersection of electrochemistry, instrumentation, and applied mathematics.[64] Such innovations would foster an environment beneficial to ML-driven discoveries in SEE.

Conclusions

Our analysis, inspired by the notable success of the AlphaFold model in biology, outlines the essential areas of development for SEE to effectively support future ML-driven discoveries. The first and most important aspect is the creation of high-quality, open-access databases, where the structural properties of individual entities are correlated with their electrochemical properties. Importantly, such databases should include both negative and positive results, and they should be well-supported and regularly checked for redundancy and biases. Automating experiments at every level, including the development of autonomous laboratory robots for nanoscale electrochemical tests and the partial automation of bottleneck stages in current data acquisition processes, will aid in database creation. Ultimately, the development of a new breed of interdisciplinary specialists, adept at identifying key challenges in SEE and applying informatics solutions, is crucial for the successful integration and advancement of ML techniques within the SEE field.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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The paper discusses the requisite skills and expertise for nanoelectrochemists to navigate future challenges and opportunities, particularly in leveraging data-driven ML for discoveries