

Crossing the Valley of Death: From Fundamental to Applied Research in Electrolysis

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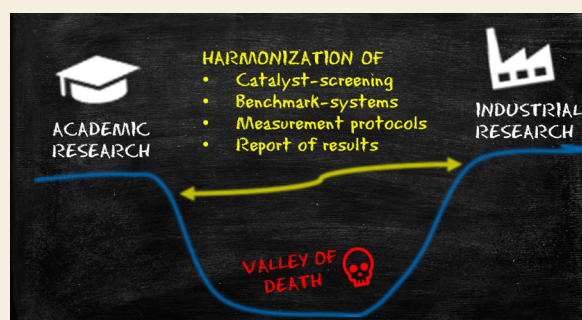
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ABSTRACT: The growing societal and political focus on the use of environmentally friendly technologies has led to an ever-increasing interest in electrolysis technologies in the scientific communities. This development is reflected by the plethora of candidate catalysts for the hydrogen and oxygen evolution reactions, as well as the CO₂ reduction reaction, reported in the literature. However, almost none of them entered the stage of application yet. Likewise, the reports on process engineering inadequately address the utilization of these catalysts, as well as electrode and cell concepts, that might be suitable for the market. Evidently, a closer collaboration between chemists and engineers from industry and academia is desirable to speed up the development of these disruptive technologies. Herein, we elucidate the critical parameters and highlight the necessary aspects to accelerate the development of industrially relevant catalysts capable of fulfilling the forthcoming challenges related to energy conversion and storage. The aim of this Perspective, composed by industrial and academic partners, is to critically question current undertakings and to encourage researchers to strike interdisciplinary research pathways.

KEYWORDS: Electrolysis, Electrocatalyst, Electrodes, Electrolyzers, Hydrogen, Carbon Dioxide



INTRODUCTION

Since 195 countries and the European Union signed the Paris Agreement in 2015 within the United Nations Framework Convention on Climate Change, a race toward a greener future and with it the substitution of fossil energy started.¹ Moreover, this endeavor is increasingly becoming a matter of public concern, e.g., resulting in the Fridays for Future activities since 2018 actively demanding a faster pace in the utilization of sustainable technologies.² While this demand to perform an immediate change is certainly an economic and societal hazardous undertaking since the technological foundations are not fully established, it should nonetheless be an incentive and inspiration for all researchers in the field of sustainable chemistry and engineering. Herein, especially electrochemical processes are expected to play a major part in limiting the temperature increase to 1.5 °C and reduce CO₂ emissions until 2030.

Among the various techniques, especially the electrochemical hydrogen production, driven by renewable energy sources, is considered to play an important role as a pacemaker to strive climate protection.³ Notably, 31 countries from around the world (representing 44% of global GDP) support national hydrogen projects.⁴ Additional 20 countries (representing another 44% of global GDP) already adopted (or are about to adopt) a national hydrogen strategy. For example, the

German hydrogen strategy defines the reduction of emissions, the diversification of energy supply, the support of national technology development, the integration of renewable energies as well as setting proper legal frameworks as key features. Hydrogen herein serves as the central molecule to achieve the climate goal by establishing a “hydrogen society” in 2050.⁵

In the light of these current developments, the progress of disruptive electrolysis technologies becomes a key undertaking and requires joint efforts from all over the world and various disciplines. From numerous discussions with colleagues in industry and academia as well as our own research undertakings, we believe that there is a large, underexplored gap between basic and application driven research to commercialize new technologies. This gap mainly originates from a discrepancy of parameters studied in basic research and those required under industrial relevant conditions as well as a missing communication between the various stakeholders.

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In this Perspective, we thus highlight important key parameters that we believe should be considered in current renewable energy research fields. We herein emphasize the requests in the three predominant related research fields, namely, (a) catalyst development, (b) electrode design, and (c) engineering of electrolysis cells. While we put special focus on water electrolysis (especially the hydrogen evolution reaction (HER)), the presented thoughts are by no means restricted to this area alone but apply equally to related applications involving electrolytic transformations such as CO₂ reduction (CO₂R). While it might seem surprising at first, the presented concepts can also be useful for galvanic transformations in, e.g., fuel cells, since the underlying processes concerning charge and mass transfer, and durability of the system—to name a few—are fundamentally related.

■ ELECTROCATALYST DEVELOPMENT

The quest for novel electrocatalysts or the advancement of existing materials is considered as one of the most crucial points in the development of electrolyzers. It is still anticipated that the capital and probably more importantly the operational expenditure of an electrolyzer can be very significantly improved by advances in catalyst development. Consequently, many research groups around the globe with variable technical/chemical backgrounds are engaged in this important field.^{6–11}

In view of many newly reported high-performance materials with their sophisticated compositions, structures, and morphologies, it may be surprising that, despite these enormous efforts, comparatively old and simple catalysts still dominate the technical/industrial applications. In addition to the many inadequately developed patenting strategies that are of high importance for commercialization but are often neglected from academics, this phenomenon can be attributed to an insufficient translation of promising materials from basic research to an applicable electrode and utilization under realistic conditions. Certainly, it is difficult to identify proper candidates within the enormous range of potential catalysts and processing options from which to choose. However, the situation is even more complicated by fundamental problems in investigating new materials and also in communicating scientific results.¹² In many cases, it almost seems as if scientific interests and technical-economic interests are presented at cross-purposes. Consequently, many potential catalyst candidates are not further investigated because initial investigations focused on essentially inadequate parameters or incorrect conclusions are drawn from electrocatalytic experiments. Nevertheless, we are convinced that a deeper dialogue between industry and basic research can lead to the uncovering of synergies that can take electrochemical catalyst development to a new and unprecedented level by pinpointing the decisive parameters to be evaluated. In the following, we list important selected issues that, in our opinion, require a more pronounced discussion in the community or a fundamental critical examination to enable the development of standard protocols that at the same time will facilitate the scientific communication.

Application Oriented vs Basic Research

In any project involving catalyst development, it should be a necessity to clearly define whether a potential technical application is really being considered for a given catalyst candidate or whether basic research topics, e.g., mechanistic

investigations, are the focus of interest. In particular, the following points should be considered when aiming for a game changer and next level catalyst for industrial processes:

(1) How complex or expensive would it be to produce the catalyst system under consideration? A catalyst with only a slightly better performance that has to be prepared in a large number of steps and/or at significantly higher cost due to extreme conditions (high pressure, high temperature, etc.) is certainly interesting in basic research but unsuitable for technical applications. In this regard, ecological and safety aspects must also be taken into account. The results of these considerations should thus be critically discussed in every publication. Likewise, considerations should be given to whether the catalyst is composed of rare components or its preparation is fraught with serious environmental concerns, a point that will increase in importance in the near future. It is conceivable that this aspect might even (partially) compensate for high cost in catalyst preparation.

(2) Another important fact to consider is that it should be plausible to prepare the catalyst in sufficient quantities to meet the demands required by the industry; bearing in mind that the electrolysis technology is anticipated to grow significantly over the coming years. Therefore, the scalability of the preparative procedure needs to be considered right from the outset and aimed ultimately toward the kilogram-tonne scale. Likewise, the various processes should be prioritized to those that can be operated continuously, such as gas phase reactions, milling etc. Another constraint is that scaled-up processes often yield catalysts with essentially different material properties. Even in the case of well-controlled processes, dispersity (the distribution of particle size, shape, surface, structure, and composition) will inevitably change during scale up.¹³ This is because temperature profiles and residence time distributions will become less defined. Hence, materials with demanding specifications and low resilience against batch-to-batch variation are certainly still interesting from a purely academic perspective but should not be regarded as “game changers” per se.

(3) In addition, durability is a major obstacle for the commercialization of catalysts: An industrially significant catalyst should have the longest possible shelf life and operational lifetime. It is understandable that this in particular is not always achieved in initial trials and is certainly hard to perform for multiple catalysts in screening experiments. But appropriate investigations should be carried out involving, e.g., postmortem analysis of catalysts to determine potentially detrimental effects and identification of effective improvements. Additionally, sufficiently large quantities of any new catalyst should be prepared which are made subject to more intensive investigation, so that a subsequent test of also aged samples is possible at least for those materials which are found promising in the initial tests.

Choosing the Right Testing Environment

An additional problem with high practical importance for the implementation of an electrocatalytic system is the construction of a proper model reactor emulating conditions of a larger scale system. Currently, many studies involving electrocatalysts for HER/(oxygen evolution reaction) OER or CO₂R are carried out utilizing rotating disc electrodes or other small-scale systems.¹⁴ While these systems can provide valuable data for the comparison of intrinsic properties and should be used to evaluate the basic properties, they are, however, very often

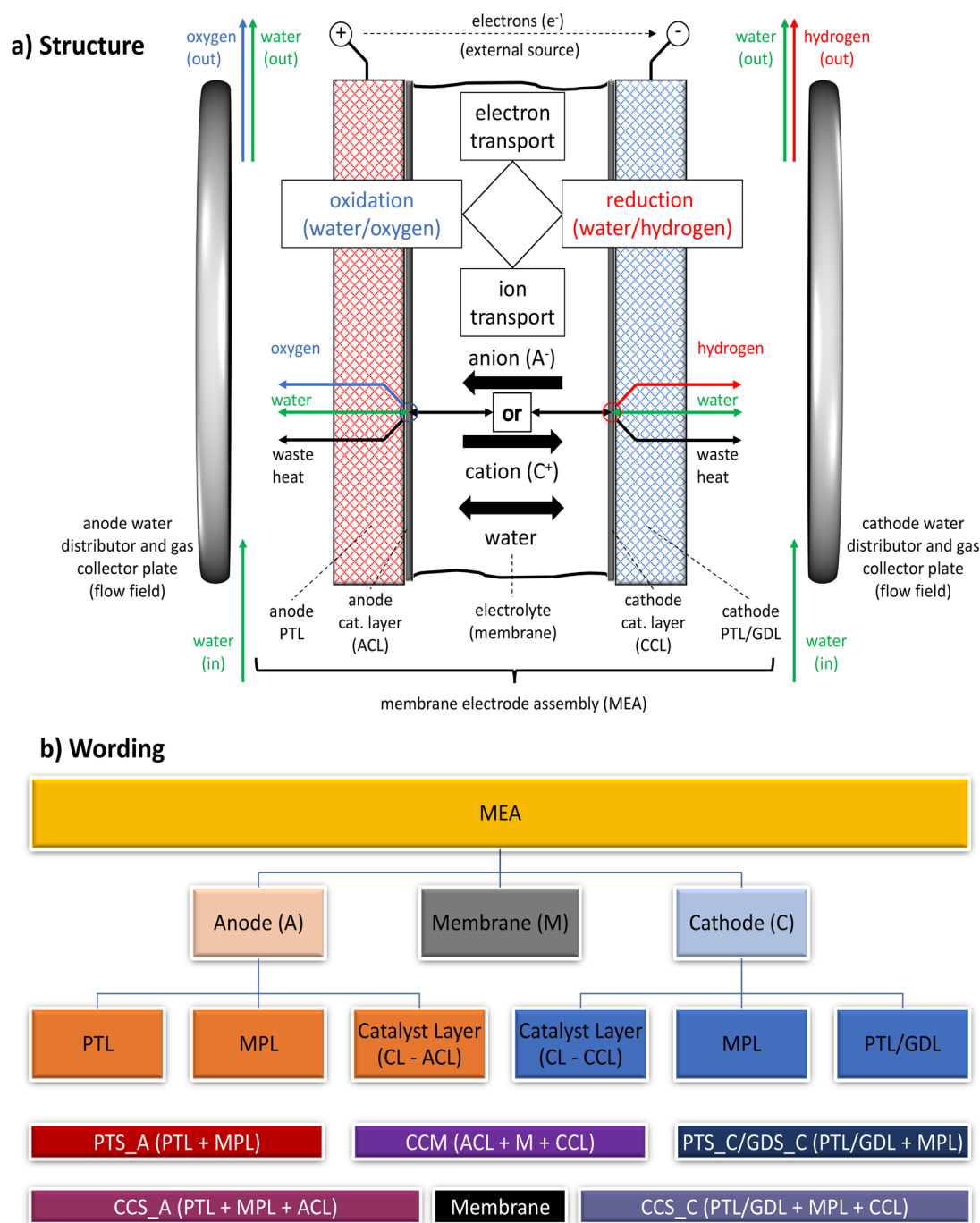


Figure 1. Setup of a MEA: (a) general structure; (b) wording. The abbreviations are as follows: PTL, porous transport layer; MPL, microporous transport layer; GDL, gas diffusion layer; PTS, porous transport system; GDS, gas diffusion system; CCS, catalyst coated substrate; CCM, catalyst coated membrane. While commonly PTLs are applied for electrolyzers utilizing liquids (e.g., PEM and alkaline cells), GDLs can typically be found when, e.g., dealing with the electrochemical reduction of CO₂.

unsuitable to judge any potential applicability and, therefore, are inappropriate. The reader should be aware that, in an industrial electrolyzer, commonly a catalyst will only be utilized in conjunction with a proper support layer/substrate and not as a “pure material”. Thus, a careful distinction between material properties and electrode properties, which is often neglected in the literature, and their interplay is of special importance for analyzing catalysts.¹⁵ Even tiny changes in the catalyst/electrode composition might lead to severe alterations of the observed properties. Strictly speaking, material properties are only encountered in cases where pure materials (e.g.,

pure metals) are employed as electrodes without any additives to improve the conductivity or any kind of binder material or ionomer (e.g., Nafion). It is thus very important to also define evaluation criteria and accordingly catalyst testing procedures which help to predict possible obstacles regarding the integration into an electrode layer. Unfortunately, very little systematic work has been done here so far.

In other words, the performance of a catalyst is tightly connected to the environment in which it is applied.^{16–18} It is to be expected that, e.g., a catalyst drop-casted on a glassy carbon electrode (which is often done in benchmark tests) will

show a very different behavior when spray-coated on a porous substrate or onto a membrane to be employed in membrane electrode assemblies (MEAs).^{19,20} With a commercial application in mind, it is therefore useful to regard the catalyst/electrode in a holistic fashion. Therefore, the ideal test world to judge catalyst performances would demand a simplified standardized model reactor for the major electrolyzer techniques such as polymer electrolyte membrane (PEM) electrolysis and anion exchange membrane (AEM) electrolysis or for the CO₂R with rather small electrode areas but a proper fabrication of electrodes (i.e., down-scaling). In that direction, such a noteworthy setup was recently presented for fuel cells²¹ as well as proton exchange membrane water electrolyzer catalysts.²²

However, there is still a clear need for an intensive discussion in the community to establish meaningful benchmark systems that allow both comparability of novel catalysts as well as extrapolation to large-scale systems.^{12,23} Then and only then, current performance criteria would become meaningful and a measure for the catalyst being a new benchmark system or not. Moreover, these statements do not refer to numerous measurements to elucidate trends or underlying reaction mechanisms. It is our firm belief that this information is likewise of utmost importance and journals as well as referees should avoid using performance numbers as a rejection or acceptance reason for these fundamental studies.

What are the performance data that a catalyst should then be tested for?

(1) *Current densities.* Many of the tested catalytic materials are currently benchmarked at rather low current densities (often between 10 and 100 mA cm⁻²). These data sets are not meaningful for any commercial processes and are obtained outside of the common process conditions that are applied.²⁴ It would be particularly desirable to at least test such a system at higher current densities of >500 mA cm⁻² (depending on the applications in question) to establish the systems' performance as well as to identify potential degradative factors.

(2) *The electrochemically active surface area.* Since current densities are frequently reported utilizing geometric areas for normalization, it should be mentioned that these values reflect electrode properties and not a material property.¹⁵ Thus, the fundamental material properties reported should be taken with severe caution as they can be easily altered by various environmental factors (see section on [Electrode Assembly](#)). Herein it is advisable to also report the mass activity of a potential new catalytic system.

(3) *Long-term experiments.* The long-term stability of an electrocatalyst and consequently of the electrolyzer is a crucial financial factor for evaluating the prospects of a novel material. So far, long-term experiments > 24 h at relevant current densities are not that frequently employed in catalyst studies. Given the fact, that an industrial setup has to remain operational for several years, degradation effects of even as low as 0.1% over 24 h are not negligible, and materials thus cannot be claimed stable. Hence, either long-time experiments have to be conducted or new rapid-aging tests have to be developed.

(4) *Selectivity/Faradaic yields.* An often-neglected parameter is the Faradaic efficiency, namely, the part of electrons that is later found in the isolated product. While this parameter is commonly analyzed nowadays for CO₂R experiments, for hydrogen evolution as well as water oxidation experiments a thorough quantification by means of product analysis is often

missing. This is, however, important to judge the efficiency, potential reoxidations, formation of hazardous intermediates, and other losses that falsify the measurements.

(5) *Reproducibility.* While it seems trivial, it appears that commonly only the very best experiments are provided and no credits are given for open discussions on reproducibility issues. A distinct error analysis and report is often missing. Such data, however, is very important to judge the catalyst performance as it will give insights/hints into potential surface or electrode composition effects, catalyst modifications, and problems with the cell setup.

Last, but not least, as will be highlighted in the section concerning cell setup, it is elementary to collect the above-mentioned data at relevant process conditions. This also means that the commonly collected room temperature data can only be an initial lead toward catalyst evaluation but is very far away to make any claim on a next technical breakthrough since electrolysis cells operate at elevated temperatures (often between 60 and 80 °C).²⁵

■ ELECTRODE ASSEMBLY

As briefly mentioned in the previous part, there are several options and parameters for testing new materials. Comparatively cheap and ubiquitously available are classical three-electrode setups like rotating ring-disc electrodes (RRDEs) or rotating disc electrodes (RDEs). However, and this needs to be stated clearly and cannot be emphasized often enough, these can only be used for initial material characterization and are certainly not able to make meaningful statements about the performance of a material in a later application. For drawing relevant conclusions as to whether a new material will become a "game-changer" and make an impact on the current energy problem, they must be incorporated into industrially relevant electrode assemblies. For this purpose, e.g., MEAs are suitable environments for extensive testing, i.e., over time and at relevant conditions using standardized (or at least carefully documented) procedures. In the widest sense, a MEA is a highly complex multicomponent, multilayer arrangement. [Figure 1](#) summarizes all components of a typical MEA, including porous transport layers (PTLs) and catalyst layers (CLs) on both the anode and cathode side. Depending on the context (more frequently used for fuel cells), the PTLs may as well be described as gas diffusion layers (GDLs) and microporous layers (MPLs). Consequently, the gas diffusion system (GDS) corresponds to a combination of GDL and MPL. The PTLs in combination with the CL represent the gas-diffusion electrode (GDE) (e.g., utilized in the electrochemical reduction of CO₂²⁶), whereas the membrane coated with the anode and cathode CL forms the catalyst coated membrane (CCM).

From [Figure 1b](#), it becomes immediately clear that the fabrication of a MEA—here plotted in a general form with electron and ion transport between the anode (oxidation) and cathode (reduction) in the center and heat and mass flows of reactants that must be transported to and products that must be transported out of the respective CL—is far from being trivial. Even if we assume a perfect connection between the materials, no problems with degradation of the catalysts, and a long-term stable and ideal separator, heat and mass transfer issues during operation remain. To overcome them, we require (1) highly performant and long-term stable active materials (see previous section), (2) electrodes that provide optimum transport of the reactants toward the CL by the PTL and

efficient transport of products (for instance by tailored hydrophobicity to work against capillary pressure and avoid flooding of the electrode), and finally (3) a good dissipation of heat to avoid thermal hotspots.

Let us assume we intend to benchmark a new active material against a reference. Then, in an ideal world, the experiment would be designed in such a way that all factors except the CL of interest remain constant. However, different active materials may have different requirements regarding their supply with reactants or dissipation of heat. Thus, the ideal structure of the optimum MEA (characteristic pore sizes, catalyst loading, electrolyte/ionomer content, and in-plane and through-plane conductivity) that is able to show the full capabilities of a new catalyst is unknown and usually approached empirically. In our hypothetical (or rather practical real-world) example, it might be that an electrode with a structure and composition optimized for catalyst A is not suitable for catalyst B. Without rational optimization—notably requiring a definition of “ideal”—the full potential of a new material cannot be explored, nor can different materials then be compared with each other. Eventually, it might be the case that an outstanding new catalyst puts requirements on the electrode structure, e.g., to prevent degradation, that cannot be fulfilled. For such a candidate, from an engineering perspective, there is no way to cross the valley of death and become technically relevant. And finally, we cannot ignore the fact that adhesion between different layers depends on the materials themselves.²⁷ So also, the contacting (assessed via the important quantity of adhesion strength) between the membrane and CL as well as CL and PTL is crucial for the MEA performance and stability. Although these are odd features that at first glance have nothing to do with a material's catalytic performance, they often determine the overall reported unoptimized properties for catalysts (see previous section) and are decisive for bringing a new material to the market and into applications.

Besides the optimization of the electrode's structure and composition, the scalability of the electrode itself is again a “place to get lost in the valley of death” and must therefore be considered. This adds an additional layer of complexity to the performance and scalability of the catalyst itself (see previous section). This is because the generation of multiple layers on top of each other is—generally independent of the chosen process (coating, spinning, deposition, etc.)—a huge challenge. In particular, if we keep in mind that the whole history of sample treatment (dispersion history, time windows, exact preparation procedures regarding the sequence of adding ingredients during formulation, application process) matters.²⁸ If these aspects are not reported appropriately, datum points for electrochemical performance, cannot be reproduced across different laboratories and prevent the exploration of causes and actions in complex parameter spaces. At the moment, compared to reports on new electrocatalytic materials, already a clearly reduced number of reports on the level of MEA is found in the literature. Showcases where such data is systematically connected to structure, polarity and composition of PTL and CCM is even rarer and showcases where this is done quantitatively including statistics and repeats are virtually nonexistent. Such data, however, is needed to fill white spots with numerical modeling and come up with more predictive approaches for electrode design.

■ THE CELL SETUP

At this point it should be no surprise to the reader that likewise the cell setup has a severe effect on the overall performance of the electrode/catalyst. Research addressing improvements of cell components is almost exclusively presented at the level of (relatively small) test cells, which are used for in situ characterization of new components. Preparation of such components is nontrivial and can be relatively time-consuming as well as expensive. In addition, degradation of the components might have a lasting negative impact on the test hardware (test cell and/or test stand). Due to these (potentially severe) repercussions, components that are tested in situ tend to be at a higher level of chemical stability than the pure materials that are newly developed and that are in the earlier stages of research. Hence, results for components that are tested in situ are considered to carry a higher level of confidence in their performance and their usability in a realistic field application. Nonetheless, there are shortcomings in the way of reporting results, of uncritical characterization of results, and/or of linking results overly optimistic or even baselessly into the overall technoeconomic framework.

It should be emphasized that, at the moment, hardly any of the available components meet all necessary requirements envisaged for future industrial application. However, in order to make results look as appealing as possible, publications sometimes focus on single, very specific performance parameters, while parameters that are crucial for industrial usability are softened down or even completely ignored. In these cases, wide reaching claims about “game changing” achievements and developments should be considered as inappropriate. The authors are aware that one cause for this current glamorized style of presentation certainly results from the substantial pressure many scientific groups at universities as well as research institutes are facing to publish results with an impact factor as high as possible whose value can be easily communicated to the scientific community. From this point of view, a fundamental discussion about publication and funding policies would be necessary which is beyond the scope of this Perspective and certainly not restricted to electrocatalysis.²⁹ However, we believe that much is already gained if a publication is preceded by a serious and honest self-assessment.

Some of those negative examples are given in the following list:

(1) *Performance is key*—obviously. So, not seldom the good performance of a PEM cell (see Figure 2 for a schematic description of PEM cell components) is used to point out the importance of the results. Based on a current/potential curve, this even seems justified, so credit where credit is due. If the good performance, however, originates from a too thin membrane, then a most likely occurring increased gas crossover (too often not discussed in the paper) would render a technical applicability impossible.

(2) *Short-term experiments*, in principle, do not allow conclusions about long-term performance. For instance, when using completely inappropriate materials for testing the longevity of an electrolyzer (the usage of carbon based PTLs on the anode side of a PEM electrolysis cell would be a classic example), results from short-term experiments can look very encouraging; and for a proof-of-concept, these might be totally acceptable. But making any conclusions for the long-term performance based on these results might be highly misleading

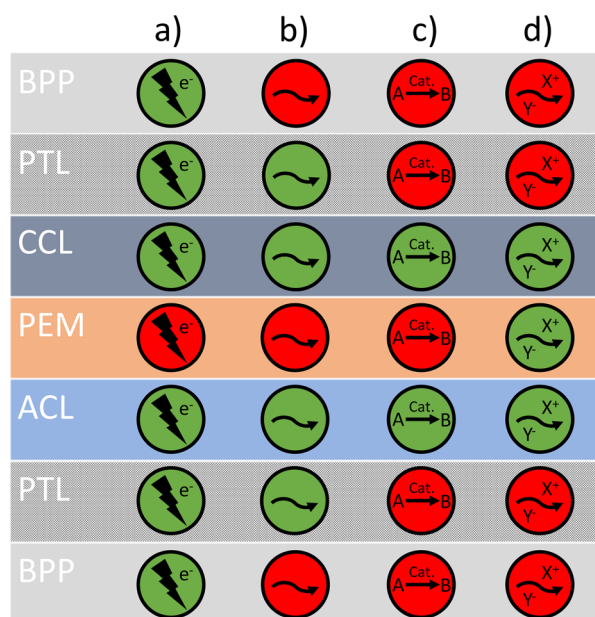


Figure 2. Structure of a PEM electrolyzer showing the different components and their functions: (a) electron conductivity; (b) diffusion/mass transport of water, hydrogen and oxygen; (c) catalytic capability; (d) ionic conductivity. The abbreviations are as follows: BPP, bipolar plate; CCL/ACL, cathodic/anodic catalyst layer.

though. And for a proof-of-concept, these might be totally acceptable results.

(3) *Long-term tests* are one of the best approaches for an in depth understanding of components but can still generate results that lead to too optimistic conclusions: To test the long-term stability of, e.g., a CCM, the amount of degradation can be reduced by expensive coatings and/or performance can be increased by catalyst loadings that are too high to be competitive in an industrial environment. While this setting is perfectly suitable to prove the stability of the CCM, the conclusion that the overall setup provides a breakthrough in the commercialization of electrolyzers would be misleading.

(4) Running tests at a *realistic temperature* of envisaged industrial operation is crucial for the significance of the obtained results for both the PEM and AEM or even CO₂R reaction. Too high temperatures can lead to an apparently good performance, especially, if the reference values have been measured at lower temperatures. However, the effect of temperature-induced degradation that becomes important in these cases is normally not monitored and the effect is normally not even discussed. Likewise, measurements at too low temperatures are not ideal. In this case, potential degradation that would occur at the temperature of operation is artificially slowed down and might remain undetected.

(5) High concentrations of KOH lead to good or very good power densities in AEM electrolysis. While the AEM has proven its merits and its potential, when being run with high KOH concentrations it is then competing in the same area as the very mature technology of alkaline electrolysis, a comparison that will be hard to win for the AEM.

(6) One of the big advantages of the AEM is the fact that it can potentially be run with nonplatinum group metals.³⁰ The use of precious metals as catalysts or electrodes in an AEM might be appropriate to demonstrate the stability of a membrane also at high current densities. But this should not be sold as a breakthrough in the commercialization.

In terms of cell setup and especially a holistic approach, it should furthermore always be kept in mind that an industrial electrolyzer of any sort is based on two half-reactions which by themselves contribute to the overall technical and economic performance of an electrolyzer.

■ GENERAL BENCHMARK CRITERIA

From the examples above it is obvious that components need to simultaneously fulfill a series of different criteria, if they should have a chance of making it into the industrial application. Based on the criteria defined within the strategic roadmap of Hydrogen Europe, none of the points summarized in Table 1 can be neglected when discussing the transferability of obtained results into the industrial application of PEM and AEM electrolysis:

Table 1. Important Key Performance Indicators (KPIs) for PEM Water Electrolyzers (SOTA: State of the Art)³¹

	SOTA	2024	2030
Industrial KPIs			
use of critical raw materials [mg/W]	2.7	1.25	0.3
degradation [%/1000 h]	0.19	0.15	0.12
electricity consumption [kWh/kg H ₂]	55	52	48
current densities [A/cm ²]	2.2	2.4	3.5
Academic KPI ^a			
temperature [°C]	60	70	80
cell voltage [V] ^b	1.9	1.8	1.75

^aBased on results obtained with SOTA materials and extrapolated performances. ^bAt temperatures and current densities given in the lines above.

Even with good intentions at heart, to provide and discuss all these values and information, the crucial question remains of how to generate those data with the resources available in an accurate and reproducible manner. The following list provides some ideas of why (while mandatory) reproducing results can be extremely challenging:

(1) *Synthesis of the candidate catalyst as a monophasic material.* Devising a preparative procedure that enables the candidate catalyst to be prepared without inclusion of undesirable phases can be a major challenge, the consequences of which are often overlooked.³² But an effective synthesis is a prerequisite to any meaningful characterization, since the presence of impurity phases can significantly affect the results and lead to a false evaluation of the catalyst's performance. Unfortunately, and as already mentioned above, this issue seldom becomes less problematic upon upscaling.

(2) *Catalyst coating.* With regard to the PEM and AEM, the coating of the candidate catalyst on membranes is a complex process designed to produce the required adhesion, dispersity, and homogeneity of the catalyst with a specified loading. However, since the details of these processes are proprietary information in the industry, it is unlikely that this aspect would be included in a publicly available protocol, emphasizing the need for an effective and trustful collaboration between academia and industry.

(3) *Preparation of each of the components before assembling the cell.* This concerns simple things as which solvents to use to cleanse the surfaces of the test cell components or more complex questions of how to prepare the MEA of a PEM cell by allowing it to swell in water for a certain temperature and time.

(4) *Sufficient accuracy of the measurement parameter.* This includes the precise spatial measurement of the temperature at which the experiment is run. This is a crucial parameter as it can differ between the inlet and outlet of the cell (which again will change depending on the current density applied), between the anode and cathode side, or where exactly the temperature sensor is placed in the system. Additional parameters that have to be properly defined are the internal gas pressure of the half-cells, the water flow rate in each of the half-cells and changes in the conductivity of the water (an increase in the conductivity hints toward degradation). Notably, some parameters are extremely hard to obtain from the actual cell assembly. The clamping pressure on the active area of the MEA is normally obtained approximately, e.g., by using pressure paper. But to then run a measurement without the pressure paper, that specific cell has to be disassembled and reassembled again. The same holds true for interfacial contact resistances. These values are obtained from as similar as possible cell assemblies but never taken from exactly the same cell. These parameters inherently carry an error, which is assumed to be small, but will still be nonzero.

(5) *Which procedure is used to obtain a “measured” value?* The signal for a measurement takes some time to converge to a steady value. How long does one wait to allow this to happen? How many values are measured in which time interval to be finally averaged into the one “measured” value reported?

(6) *Repeating measurements help to ensure accuracy.* The number of repeated measurements can be crucial for the quality of the recorded data. To record a polarization curve once from high to low and immediately afterward from low to high will only in a few cases provide very different results. Disassembling and reassembling the cell with the same materials or utilizing multiple batches of the same catalyst and processing them into electrodes is more time-consuming and more expensive but helps much more to identify the influence of an individual catalyst/electrode and cell assembly. The latter can help to identify errors during electrode layer processing and in the assembly process, the former most likely does not.

The overall objective for research aimed at industrial applications is to establish universally recognized characterization protocols with well-defined criteria that can be readily implemented by workers in the field. Notably, certain work has already been initiated in this area. For example, Bender et al.³³ have reported the results of a round robin test for PEM water electrolyzers conducted at five laboratories; Bertucciolo et al.³⁴ highlighted various recommendations for R&D priorities in water electrolysis, while Tsotridis and Pilenga³⁵ proposed a harmonized terminology for low-temperature water electrolysis for energy-storage applications.

However, these tests and setups are a drop in the bucket and more researchers need to follow their example. The aim of course has to be the development of robust and standardized or at least harmonized characterization protocols. This in turn seems, as experience has shown, to require at least a minimum amount of harmonized hardware (not only with respect to the test cell, but also regarding the test stand and used sensors) in order to obtain comparable results. So long as there is no standardized measurement protocol, we are left with a “best-practice approach”. For this, the choices made need to be clearly documented and critically checked, whether they are suitable for the job at hand. And even then, it is good practice to provide a reference measurement, run with known

(preferentially commercially available) materials/components and using the same settings and the same test hardware as used in the later measurements for the new materials/components.

CONCLUSION

As in all fields of science and technology, we need to be critical and realistic about our own experimental data, especially where such data are used to promote the exploitation of the material or process in question. We thus encourage researchers to venture beyond their immediate field and establish more intensive collaborations among the various disciplines to strive toward this common and urgent goal. The current limitations and hurdles are far from being trivial and can only be achieved by joint, interdisciplinary efforts from both academia and industry, and a research environment where failure is integral part of the culture, reported, and recognized. One group alone cannot perform the large number of experiments that are needed on various levels to really drive electrocatalysis toward this ultimate goal. Ideally, developments start hand-in-hand *qua* design, from day one. Our recommendations for future research in this field are therefore as follows: (1) Consider scale up already during synthesis. In case synthesis protocols cannot be changed and materials turn out to be true outperformers (on MEA level), new processes must be developed. (2) Consider dispersity already on the lab scale and application of scalable procedures (e.g., dispersion by scalable concepts instead of ultrasound, more efforts from the engineering side with regard to down-scaling). (3) Test new materials as soon as possible in GDEs, ideally even at the MEA level in downsized industrial relevant electrolysis cells. (4) Constantly check experimental repeatability as well as reproducibility across different laboratories. Finally, (5) the community should work on standardized experimental setups and protocols for catalyst/cell testing that go beyond the currently established (or rather not well established) standard (which is notably quite typical also in other fields of application, e.g., in the evaluation of photovoltaics as well as medicinal chemistry/drug developments). Therefore, suggestions and discussions within the community are clearly encouraged by the authors.

These points imply that all materials and methods down to the smallest aspect of a certain procedure are accurately published together with the results. Along this line, catalysts/electrodes/cells should be critically scrutinized with regard to their future potential with a critical and honest assessment.

In the authors' view, the serious challenges of the fundamental climate issue far outweigh a few additional impact points generated by euphemistic assessments. It needs to be emphasized that new materials must be processed into MEAs and tested appropriately with regard to electrochemical performance and stability as soon as possible. Putting all these aspects together, it becomes clear that proper electrocatalyst development is an interdisciplinary endeavor that requires chemists, material scientists, and engineers to work hand-in-hand along the whole design chain. This could be also integrated structurally into research institutions, for instance, by the generation of joint research centers or even research departments where (electro)chemistry, materials science, physics, chemical, and electrical and mechanical engineering are under one roof and scientists are working together in close connection to partners from industry.

In conclusion, a serial design of first developing new materials by chemists and then processing them by engineers is

not sufficient. We must act now together to make an impact and to enable the Paris Agreement to be a success.

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Notes

The authors declare no competing financial interest.

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REFERENCES

- (1) UNFCCC. *The Paris Agreement*. <https://unfccc.int/process-and-meetings/the-paris-agreement/the-paris-agreement> (accessed 2021-02-25).
- (2) Fisher, D. R. The Broader Importance of #FridaysForFuture. *Nat. Clim. Change* **2019**, *9* (6), 430–431.

- (3) van Renssen, S. The Hydrogen Solution? *Nat. Clim. Change* **2020**, *10* (9), 799–801.

- (4) Albrecht, U.; Bünger, U.; Raksha, T.; Michalski, J.; Zerhusen, J. INTERNATIONAL HYDROGEN STRATEGIES, A Study Commissioned by and in Cooperation with the World Energy Council Germany, 2020.

- (5) Federal Ministry for Economics Affairs and Energy. *The National Hydrogen Strategy*. <https://www.bmwi.de/Redaktion/EN/Publikationen/Energie/the-national-hydrogen-strategy.html> (accessed 2021-02-15).

- (6) Wächter, M.; González, L.; Dietzek, B.; Turchanin, A.; Roth, C. Functional Materials: Making the World Go Round. *Phys. Chem. Chem. Phys.* **2019**, *21* (18), 8988–8991.

- (7) Zhu, J.; Hu, L.; Zhao, P.; Lee, L. Y. S.; Wong, K.-Y. Recent Advances in Electrocatalytic Hydrogen Evolution Using Nanoparticles. *Chem. Rev.* **2020**, *120* (2), 851–918.

- (8) Amin, H. M. A.; Apfel, U.-P. Metal-Rich Chalcogenides as Sustainable Electrocatalysts for Oxygen Evolution and Reduction: State of the Art and Future Perspectives. *Eur. J. Inorg. Chem.* **2020**, *2020* (28), 2679–2690.

- (9) Boutin, E.; Merakeb, L.; Ma, B.; Boudy, B.; Wang, M.; Bonin, J.; Anxolabéhère-Mallart, E.; Robert, M. Molecular Catalysis of CO₂ Reduction: Recent Advances and Perspectives in Electrochemical and Light-Driven Processes with Selected Fe, Ni and Co Aza Macrocyclic and Polypyridine Complexes. *Chem. Soc. Rev.* **2020**, *49* (16), 5772–5809.

- (10) Nitopi, S.; Bertheussen, E.; Scott, S. B.; Liu, X.; Engstfeld, A. K.; Høch, S.; Seger, B.; Stephens, I. E. L.; Chan, K.; Hahn, C.; Nørskov, J. K.; Jaramillo, T. F.; Chorkendorff, I. Progress and Perspectives of Electrochemical CO₂ Reduction on Copper in Aqueous Electrolyte. *Chem. Rev.* **2019**, *119* (12), 7610–7672.

- (11) Zou, X.; Zhang, Y. Noble Metal-Free Hydrogen Evolution Catalysts for Water Splitting. *Chem. Soc. Rev.* **2015**, *44* (15), 5148–5180.

- (12) Masa, J.; Andronescu, C.; Schuhmann, W. Electrocatalysis as the Nexus for Sustainable Renewable Energy: The Gordian Knot of Activity, Stability, and Selectivity. *Angew. Chem., Int. Ed.* **2020**, *59* (36), 15298–15312.

- (13) Peukert, W.; Segets, D.; Pflug, L.; Leugering, G. Unified Design Strategies for Particulate Products. In *Advances in Chemical Engineering*; Marin, G. B., Li, J., Eds.; Mesoscale Modeling in Chemical Engineering Part I; Academic Press: London, 2015; Vol. 46, Ch. 1, pp 1–81.

- (14) Shao, M.; Chang, Q.; Dodelet, J.-P.; Chenitz, R. Recent Advances in Electrocatalysts for Oxygen Reduction Reaction. *Chem. Rev.* **2016**, *116* (6), 3594–3657.

- (15) Siegmund, D.; Blanc, N.; Smialkowski, M.; Tschulik, K.; Apfel, U. Metal-Rich Chalcogenides for Electrocatalytic Hydrogen Evolution: Activity of Electrodes and Bulk Materials. *ChemElectroChem* **2020**, *7* (7), 1514–1527.

- (16) Higgins, D.; Hahn, C.; Xiang, C.; Jaramillo, T. F.; Weber, A. Z. Gas-Diffusion Electrodes for Carbon Dioxide Reduction: A New Paradigm. *ACS Energy Lett.* **2019**, *4* (1), 317–324.

- (17) Yang, K.; Kas, R.; Smith, W. A.; Burdyny, T. Role of the Carbon-Based Gas Diffusion Layer on Flooding in a Gas Diffusion Electrode Cell for Electrochemical CO₂ Reduction. *ACS Energy Lett.* **2021**, *6* (1), 33–40.

- (18) Garcia de Arquer, F. P.; Dinh, C.-T.; Ozden, A.; Wicks, J.; McCallum, C.; Kirmani, A. R.; Nam, D.-H.; Gabardo, C.; Seifitokaldani, A.; Wang, X.; Li, Y. C.; Li, F.; Edwards, J.; Richter, L. J.; Thorpe, S. J.; Sinton, D.; Sargent, E. H. CO₂ Electrolysis to Multicarbon Products at Activities Greater than 1 A cm⁻². *Science* **2020**, *367* (6478), 661–666.

- (19) Konkena, B.; Junge, P.; Sinev, I.; Piontek, S.; Khavryuchenko, O.; Durholt, J. P.; Schmid, R.; Tuysuz, H.; Muhler, M.; Schuhmann, W.; Apfel, U.-P. Pentlandite Rocks as Sustainable and Stable Efficient Electrocatalysts for Hydrogen Generation. *Nat. Commun.* **2016**, *7*, 12269.

- (20) Tetzlaff, D.; Pellumbi, K.; Baier, D. M.; Hoof, L.; Shastry Barkur, H.; Smialkowski, M.; Amin, H. M. A.; Gratz, S.; Siegmund, D.; Borchardt, L.; Apfel, U.-P. Sustainable and Rapid Preparation of Nanosized Fe/Ni-Pentlandite Particles by Mechanochemistry. *Chem. Sci.* **2020**, *11* (47), 12835–12842.
- (21) Inaba, M.; Jensen, A. W.; Sievers, G. W.; Escudero-Escribano, M.; Zana, A.; Arenz, M. Benchmarking High Surface Area Electrocatalysts in a Gas Diffusion Electrode: Measurement of Oxygen Reduction Activities under Realistic Conditions. *Energy Environ. Sci.* **2018**, *11* (4), 988–994.
- (22) Schröder, J.; Mints, V. A.; Bornet, A.; Berner, E.; Fathi Tovini, M.; Quinson, J.; Wiberg, G. K. H.; Bizzotto, F.; El-Sayed, H. A.; Arenz, M. The Gas Diffusion Electrode Setup as Straightforward Testing Device for Proton Exchange Membrane Water Electrolyzer Catalysts. *JACS Au* **2021**, *1*, 247.
- (23) Kibria, M. G.; Edwards, J. P.; Gabardo, C. M.; Dinh, C.-T.; Seifitokaldani, A.; Sinton, D.; Sargent, E. H. Electrochemical CO₂ Reduction into Chemical Feedstocks: From Mechanistic Electrocatalysis Models to System Design. *Adv. Mater.* **2019**, *31* (31), 1807166.
- (24) Burdyny, T.; Smith, W. A. CO₂ Reduction on Gas-Diffusion Electrodes and Why Catalytic Performance Must Be Assessed at Commercially-Relevant Conditions. *Energy Environ. Sci.* **2019**, *12* (5), 1442–1453.
- (25) Schmidt, O.; Gambhir, A.; Staffell, I.; Hawkes, A.; Nelson, J.; Few, S. Future Cost and Performance of Water Electrolysis: An Expert Elicitation Study. *Int. J. Hydrogen Energy* **2017**, *42* (52), 30470–30492.
- (26) Junge Puring, K.; Siegmund, D.; Timm, J.; Mollenbruck, F.; Schemme, S.; Marschall, R.; Apfel, U.P. Electrochemical CO₂ Reduction: Tailoring Catalyst Layers in Gas Diffusion Electrodes. *Adv. Sustainable Syst.* **2020**, *5* (1), 2000088.
- (27) Leicht, H.; Orf, L.; Hesselbach, J.; Vudugula, H.; Kraus, E.; Baudrit, B.; Hochrein, T.; Bastian, M. Adhesive Bonding of 3D-Printed Plastic Components. *J. Adhes.* **2020**, *96* (1–4), 48–63.
- (28) Bapat, S.; Segets, D. Sedimentation Dynamics of Colloidal Formulations through Direct Visualization: Implications for Fuel Cell Catalyst Inks. *ACS Appl. Nano Mater.* **2020**, *3* (8), 7384–7391.
- (29) Abbott, S. Solubility, Similarity, and Compatibility: A General-Purpose Theory for the Formulator. *Curr. Opin. Colloid Interface Sci.* **2020**, *48*, 65–76.
- (30) Wang, L.; Weissbach, T.; Reissner, R.; Ansar, A.; Gago, A. S.; Holdcroft, S.; Friedrich, K. A. High Performance Anion Exchange Membrane Electrolysis Using Plasma-Sprayed, Non-Precious-Metal Electrodes. *ACS Appl. Energy Mater.* **2019**, *2* (11), 7903–7912.
- (31) EERA Key Performance Indicators (KPIs) for FCH Research and Innovation, 2020–2030. <https://hydrogeneurope.eu/news/eera-jp-fch-and-hydrogen-europe-research-publish-joint-effort-research-kpis-hydrogen> (accessed 2021-02-28).
- (32) Warner, T. E. *Synthesis, Properties and Mineralogy of Important Inorganic Materials*; John Wiley & Sons Ltd.: Chichester, 2011; pp 1–288.
- (33) Bender, G.; Carmo, M.; Smolinka, T.; Gago, A.; Danilovic, N.; Mueller, M.; Ganci, F.; Fallisch, A.; Lettenmeier, P.; Friedrich, K. A.; Ayers, K.; Pivovar, B.; Mergel, J.; Stolten, D. Initial Approaches in Benchmarking and Round Robin Testing for Proton Exchange Membrane Water Electrolyzers. *Int. J. Hydrogen Energy* **2019**, *44* (18), 9174–9187.
- (34) Bertucciolo, L.; Chan, A.; Hart, D.; Lehner, F.; Madden, B.; Standen, E. Development of Water Electrolysis in the European Union. Final Report. *Fuel Cells and Hydrogen Joint Undertaking*. <https://www.fch.europa.eu/node/783> (accessed 2021-02-25).
- (35) Tsotridis, G.; Pilenga, A. EU harmonised terminology for low temperature water electrolysis for energy storage applications. *EU Science Hub*. <https://ec.europa.eu/jrc/en/publication/eu-harmonised-terminology-low-temperature-water-electrolysis-energy-storage-applications> (accessed 2021-02-25).