3rd Symposium on Insights into Gas Diffusion Electrodes: From Fundamentals to Industrial Applications & Beyond the OER

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Plenary Lectures

Value-generating Anode Reactions in Unconventional Media

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Most of the work in electrolysis focusses on the generation of hydrogen on the cathode, typically under acidic or alkaline conditions. Anode reactions, predominantly the oxygen evolution reaction, are mostly only of interest, because they lead to high overpotential, and the target is reduction of the overpotential. However, anode reactions can be turned into an advantage, if the oxygen evolution reaction is replaced by a value-generating reaction. In future energy systems the required amounts of hydrogen are predicted to be so high that essentially no anode reaction would reach similar volumes. However, producing value at the anode could facilitate ramp-up of a partly hydrogen-based energy infrastructure by increasing overall revenue from the electrolysis process.

We have studied different, potentially interesting, anode reactions, partly in unconventional media and under rather demanding conditions in specially developed electrolysis cells [1]. These include the synthesis of methylbisulfate by methane oxidation in oleum. Under adapted reaction conditions, also the generation of methanesulfonic acid was possible [2]. Based on the experience with the electrolysis cell tolerating oleum as electrolyte at high pressures and elevated temperatures, a new cell was constructed allowing to work in liquid ammonia, also at high pressures. The goal of the work in liquid ammonia is related to the potential use of ammonia in future energy systems. One way of ammonia splitting is the electrochemical pathway, in which, analogous to water splitting, ammonia is split into hydrogen on the cathode and nitrogen on the anode. However, if instead of nitrogen oxidized nitrogen species can be formed, hydrogen evolution could be combined with the generation of potential fertilizer compounds, which are needed globally on very large scale. First success was achieved with the formation of nitrite and nitrate by activated oxygen species generated at the cathode, but this is on the expense of the hydrogen evolution [3]. However, with BDD as anode at least moderate amounts of nitrite and nitrate can be formed anodically, suggesting that the original idea is at least feasible [4]. Anodic formation of nitrite and nitrate had been shown in aqueous environment by the Schuhmann-group using a gas diffusion electrode [5].

In aqueous environments an interesting value-generating anode reaction is the oxidation of biomass-based 5-hydroxymethylfurfural (HMF) to furandicarboxylic acid (FDCA). FDCA could be an interesting monomer for the high-volume polymer polyethylene terephthalate. Under highly alkaline conditions, HMF is quickly converted to Cannizzaro-products which stabilizes the substrate against degradation to humins [6]. With a Fe/Co-modified nickel foam electrode, oxidation of HMF to FDCA is possible at industrially relevant current densities with excellent Faradaic efficiency (FE) and yield [7], and the reaction was successfully transferred to a flow cell configuration, producing syngas from CO_2 at the cathode. Protection of HMF via acetals is an alternative to the prevention of HMF degradation via the Cannizzaro reaction, and also via this pathway, excellent yields and FE could be achieved.

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Overcoming the Stability Challenge: Metal Based Gas Diffusion Electrodes for Long-term CO_2 Electroreduction

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Electrochemical reduction allows converting CO_2 into valuable products serving as fuels and feedstock chemicals for many industrial applications. The most well-known products from the electrochemical CO_2 reduction (ECR) include acids (formic acids), alcohols (such as methanol and ethanol), and hydrocarbons (such as ethylene and methane). Sn and Bi-based cathodes have extensively been reported for formic acid/formate (FA) production from ECR. This work presents the activities of VITO based on a unique way to prepare porous self-sustaining metal-based (Sn/ Bi) gas diffusion electrodes (GDEs) that are utilized in ECR with high faradaic efficiency (~80-90%) towards formate, which can be maintained for operational times of 2500 –4000 hours at a current density of 100 mA/cm^2 by utilizing the process and material stabilization techniques. Current efforts, potential approaches and inherent challenges related to the upscaling of CO_2 electrolyzers for industrial use will be discussed and illustrated.

The Use of Gas Diffusion Electrodes in Oxygen Reduction Reaction

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Oxygen reduction reaction is a vital reaction in fuel cells' cathodic compartment. This reaction is normally catalysed by Pt. In my talk I will present the design of alternative N-Fe-C catalysts as a replacement for Pt catalyst. I will present the synthetic design and electrochemical evaluation and most importantly the degradation of these catalysts studied using in operando ICP-MS coupled with gas diffusion electrodes. I will also present the influence of different commercial ionomers on the performance of such catalysts in addition to designing new hierarchical carbon materials to support transport in gas diffusion electrodes when loaded with active sites like Pt or Fe-N sites.

Keynote Lectures

Flooding Revisited: Electrolyte Management Ensures Robust Electrochemical CO_2 Reduction

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The electrochemical reduction of CO_2 is expected to play a role in closing the artificial carbon cycle, using a harmful greenhouse gas as feedstock for valuable chemicals. Building on recent achievements, high reaction rate and selectivity can be routinely achieved. Measurements at high current densities, however, brought several further scientific challenges to daylight, mostly regarding process stability. A key factor is maintaining ideal chemical conditions at the cathode, by ensuring the presence of cations and water at the catalyst surface, while minimizing the diffusion length of CO_2 in liquid phase. Here we demonstrate that the cation buildup at the cathode depends heavily on the applied conditions. There is an optimal cation concentration at the cathode, where CO_2 reduction occurs at the highest rate. Below this concentration, the activity of the catalyst decreases. At higher concentrations, the cathode support carbon paper becomes an active catalyst for the parasitic hydrogen evolution reaction, and at very high concentrations, the vigorously forming hydrogen blocks the path of CO_2 to the catalyst. These effects are reversible, and their extent can be quantified from EIS measurements. Our findings pave the way for long-term operation of CO_2 electrolyzers under continuously adjusted reaction conditions.

I will also discuss that flooding, one of the main performance fading mechanisms of CO_2 electrolysers, is vaguely defined, and often used for very different phenomena that cause cell/stack failure. The term itself is also controversial, as a fully wet electrode is often observed after high-performing zero-gap electrolyser cells are disassembled. To resolve this apparent contradiction, we investigated the cation balance in a zero-gap CO_2 electrolyser cell operated under different conditions, and also actively controlled cation concentration in the cathode compartment to study its effect on the electrolyser performance. We demonstrated that flooding in CO_2 electrolysers is not only related to the excess amount of water in the GDE, but rather to the presence of an electrolyte solution with high enough concentration. To operate the cell efficiently, the electrolyte concentration in the cathode GDE must be kept within an optimal range.

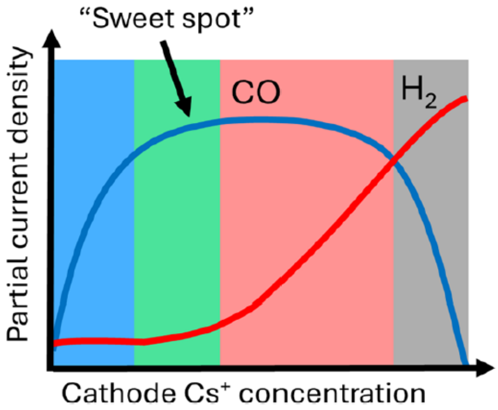

Figure 1. *Effect of the cation concentration in the cathode GDE on the CO_2 reduction performance.*

Electrooxidation of Alcohols as Alternative Anode Reaction in Alkaline Water Electrolysis

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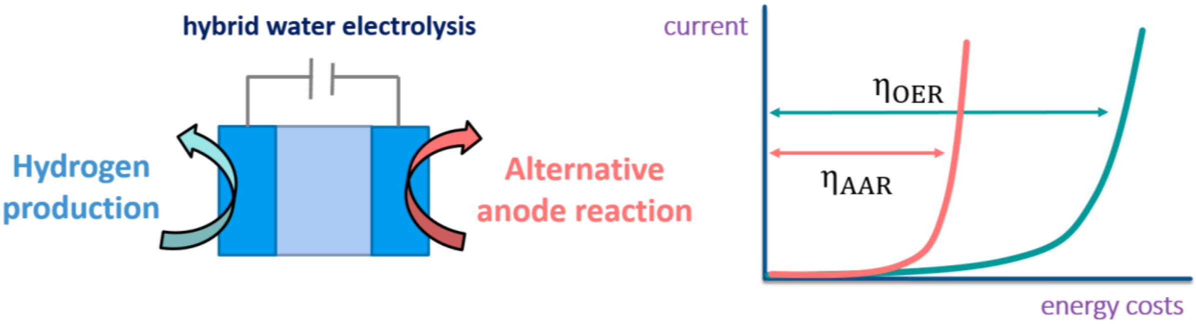
Compared to the conventional but sluggish oxygen evolution reaction (OER), alternative anode reactions (AAR) are highly advantageous in alkaline water electrolysis. On the one hand, hydrogen production can be achieved with lower energy costs, and on the other hand, it is possible to obtain value-added products at the anode from the electrooxidation of abundant compounds such as bio-based alcohols and sugars. This approach is known as hybrid water electrolysis. In this talk, I will illustrate how the energy costs for producing hydrogen can be reduced via the alcohol oxidation reaction (AOR) by comparing the performance of a series of $LaFe_{1-x}Co_xO_3$ perovskites towards the oxygen evolution, the oxidation of glycerol and the oxidation of isopropanol.[1] As a second example, the influence of electrode potential and electrolyte composition on the performance of Ni oxide nanoparticles towards the oxidation of glycerol will be discussed,[2] showing the trade-off between activity and selectivity of the AOR, and highlighting the need for identifying suitable catalysts and electrolysis conditions to minimize this trade-off.[3] Finally, the competition between the OER and the AOR will be discussed based on Differential Electrochemical Mass Spectrometry and in situ Raman spectroscopy analyses,[4] and, with this, future prospects for the emerging field of hybrid water electrolysis will be presented.



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Transferring Insights obtained on 2D Metal Foams Synthesized by Dynamic Hydrogen Bubble Templating to 3D Gas Diffusion Electrodes

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As we move towards clean and sustainable energy production, reducing greenhouse gas emissions is of paramount importance to our society. The electrochemical reduction of carbon dioxide (CO2RR) to valuable products can contribute to this goal, especially when using renewable energy sources such as wind and solar. However, CO_2 is a sluggish molecule and its reaction towards CO, formic acid or C_2 products such as ethene is plagued by very similar overpotentials and low selectivity. The electrocatalyst material plays a pivotal role in defining the product spectrum, i.e. on Ag mostly CO is being formed, whereas Cu can catalyze C-C bond formation leading to C_{2+} products.

Very often, studies reported in the literature focus on 2D model electrocatalysts in so-called H-cell set-ups. While these are essential to obtain fundamental insights, they do not perfectly mimic technically relevant conditions. For example, high current densities, high conversion rates and the efficiency of the reduction process itself are required to make the process industrially viable. All of these can only be achieved by using gas diffusion electrodes (GDE) in a flow cell configuration, coming at the expense of additional challenges, such as feed crossover, salt precipitation, and flooding.

In this work, we used the Dynamic Hydrogen Bubble Templating (DHBT) method, which does not require solvents and produces metallic foam structures in a very controlled fashion. By polarising a substrate foil sufficiently negatively, metal ions added to the electrolyte are reduced and electrochemically deposited, while at the same time the bubbles generated by the parasitic hydrogen evolution reaction (HER) act as a dynamically dissolving negative template around which the metal can grow, forming macroporous layers and nanoscale interconnecting foam walls. The obtained 2D model structures were investigated for their electrocatalytic performance in an H-cell with coupled GC, IGC and subsequent HPLC analysis. We found that it is possible to facilely transfer and maintain these Ag (and recently also Cu) 2D morphologies onto a 3D GDE, when replacing the smooth substrate foil by a metal-sputtered PET/PTFE mesh. These samples allow us to study the intricate structures in flow cells, which can be operated at industrially-relevant current densities.

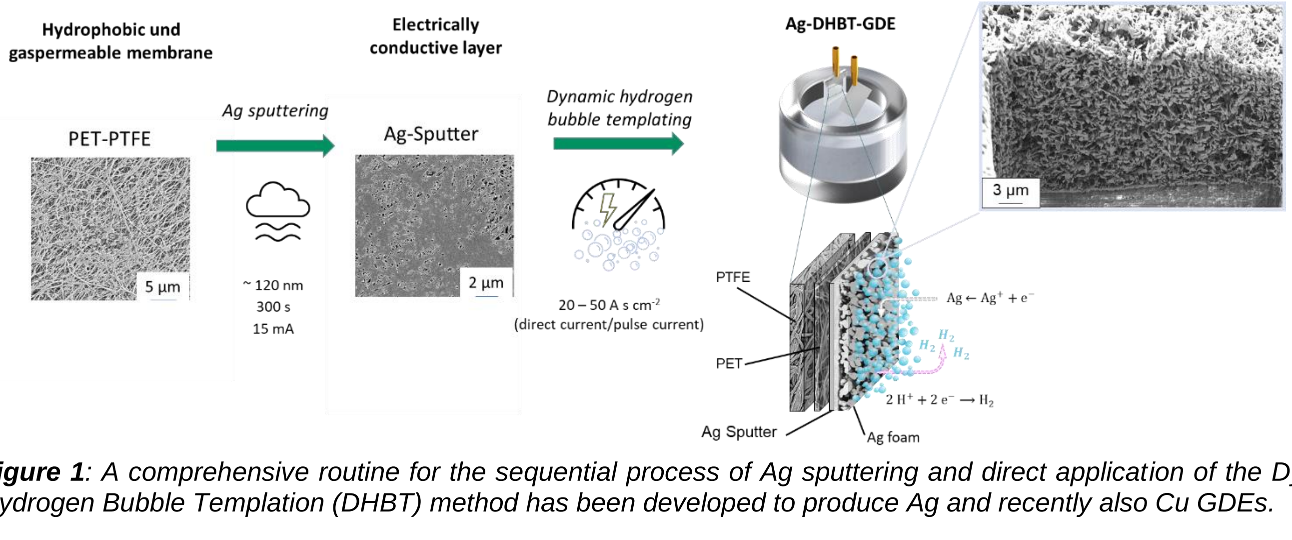

***Figure 1**: A comprehensive routine for the sequential process of Ag sputtering and direct application of the Dynamic Hydrogen Bubble Templation (DHBT) method has been developed to produce Ag and recently also Cu GDEs.*

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Invited Lectures

Electrolysis at 0.3 V: Electrochemical Liquid Organic Hydrogen Carriers

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The transition to zero-emission heavy transport demands compact, high-efficiency energy systems that outperform conventional hydrogen storage in cost, safety, and logistics. This talk presents the concept of an electrochemical liquid organic hydrogen carrier (LOHC) in which hydrogen is reversibly stored in a liquid-phase couple using low-voltage electrolysis. Thermodynamic analysis highlights the low cell potential required for liberate hydrogen, offering a compelling alternative to water electrolysis – essentially by replacing the OER we can unlock new methods of effective storing hydrogen. We explore the integration of this LOHC system with fuel cells, including approaches for electrochemical recharging and system-level efficiency. Results from both low- and high-temperature cell operation will be discussed, with emphasis on performance, durability, and materials compatibility. The concept, under development at Ternary, has advanced from lab-scale demonstration to system prototyping, driven by strong industry demand for scalable, zero-emission solutions. This electrochemical approach offers a promising pathway toward circular hydrogen storage without compression, liquefaction, or complex thermal management.

Electrolyte-Driven Selectivity: Tuning the CO_2 Reduction Interface Across Current Density and pH

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Acidic CO_2 electrolysis offers a promising method to convert CO_2 into valuable chemicals and fuels, providing advantages over neutral or alkaline systems, such as lower energy consumption and higher carbon utilization. Despite these benefits, the optimal electrolyte for acidic media remains unclear. Key challenges include ensuring catalyst stability and optimizing operating conditions, like current density and pH, to maximize efficiency and product yield. In this study, we explore CO_2 reduction on copper nanocubes supported on gas diffusion electrodes in seven different acidic electrolytes. We investigate the impact of supporting anions and their solution chemistry on reaction selectivity across current densities of 10–160 mA/cm². The catalyst's chemical state and evolution are monitored using operando X-ray Absorption Spectroscopy (XAS), while morphological changes are analyzed via Scanning Electron Microscopy (SEM). Our results show that selectivity correlates with the pK_a and diffusion coefficient of the acids, affecting local pH and determining optimal operating currents for each electrolyte. In contrast, morphological and chemical state changes seem to play a less significant role. Finally, we provide guidelines for selecting electrolytes in low-pH CO_2 electrolysis, which is crucial for advancing acidic CO_2 reduction technologies.

Electrochemical Hydrogen Pumps – The Impact of GDE Properties on Performance

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Electrochemical hydrogen pumps (EHPs) are a promising technology for separating H_2 from gas mixtures. This work implements novel proton-conducting binders into gas diffusion electrodes (GDEs) and investigates full-cell EHPs with a phosphoric acid-doped polybenzimidazole membrane[1]. The morphological GDE properties are investigated by scanning electron microscopy (SEM), energy dispersive X-ray spectroscopy (EDX), argon gas sorption, and distribution of relaxation times (DRT) analysis[2]. These methods reveal an extremely high catalyst layer porosity with phosphonated poly(pentafluorstyrene) (PWN70) ionomer. Adding Triton X-100 to the catalyst ink improves the distribution of poly(pentafluorostyrene) imidazole binder, increasing electrode porosity and cell performance. Furthermore, the hydrophobicity of all catalyst layers is probed by dynamic vapor sorption. The EHPs show 99.98% H_2 purity and 100% H_2 recovery from a reformate gas mix at 95% power efficiency at 200 °C. The durability test at 1.6 A cm^2 proves that the electrodes are stable. This clearly shows that the binders used for EHPs are suitable.

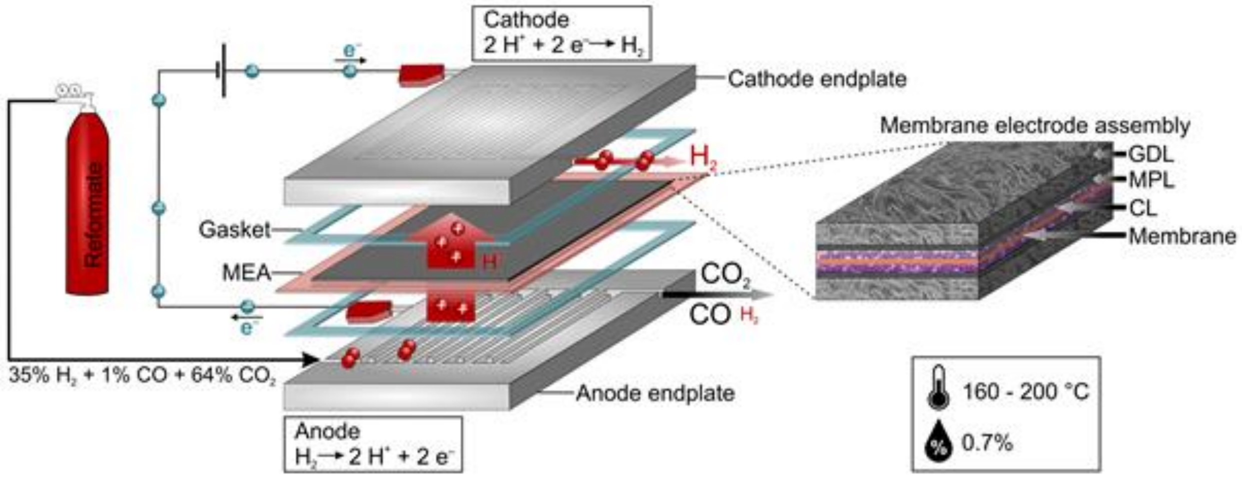

Figure 1: *Schematic illustration of the electrochemical hydrogen pump.*

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Oral Contributions

Understanding factors influencing selectivity in electrochemical conversion of CO_2 and CO using X-ray based operando methods

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Poor control over product selectivity is a central challenge in electrochemical conversion of CO_2 and CO to added-value products. Herein we report our efforts to understand and control several factors influencing selectivity in gas diffusion electrode CO_2 electrolyzers. We find that the electrolyte composition (e.g. cation type and concentration), cell configuration, and operational modes (e.g. pulsing) each significantly influence reaction selectivity. Methods such as *operando* X-ray absorption spectroscopy and X-ray radiography/tomography enable novel insights into catalyst structure and cell-level phenomena occuring under real operating conditions.

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Quantification of Gaseous Products Evolved During CO_2 Electroreduction at Industrially Relevant Current Densities Enabled by Gas Diffusion Electrodes

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Gas diffusion electrodes play an essential role in the conversion of gaseous reactants such as CO_2, in an aqueous-based electrolyte. The increased concentration of CO_2 that allows its conversion at high current densities enables the synthesis of essential chemicals, one of them being CO, at industrially relevant current densities. Fabrication of robust GDEs, as well as the establishment of robust measurement protocols that allow the proper quantification of the products formed in high amounts, are required. In recent years, several measurement protocols that enable the rigorous quantification of CO_2 electroreduction products have been proposed in the literature. While more than ten years ago, the challenge was to detect a low amount of a formed product, today bigger amounts are formed using GDEs. Several important aspects discussed in the literature are the importance of using the gas flow stream after the reactor in calibrating the gas chromatographs in a range that fits the amount of products formed.[1,2]

Here, we discuss the challenges in product quantification via online gas chromatography (GC) that can reach high selectivity towards one product at industrially relevant current density. As model electrodes, we used previously developed GDEs based on Ni and Fe catalysts that enabled the selective synthesis of syngas (CO and H_2 mixtures) at current densities of up to 400 mA cm^{-2}.[3] Two different gas chromatographs (GCs) purchased from the same company were used to detect the formed gaseous products online. Depending on the online chromatography system used for the product quantification, we observed different variations in the total FEs from the expected 100% value, that, depending on the used setup, correlates with an increased amount of CO or H_2 production, respectively. To understand the differences in the recorded values from the two instruments, we performed a study in which the analysis of the limit of detection of the detectors as well as the saturation of the columns with the two products was performed in detail. The factors that impact the quantification of gaseous products via online chromatography will be presented. A new measurement protocol that allows the same amount of CO and H_2 formed during CO_2 electroreduction to be detected on the two different instruments will be shown.

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Nano-carbon-supported Molecular Electrocatalyst Modified Gas Diffusion Electrode for Highly Selective and Efficient CO_2 Conversion

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Renewable energy-driven electrocatalytic CO_2 reduction has been considered a promising approach for upcycling CO_2 into valuable chemicals, enabling a carbon-neutral economy. The success of such a system relies on the development of cost-effective catalysts that can efficiently and selectively catalyze the CO_2 reduction reaction (CO_2RR). Among the developed electrocatalyst materials, molecular complexes, such as cobalt phthalocyanine and cobalt tetraphenylporphyrin, have received great attention in recent years due to their unique properties[1, 2], including (i) capability of converting CO_2 into CO with high selectivity at the expense of low overpotential, (ii) molecular-level size, which can provide extremely high active sites per unit area, and (iii) a clear and controllable structure-activity relationship. However, these molecular complexes have a high tendency to form aggregates, especially at high-loading amounts on the electrode surface, which significantly deteriorates the overall catalytic performance due to the loss of active surface area and low electronic conductivity of aggregates[3]. In this research, we synthesize a nano-carbon-supported molecular catalyst (CNT-CoPc) using a non-toxic solvent with better dispersibility instead of the harmful organic solvent DMF. This catalyst demonstrates high selectivity for CO_2-to-CO conversion, achieving a Faradaic efficiency of 95.0% and a turnover frequency of 3.5 s^{-1} at an overpotential of 0.59 V in a H-type reactor. In addition, an effective strategy was developed to enhance the exposure of molecular catalyst active sites on the gas diffusion electrode (GDE) surface. The modified GDE exhibits outstanding eCO_2RR performance, achieving a CO current efficiency (CE_{CO}) of ≥ 90% at a current density of −100 mA cm^{-2} and maintaining CE_{CO} ≥ 80% over 12 hours. Furthermore, the electrode demonstrates competitive activity under low (40%) or diluted (20%) CO_2 conditions, maintaining CE_{CO} ≥75% and achieving 65% CO_2 conversion at a current of 0.625 A in a flow-type system.

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A Gas Diffusion Electrode Like Approach to Electrochemical Aldehyde Oxidation: Electrochemistry at the Liquid|Liquid|Solid Phase Boundary

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The use of gas diffusion electrodes for the electrochemical conversion of gaseous reactants at the gas|liquid|solid triple phase boundary is well established. However, it is not well-known that similar arrangements can be used to achieve the electrochemical conversion of liquid reactants at the liquid|liquid|solid triple phase boundary. In our presentation we are going to show that a GDE-like arrangement can be used to convert aldehydes selectively into carboxylates with current densities relevant for technical processes (300 mA/cm^2), while achieving Faradaic Efficiencies and yields close to 100%. We are also going to discuss why this reaction is relevant for the up-conversion of biomass to fuels and commodity chemicals.

Although the electrochemical oxidation of aldehydes is kinetically facile, the reaction is challenging because OH^- is an essential reactant and high current densities are only achieved in very alkaline electrolytes [1]. However, OH^- also induces chemical side-reactions such as aldol condensation that occur in the bulk electrolyte if conventional electrolysis arrangements are chosen in which the aldehyde is directly dissolved in the electrolyte. Accordingly, substantial amounts of the aldehyde enter a waste stream. Worse still, the formed decomposition products precipitate on the electrode leading to its rapid deactivation. In our presentation we demonstrate that this issue can be overcome when the reaction is conducted at the liquid|liquid interface between an alkaline electrolyte and an immiscible organic solvent featuring the aldehyde. In this arrangement the aldehyde and OH^- do not reside in the same phase and the decomposition reaction cannot occur in the bulk electrolyte. The electrochemical reaction is possible nonetheless when a porous electrode is placed at the liquid|liquid interface, where both reactants (i.e. aldehyde and hydroxide) are available

We will show that in this arrangement decomposition reactions can be avoided also at the liquid|liquid interface. That is, our DEMS and RRDE results show that the aldehyde must transition from the organic into the aqueous phase prior to the electrochemical step. In principle, this renders the aldehyde susceptible to OH^- induced decomposition reactions. Since aqueous and organic phase meet in the direct vicinity of the metal catalyst the aldehyde must only diffuse over a short distance before it undergoes the electrochemical reaction. Accordingly, there is only a short dwell-time of the aldehyde in the aqueous phase, which gives it little to no chance to engage in decomposition reactions.

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Nanostructured and multi-functional catalysts for the electrochemical reduction of CO_2

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The overreliance on fossil fuels has determined a rapid rise in atmospheric carbon dioxide concentrations and disrupted the natural balance of the carbon cycle. One way to close the carbon loop regards the Electrochemical Reduction of CO_2 (CO_2RR). Taking advantage of the Proton-Coupled Electron Transfer mechanism (PCET) in aqueous environment, it is possible to work at ambient temperature and pressure. However, the high thermodynamic stability of the molecule itself and its limited solubility in that media, makes the reaction poorly selective to one product and affected by a side-process, the Hydrogen Evolution Reaction (HER). Due to these problematics, the design of the right catalyst coupled with their use in efficient set-up and electrochemical cells has a fundamental importance for the direction of the reaction to get a certain CO_2RR product selectivity with high efficiencies, avoiding H_2 production [1]. Herein, cerium is studied as cerium dioxide (CeO_2) which forms, in a reducing environment, oxygen deficient phases with non-stoichiometric CeO_{2-x} oxides ($0 < x < 0.5$). Therefore, it will act as the co-catalyst for the electrochemical CO_2 reduction reaction able to absorb its oxygen on vacancies favouring formic acid production. Its coupling with carbon nitride-based support would improve the electron transfer process. Moreover, rare earth (RE) metals doping of cerium oxide were studied to evaluate their effect on oxygen vacancies and formation of C_+ products. The focus is given to Praseodymium (Pr), Gadolinium (Gd) and Yttrium (Y) REM doping synthetized by Prof. Tiziano Montini from University of Trieste [2]. Their performances were evaluated in a flow cell device, as electrochemical cell, with Gas Diffusion Electrode (GDE) configuration to overcome mass transport. CO_2RR gas products were quantified online through a Gas Chromatograph (GC) while those liquids were analysed with both Ionic Chromatography (IC) and ^1H-NMR spectroscopy.

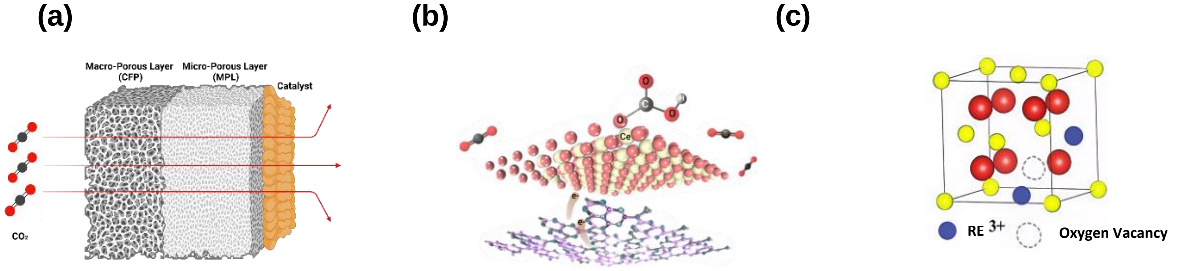

***Figure 2**. (a) Gas Diffusion Electrode structure. (b) Design of the catalyst: CeO_2 as co-catalyst supported on g-C_3N_4. (c) Rare Earth Metal doping effect on CeO_2.*

The catalysts were tested in chronoamperometry at both lower (from -0.5 V to -0.9 V vs. RHE) and higher (-1.1 V vs. RHE) overpotentials. The results obtained proved the ability of CeO_2 in directing the CO_2RR toward formic acid production within the entire potential window while, REM dopants improve C_{2+} products formation. In particular, Praseodymium is the best one in propanol production demonstrating its improved coupling and oxygen vacancies formation [3].

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Electrochemical Valorization of Glycerol and Concomitant H_2 Production by a Cobalt-based Metal Organic Framework

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Hydrogen generation by water electrolysis, powered through renewable energy, has been identified as a strong candidate for a future-proof energy vector and green chemical feedstock. However, large-scale applications and high energy conversion efficiencies are hampered by the sluggish anodic oxygen evolution reaction (OER). Alternative anodic reactions, such as the glycerol oxidation reaction (GOR), could provide a feasible alternative to the OER, decreasing the energy requirements for hydrogen production while anodically generating (valuable) organic oxidation products. However, alternative anodic reactions often suffer from a high activity-selectivity trade-off.[1] To address this, the design of suitable electrocatalytic materials is needed. In this context, metal-organic framework cobalt zeolitic imidazolate (Co-ZIF9(III)) , previously reported to be an effective catalyst for the OER [2], was prepared via a facile mechanical synthesis and evaluated as electrocatalyst for the GOR. Rotating disk electrode (RDE) voltammetry was performed to assess the activity of the catalyst in terms of overpotential and current density. Furthermore, chronoamperometric experiments were performed in a homemade flow cell to assess its stability and generated product distribution. Co-ZIF9(III) was found to be an effective catalyst upon the introduction of glycerol in the electrolyte, with a reduction in overpotential of up to 180 mV at a current density of 10 mA cm^{-2}, compared to the OER. Moreover, differential electrochemical mass spectrometry (DEMS) measurements were performed to assess potential inhibition of the OER reaction upon introduction of glycerol in the electrolyte. In addition, in-situ Raman spectroscopy was performed on systems in the presence and absence of glycerol, these results were compared. The impact of electrolyte composition on the selectivity was further demonstrated by conducting chronoamperometry under various conditions followed by HPLC, resulting in varied product mixtures. A higher glycerol concentration seemed to impede carbon-carbon bond scission, resulting in higher faradaic efficiencies for longer-chain carbon products. These results suggest a relationship between electrolyte composition and the distribution of products obtained, which can be optimized toward a target product.

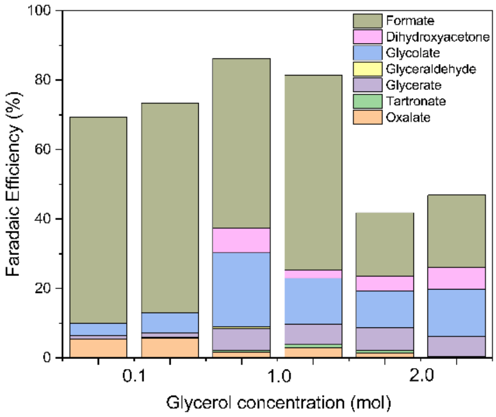

Figure 1. *Faradaic efficiencies for GOR products for different electrolyte compositions obtained after chronoamperometry (1.5 V vs RHE) for 24 hours.*

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Exploiting the Functionality of CeO_2@CNH Catalysts Towards Enhanced CO_2RR Performance

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The electrochemical reduction of CO_2 (CO_2RR) is a promising strategy for both greenhouse gas mitigation and the sustainable production of value-added chemicals and fuels. However, the development of efficient catalysts and advanced reactor configurations remains crucial to enhancing performance. In this work, we present a comparative study of cerium oxide (CeO_2)–carbon nanohorn (CNH) hybrid catalysts synthesized via two distinct routes: a conventional sol–gel method and a novel solvothermal approach.[1] The solvothermal method yields nanoflower with smaller, more homogeneously dispersed CeO_2 nanoparticles and improved integration with the CNH scaffold, resulting in an increased current density. This enhanced performance is attributed to better electron transport, higher metal oxide loading, and increased active surface area for CO_2 adsorption and conversion.[2] In parallel, we systematically investigate the influence of the electrochemical reactor design on CO_2RR performance. Experiments conducted in both static H-cell systems and gas diffusion electrode (GDE)-based flow cells reveal that the GDE configuration significantly boosts reaction rates and product selectivity. Notably, the GDE setup enables the formation of higher-value C_2 products such as ethanol and acetaldehyde, highlighting the importance of mass transport and CO_2 delivery strategies.[3]

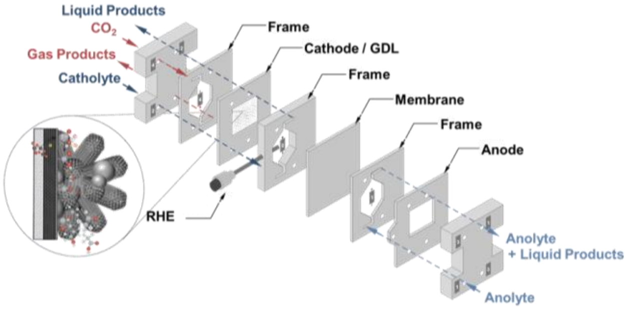

Figure 1. *MicroFlowCell system and CeO_2@CNH nanoflower.*

These results highlight the impact of CO_2 delivery methods on product selectivity and demonstrate the potential of combining nanostructured catalysts with advanced mass transport systems for efficient CO_2 conversion, paving the way for scalable, energy-efficient carbon utilization technologies.

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Electrode Design in Alkaline Ethanol Fuel Cells: Insights from Half-Cell GDE Studies

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The performance of alkaline direct ethanol fuel cells (ADEFCs) is strongly influenced by the physical and chemical properties of the electrodes, particularly the catalyst layer morphology, thickness, and deposition technique. In this study, the impact of electrode architecture on electrochemical performance was systematically evaluated under half-cell gas diffusion electrode (GDE) conditions, focusing on Pt-free catalyst systems relevant for ADEFC operation [1,2].

In a first investigation [1], the influence of catalyst layer thickness on the ethanol oxidation reaction (EOR) and oxygen reduction reaction (ORR) was examined. Gas diffusion electrodes with varying loadings of PdNiBi/C (anode) and Ag–MnO_x/C (cathode) were fabricated by ultrasonic spray coating and electrochemically characterized in a three-electrode half-cell configuration. The half-cell GDE measurements reveal a clear enhancement of both ORR and EOR activities with increasing temperature. Higher temperatures improve diffusion and reaction kinetics, leading to significantly higher current densities for both anodes and cathodes. Thicker catalyst layers particularly benefit from these conditions.

A complementary study [2] focused on the influence of electrode fabrication methods on catalyst layer structure and activity. Using graphene-supported catalysts, four deposition techniques (spray coating, drop coating, brush coating, and roll coating) were evaluated with respect to morphology and electrochemical performance. Among the tested configurations, the brush-coated cathode and the ultrasonic spray-coated anode exhibited superior activity. These findings can be attributed not only to the catalyst properties but also to the nature of the gas diffusion layers and the contrasting requirements regarding hydrophilicity and hydrophobicity at the anode and cathode. Consequently, both the structural integrity of the catalyst layer and the suitability of the deposition technique are dependent on the specific functional role of the electrode.

These findings highlight the importance of optimizing both electrode structure and fabrication parameters for high-performance, Pt-free ADEFC systems. The half-cell GDE approach proved to be a reliable tool for isolating and studying individual factors that influence catalyst layer behavior, and serves as a valuable platform for electrode development.

Acknowledgements:

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Electrochemical Impedance Spectroscopy as a Window into Electrochemical CO_2 Reduction Mechanism

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The electrochemical reduction of CO_2 (eCO_2RR) into value-added products has emerged as a promising approach for renewable energy storage and carbon recycling. Among various electrocatalysts, silver is particularly attractive due to its ability to selectively produce carbon monoxide (CO) and hydrogen (H_2) during electrolysis. However, for enhanced efficiency and commercial viability, the suppression of the competing Hydrogen Evolution Reaction (HER) is essential. Our previous work established that CO selectivity under dynamic (pulsed electrolysis) conditions is influenced by the kinetic parameters of two competing reactions, where the reaction with a higher charge transfer coefficient value shows greater selectivity under dynamic conditions[1]. In a separate study, HER was found to exhibit a distinctive Z-shaped response linked to the roles of proton donors and the kinetic interplay between HER and eCO_2RR[2].

To deepen the kinetic understanding of electrochemical CO_2 reduction (eCO_2RR), Electrochemical Impedance Spectroscopy (EIS) is employed in this study to investigate the underlying processes in the frequency domain. The previously published continuum microkinetic model is extended to simulate EIS spectra, which are then compared with experimental measurements conducted using a three-electrode setup on a silver rotating disk electrode (RDE). The simulated and experimental EIS spectra were found to exhibit two distinct features: one arising from the combined kinetics of the two reactions and another attributed to adsorption kinetics depended on the applied overpotential, as illustrated in Figure 1. Additionally, a sensitivity analysis was carried out to assess the influence of various parameters on the EIS response. These findings highlight the potential of combining microkinetic modeling with EIS analysis to gain mechanistic insights and demonstrate the ability to guide characterization of more selective and efficient catalysts for eCO2RR.

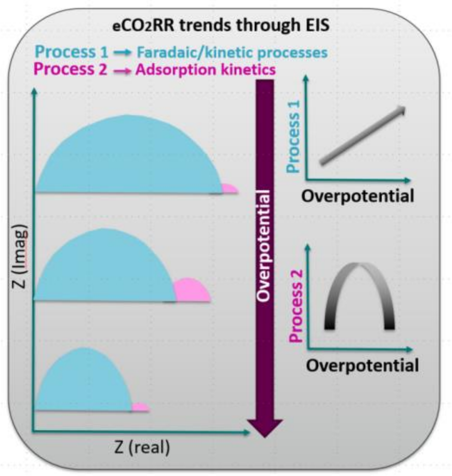

Figure 1: *Distinct Processes observed in EIS spectra during eCO2RR as a function of overpotential*

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Boron-Doped Diamond Mesh Electrodes for Zero-Gap Electrolysis

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In this contribution, I will discuss the use of boron-doped diamond (BDD) electrodes in a zero-gap electrolyzer configuration for alternative anodic processes, with focus on anodic production of H_2O_2.

Recent studies have proved that BDD anodes feature excellent stability and catalytic properties for H_2O_2 electrosynthesis. BDD electrodes, however, are usually tested in the form of non-porous electrodes in h-cells [1,2]. I will present our work on a zero-gap PEM electrolyzer with a BDD-coated Nb mesh anode [3] and show the feasibility of anodic H_2O_2 formation by partial water oxidation in a flow-cell configuration, pairing the anode reaction with cathodic hydrogen evolution. In addition, I will discuss the effect of process parameters (e.g., recirculated vs. single-pass anolyte flow, flow rate, etc.) and show that pulsed electrolysis significantly enhances the product (H_2O_2) formation efficiency. Particularly, we systematically investigated the on-to-off time ratio and amplitude of the current-pulse cycles and achieved a 70 % increase in Faradaic efficiency to H_2O_2 compared to constant-current electrolysis at industrially relevant current densities (i. e.,150 mA cm^{-2}) [4]. In future work, we aim to tune membrane-electrode-assembly components (e.g., BDD morphology and properties), cell hardware (flow field design), and process parameters (current pulse frequency, shape, etc.), to further increase the cell performance. Another important aspect to investigate is the long-term stability of the cell under dynamic operation.

Finally, I will touch upon the perspective application of BDD anodes in CH_4 activation [5] and CO_2 reduction [6].

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Microenvironment effects from first-principles multiscale modelling of electrochemical CO_2 reduction

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Electrochemical CO_2 reduction (eCO_2R) is a promising route to achieve net-zero emissions, but optimizing and scaling up electrode/electrolyte interfaces remains challenging. Along with catalyst design, electrolyte microenvironments, especially cations, play a crucial role in improving the Faradaic efficiencies [1,2]. However, tools for optimizing electrolyte effects are still lacking due to complex interplay between surface kinetics and transport processes [3]. In this work, we develop an ab-initio multiscale model coupling DFT-derived microkinetics with a continuum-scale transport model to study eCO_2R on Ag surfaces for both liquid electrolytes and ionomers [4]. By explicity accounting for the cations across all scales, the current density trends across different cation buffer identities align with experiments. Low concentration buffers show a favorable trade-off between Faradaic efficiencies and carbonate precipitation, showing optimal performance. Further, we observe a volcano dependence for current densities as a function of buffer concentrations, due to the inverse relationship between concentration of cations and CO_2 at the Outer Helmholtz Plane. The ionomers can break this dependence due to fixed charges on their backbones, however, water management becomes crucial to go towards high current densities. Overall, this study paves the way towards rational electrolyte design in eCO_2R.

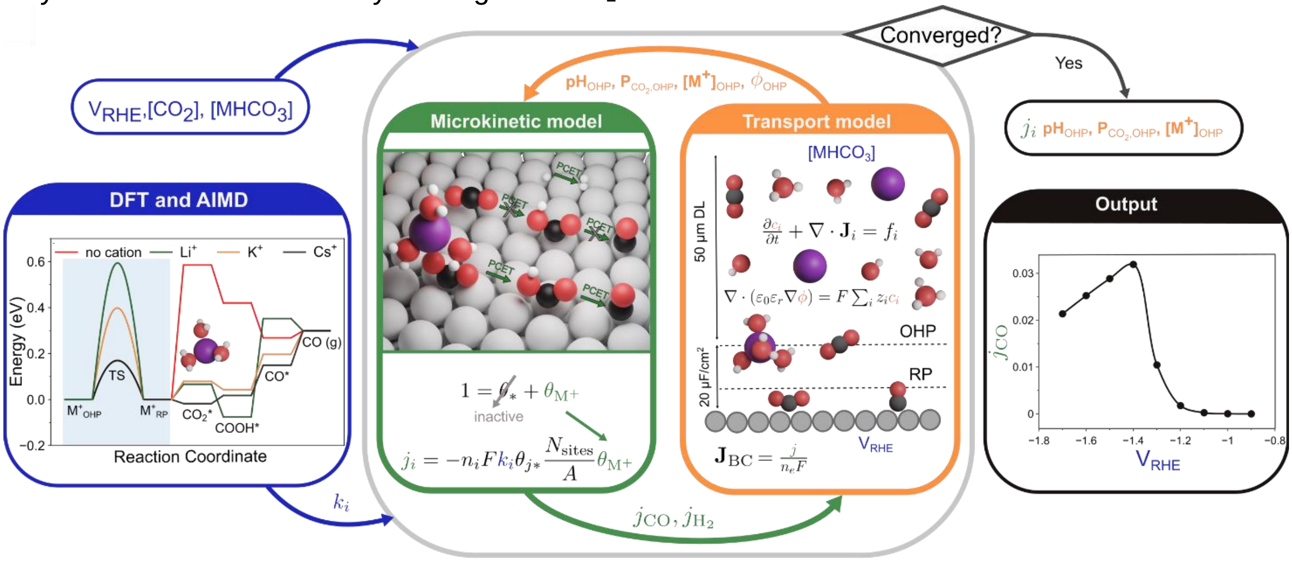

***Figure 1**: Overview of multiscale methodology for electrochemical CO_2 reduction to CO.*

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Fundamental Insights into the Nitrogen Oxidation Reaction over Pd-Based Electrodes

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The conventional nitrate synthesis industry combines steam reforming with the Haber-Bosch and Ostwald processes, which require extreme conditions with huge energy consumption and greenhouse gas emission. In very recent years, a few experimental studies have reported that Pd-based electrocatalysts have affinity for the nitrogen oxidation reaction (NOR) [1–4], thus offering the exciting opportunity to directly convert dinitrogen from the air into nitrate under applied bias:

$$\mathrm{N_2 + 6H_2O \rightarrow 2NO_3^- + 12H^+ + 10e^-}, U^0_{\mathrm{NOR}} = 1.08\ \mathrm{V\ vs.RHE@pH} = 14.$$

Therefore, the NOR could lead to a sustainable formation of nitrates by taking the required electricity from renewable energy sources and running the process under near-ambient conditions. Regrettably, the NOR is impaired by a selectivity problem since the competing oxygen evolution reaction (OER),

$$\mathrm{2H_2O \rightarrow O_2 + 4H^+ + 4e^-}, U^0_{\mathrm{OER}} = 1.23\ \mathrm{V\ vs.RHE},$$

is both thermodynamically (lower equilibrium potential) and kinetically (less electrons transferred) preferred over the NOR. This makes the development of selective NOR catalysts challenging and rewarding at the same time. In this context, density functional theory (DFT) calculations have been successfully employed to provide fundamental insights into possible NOR mechanisms on metal oxides [5,6]. While these studies represent a significant step toward the rational design of selective NOR catalysts, atomic-scale information on the metal termination of electrocatalysts remains unknown.

By combining DFT-based modelling with the computational hydrogen electrode approach [7], we present fundamental insight into the NOR and OER over Pd-based surfaces. We construct surface Pourbaix diagrams and free-energy diagrams along the reaction coordinate that are evaluated by advanced screening protocols [8–10]. A mechanism of the NOR on Pd(111) surfaces is proposed, including side reactions for the formation of N_2O, N_2O_2, NO, and NO_2^- besides the NO_3^- formation. Our results are compared with the proposed mechanism for metal oxides, which involves a series of chemical and electrochemical steps. The reported findings open new possibilities to improve the selective conversion of N_2 to nitrogen-based compounds over metal-based electrodes while suppressing the parasitic OER.

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Enhancing Microenvironment for CO_2 Reduction: Advanced Silver Catalysts and Ionomer Binders for Improved Carbon Monoxide Production

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Carbon monoxide is a vital industrial base chemical that can be transformed into various valuable compounds through methanol or Fischer-Tropsch synthesis [1]. This potential drives interest in the electrocatalytic reduction of CO_2, mainly using silver catalysts recognized for their high Faradaic efficiency in generating carbon monoxide. Gas diffusion electrodes (GDEs) offer promise in facilitating CO_2 reduction reaction (CORR) by supporting the three-phase boundary zone, but limitations in increasing current density challenge industrial application [2]. This affects the capacitive double layer at the electrolyte-electrode interface, leading to potential electrolyte flooding and impaired CO_2 mass transfer due to diminished hydrophobic properties.

This study explores binder materials incorporating ion-conducting polymers (Ionomers) to improve Faradaic efficiency at high current densities. These polymers, featuring hydrophobic backbones and functional group side chains, create an optimal microenvironment at the electrolyte-catalyst interface (Fig. 1). The promising effects of ionomers have been demonstrated by Hoffmann et al. [3], a team from Bayreuth University who are our partners in this project. Building on this research, we investigate various ionomers, including anion and cation exchange types such as Sustainion and Nafion, at different concentrations and compare them with commercial Covestro electrodes. Given the high cost of silver, we initially experimented with silver flakes. Subsequently, our project partner produced silver foam, offering a larger surface area structure. This increased surface area is crucial for enhancing the exposure of active sites and improving the binding efficiency for CO dimerization.

Chronoamperometry tests will assess electrode performance at varying current densities, while electrochemical impedance spectroscopy (EIS) will evaluate the capacitive double layer. The research will utilize characterization techniques to investigate electrode morphology and surface chemistry.

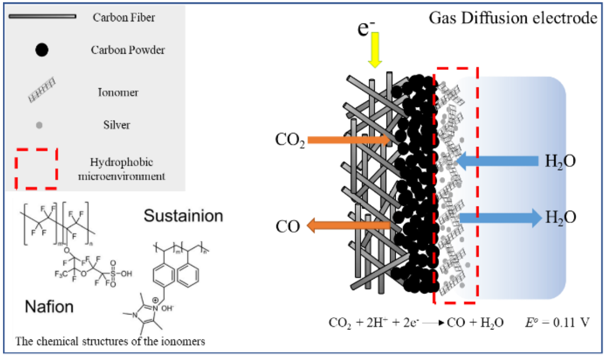

***Figure 1**. A hydrophobic environment enhances the three-phase boundary for CO2RR, while a hydrophilic environment may cause GDE flooding and boost H_2 generation.*

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Ni Foam-Based Gas Diffusion Electrodes for the Electrooxidation of Gaseous Ammonia to Nitrite and Nitrate

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Due to its high hydrogen content and carbon-free nature, ammonia is emerging as a promising hydrogen carrier, offering a solution to the challenges associated with hydrogen storage and transportation. The electrocatalytic ammonia oxidation reaction (AmOR) presents a viable hydrogen recovery strategy, enabling the production of environmentally neutral nitrogen or oxidized nitrogen species, such as nitrite (NO_2^-) and nitrate (NO_3^-), which are conventionally obtained through the energy-intensive Ostwald process.[1] A selective AmOR process in an electrolyzer setup could provide dual benefits: replacing the energetically demanding oxygen evolution reaction (OER) while simultaneously generating value-added products at the anode and while concomitantly producing hydrogen at the cathode.

Most published studies on AmOR for nitrite and nitrate production have focused on aqueous ammonia-containing compounds.[2] However, the direct oxidation of gaseous ammonia, particularly in the context of ammonia as a hydrogen carrier, could significantly simplify the process. To explore this approach, we introduced a proof-of-concept electrolyzer cell in which gaseous ammonia is directly oxidized using specially designed Ni foam-based gas diffusion electrodes (GDEs).[3] Using an airbrush-type spray coater, a multi-metal catalyst layer of the desired composition is deposited onto the Ni foam substrate, creating a high-surface-area catalyst layer. A porous gas diffusion layer is then formed by spraying a carbon-based suspension onto a PEEK mesh.[4]

A variety of GDEs with different catalyst layers were prepared and investigated for AmOR. The results demonstrated that, under suitable reaction conditions, high NO_2^-/NO_3^- selectivity can be achieved, while effectively suppressing oxygen formation via the undesirable OER and minimal nitrogen production.

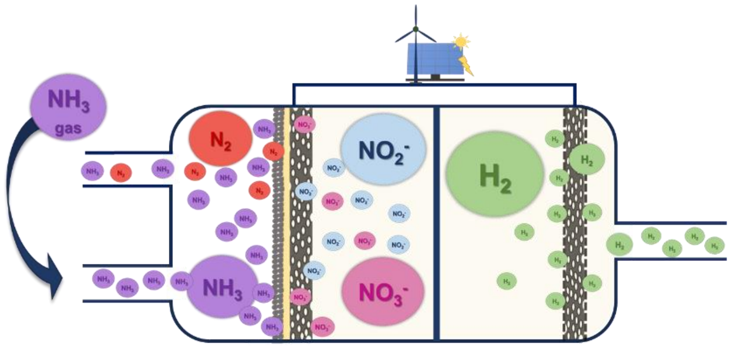

***Figure 1**: Proof-of-concept electrolyser scheme for electrooxidation of gaseous ammonia using gas diffusion electrode. Adapted from Ref. [3].*

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Stability of Ionomers in Membrane Electrode Assemblies for Electrochemical CO_2 Reduction at Elevated Temperatures

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Electrochemical CO_2 reduction presents a promising pathway for decarbonization while enabling the sustainable production of valuable chemicals. By using renewable energy, CO_2 can be electrochemically converted e. g. into hydrocarbons or alcohols, depending on the catalyst material. Silver-based catalysts exhibit high selectivity for carbon monoxide formation, with hydrogen as the only byproduct occurring in significant amounts.

Gas diffusion electrodes (GDEs) help mitigate mass transport limitations associated with CO_2 solubility, still enabling high Faradaic efficiencies for CO at elevated current densities [1]. However, challenges such as low energy efficiency and fast electrode degradation hinder the industrial feasibility of the process. Membrane electrode assemblies (MEAs) offer a strategy to reduce internal cell resistances caused by the electrolyte gaps in conventional flow-cell setups with GDEs. Nevertheless, issues such as salt precipitation become more severe in this setup [2], obstructing CO_2 diffusion pathways and therefore inhibiting CO production. Among others, the type of membrane and ionomer used for manufacturing the MEA strongly influence the extent of salt deposition on the cathode. Fig. 1, left, exhibits the resulting performance differences of MEAs with varying anion exchange ionomers and membranes.

While the results shown here were obtained at room temperature, industrial electrolyzers will inevitably function at moderately elevated temperatures due to heat generation from the system and peripheral components [3]. Temperature affects multiple factors, including mass transport, conductivity, and overall performance, but also the stability of ionomers. As depicted in fig. 1, right, even a 10 °C increase can significantly shift MEA performance, with more pronounced effects expected at operational temperatures between 50 °C and 60 °C. This study systematically investigates the influence of temperature on MEA performance, with a particular emphasis on the thermal stability of different ionomer materials.

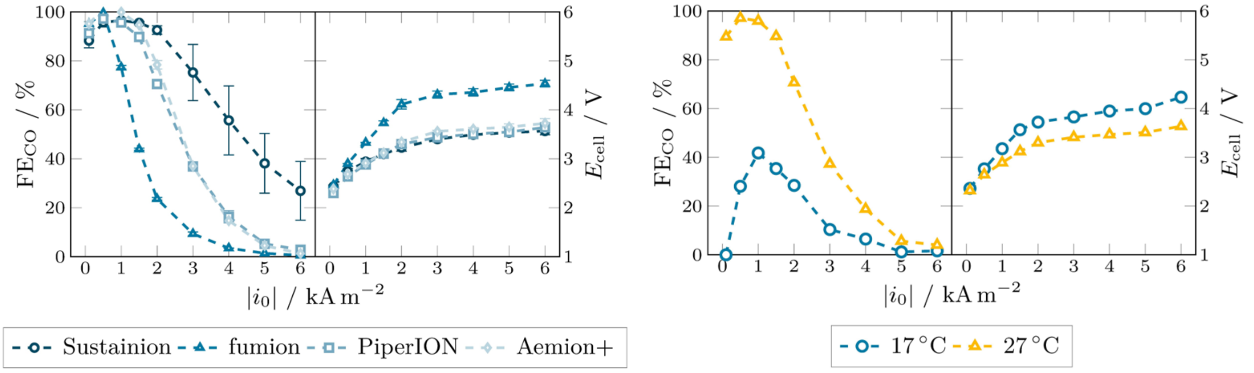

***Figure 1**: Faradaic efficiency for CO and cell potential obtained from galvanostatic step experiments in MEA setup at room temperature using different ionomers and corresponding membranes (left) and at different temperatures using PiperION ionomer and membranes (right).*

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Membrane Assembly Electrodes for High Power Direct Ammonia Fuel Cells

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Renewable hydrogen energy plays a key role in the implementation of zero carbon emission in green electricity. Hydrogen energy includes ammonia is an alternative form for such green energy. Ammonia with advantages of storage and transportation mobility are noticed particularly for power generation [1, 2]. This report will examine several key variables that directly impact the performance of low-temperature direct ammonia fuel cells (DAFC) using a cross-comparison method to achieve optimal test parameter settings. Additionally, the study explores the use of non-precious metal catalysts instead of traditional platinum catalysts at the cathode, aiming to reduce costs while maintaining high power density. For the cathode catalyst, iron (Fe) and copper (Cu) were tested on different carbon carriers, and the successful preparation of the FeCuN/C electrocatalyst was verified through SEM, EDS, and XRD analyses. The electrochemical results from cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) indicated that FeCuN/C exhibited more distinct redox peaks and the lowest impedance of 22.15Ω. At the anode, 40wt% PtIr/C was selected for its proven high efficiency and stability. Both anode and cathode are combined with an anion exchange membrane (AEM) to composed a membrane electrode assembly (MEA). The power density of the DAFC was measured in a mixed solution of ammonia (NH_3) and potassium hydroxide (KOH) under O_2, achieving a maximum power density of 269 mW/cm² at a working temperature of 110°C and a catalyst load of 2 mg/cm² for FeCuN/C. To improve the power density, Pd and Co were introduced, resulting in the synthesis of a novel 15wt% PdAgCo/C cathode catalyst, which increased the power density to 74 mW/cm². Ultimately, by adjusting the loading of both the anode and cathode to 2 mg/cm², the power density reached a maximum of 332 mW/cm². This study can implement to use ammonia as a fuel for DAFC and further to construct ammonia fuel electrical generators.

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Enhancing Hydrophobicity in Sustainable Gas Diffusion Electrodes for Electrochemical CO_2 Reduction

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A promising technology for the reuse of emitted carbon dioxide (CO_2) is the electrochemical CO_2 reduction (eCO_2R) [1]. A crucial component of the CO_2 electrolyzer is the gas diffusion electrode (GDE). A GDE provides a large three-phase boundary, helps to overcome the solubility problems of CO_2 in aqueous electrolytes and increases the achievable current density [2]. In these GDEs, PTFE is used as a polymeric binder to stabilize the GDE while also improving the hydrophobicity [3]. Due to the environmental impact of PFAS, which are necessary for PTFE production, the European Union is planning to ban these PFAS [4]. Therefore, a replacement of PTFE as polymeric binder must be found.

In initial tests, PEEK has been used as alternative for PTFE, because it offers similar thermal and chemical stability. Thus PEEK is suitable for the production process of Moussallem et al. [3], which includes thermal treatment at elevated temperatures. However, PEEK has a lower hydrophobicity than PTFE, resulting in a GDE that was prone by electrolyte flooding. In this work, an approach to increase the hydrophobicity further is presented. The incorporation of superhydrophobic methyl MQ silicone resin-coated SiO_2 particles ($MeMQ/SiO_2$) into the GDE has been demonstrated to enhance its hydrophobicity, thereby facilitating the formation of a sufficiently large three-phase boundary within the GDE. The left-hand diagram of figure 1 reveals a similar initial performance of this GDE compared to a baseline PTFE-GDE. However, the GDE underwent rapid degradation due to side reactions, as can be seen from the right-hand diagram in figure 1, after which only the undesirable hydrogen evolution reaction took place. In future work, the production process will be changed in order to exclude thermal treatment of the GDE. This way, other promising but temperature-sensitive superhydrophobic particles can be also tested.

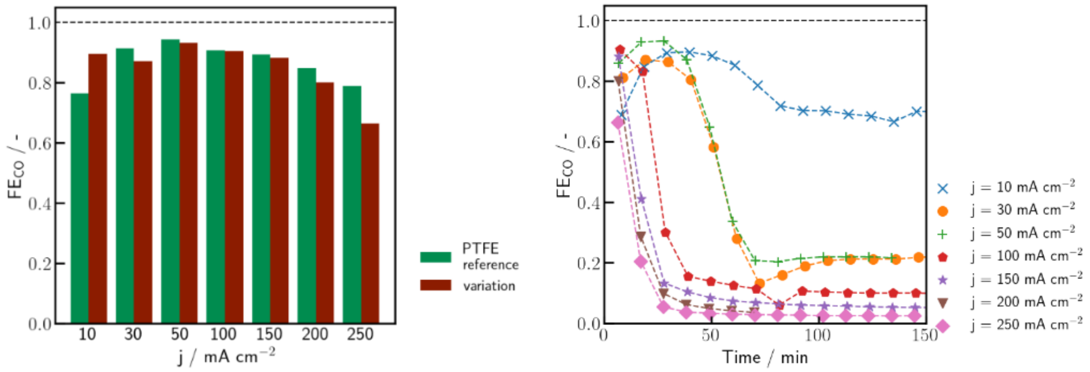

Figure 3: *Initial Faradaic efficiency for CO for a GDE with increased hydrophobicity compared to a baseline PTFE-GDE (left) and Faradaic efficiency for the new GDE as a function of time (right)*

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Coupling Electrocatalytic CO_2 Reduction with Ethanol Oxidation for High-Yield Acetic Acid Production in a Dual-Reactor System

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Electrochemical conversion of CO_2 into value-added chemicals coupled with organic oxidation reactions presents a promising pathway for sustainable chemical synthesis and efficient carbon utilization.[1] This study demonstrates an integrated two-reactor system for the efficient production of acetic acid by coupling electrocatalytic CO_2 reduction with electrocatalytic ethanol oxidation. In reactor 1 of a model electrolyzer system, CO_2 is electrochemically reduced to CO at high current densities at a catalyst-modified gas diffusion electrode, while ethanol is concomitantly oxidized to acetate at the anode.

At a current density of 400 mA cm^{-2}, the Faradaic efficiency (FE) for CO production reaches 99%, while the FE for acetate formation during ethanol oxidation is nearly 80%, with a production rate of 45 µmol min^{-1} cm^{-2}. The CO_2/CO mixture is then utilized in **reactor 2** to enhance the production rate of C_{2+} products using a structurally defective metal-organic framework. During this tandem cell system **reactor 1** was working at 500 mA cm^{-2} and **reactor 2** was operated at -2.1 V vs Ag/AgCl (3 M KCl). Compared with the single cell system the tandem cell showed an increase in acetate production from 0.573 µmol min^{-1} cm^{-2} to 2.12 µmol min^{-1} cm^{-2} while the production rate for ethylene has increased from 1.46 µmol min^{-1} cm^{-2} to 2.5 µmol min^{-1} cm^{-2}. Additionally, the ethanol formed at the cathode is recirculated towards the anode, which undergoes further conversion towards acetate. This dual-reactor configuration not only maximizes the utilization of CO_2 but also minimizes energy consumption by coupling the anodic ethanol oxidation with cathodic CO_2 reduction.

This work provides an efficient strategy for producing acetic acid, a valuable chemical feedstock while addressing CO_2 emissions and energy efficiency challenges in electrochemical systems.[2] The system achieves a high acetic acid yield, demonstrating significant improvements in both FE and overall production rates of acetic acid compared to conventional single-reactor approaches. This approach highlights the potential of coupled electrochemical processes for sustainable chemical manufacturing and carbon valorization.

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Acknowledgement:

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Optimizing GDE Catalyst Layer Composition for Durable CO_2 Electroreduction to Formate

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The electrochemical reduction of carbon dioxide ($ERCO_2$) to formate offers a sustainable and scalable route for carbon capture and utilization, in line with global decarbonization goals. However, the long-term stability of gas diffusion electrodes (GDEs) remains a critical challenge for the industrial application of this technology [1]. In this work, we systematically investigate how variations in catalyst layer (CL) composition affect both the performance and durability of GDEs for selective formate production. The parameters explored include ionomer type, catalyst-to-ionomer ratio, and the incorporation of hydrophobic additives such as polytetrafluoroethylene (PTFE).

Bismuth subcarbonate ($(BiO)_2CO_3$) is chosen as the active catalytic material because of its high selectivity toward formate under mild electrochemical conditions. We initiallycompare the performance of two ionomers: Nafion, a proton-conducting ionomer, and Sustainion, an anion-conducting ionomer. Our results indicate that Nafion-based GDEs exhibit good selectivity at low ionomer concentrations, increasing the Nafion content leads to catalyst agglomeration, pore blockage, and an enhanced hydrogen evolution reaction (HER). In contrast, Sustainion-based GDEs maintain high Faradaic efficiencies (FE) for formate across a wider range of compositions by effectively suppressing HER. Nevertheless, excessive ionomer loadingwith Sustainioncan impede CO_2 transport by clogging the porous structure of the CL, thereby reducing catalytic efficiency [2].

To overcome these limitations and enhance long-term stability, we introduce PTFE into the CL formulation alongside Sustainion. By carefully tuning the PTFE content, we achieve an ideal balance between hydrophobicity and ion transport, thereby mitigating electrolyte flooding and ensuring consistent CO_2 access to the active sites. This optimized design enables continuous electrolysis for 24 hours while maintaining a high FE for formate (~85%) and suppressing HER to below 10%. In contrast, GDEs without PTFE—irrespective of whether they employ Nafion or Sustainion—suffer from progressive electrolyte intrusion, resulting in reduced formate selectivity and increased HER over time. These findings underscore the importance of rational CL design in the development of high-performance, durable GDEs for $ERCO_2$ applications. By leveraging the benefits of anion-conducting ionomers and incorporating hydrophobic additives, this study provides valuable insights and a practical approach for advancing $ERCO_2$-to-formate technologies toward industrial relevance.

Acknowledgments:
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Exploring oxygen depolarized cathode for CO_2 electrolysis in zero-gap cell

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CO_2 electrolysers utilize renewable energy to convert CO_2 into valuable products, demonstrating significant potential in addressing both environmental and energy challenges. Among them, gas diffusion electrodes (GDEs) are a key component which aids in transport of gaseous reactants to the catalyst surface thereby boosting reaction rates. In general, GDEs require high conductivity, pore structure for efficient gas permeability, and proper hydrophobic-hydrophilic balance to prevent flooding. Conventional GDEs are affected by electrowetting wherein the porous network is flooded at high current densities hindering CO_2 transport. The chlor-alkali industry has accumulated extensive experience in addressing these challenges during the development of oxygen depolarized cathodes (ODCs) [1]. Adaptation of this expertise into CO_2 electrolysis in flow cells has been demonstrated to be feasible [2], however, direct implementation in a zero-gap configuration remains challenging. [3],[2]. Indeed, it requires optimization of the process parameters and precise control of water management in the GDE along with the membrane. In this study, we explore a silver based ODC for CO_2 electrolysis toward CO production in membrane electrode assembly (MEA) configuration. Preliminary results show that the ODC was capable of reaching 3 kA/m^2 with 60% faradaic efficiency (FE) of CO at a cell potential of 3.03 V (Fig 1a). Without any mitigation, the FE of CO maintained around 86% up to 6 hours at 1 kA/m² (Fig 1b) promising long term stability of the overall cell. Additionally, at elevated temperatures, the potential was reduced to 2.2 V at 1 kA/m^2 (Fig 1c) lower than previously reported values[3].This study further aims to optimize reaction conditions to ensure stable long-term CO_2 electrolysis in a MEA cell using this convential ODC.

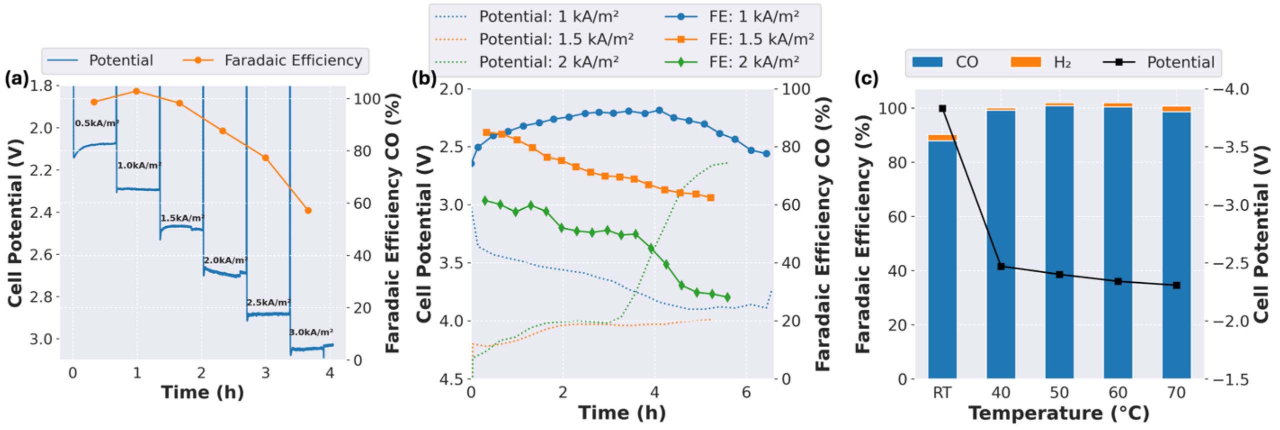

***Figure** 4 Catalytic performance of the oxygen depolarized cathode (ODC) for CO_2 electrolysis with 35 S.mL/min CO_2 supply and 1M KOH anolyte: (a)Quick ramp up at different current density up to 3 kA/m² with. (b) 6-hours electrolysis at three different current densities. (c)Testing at different temperatures and 1 kA/m².*

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Engineering novel gas diffusion electrode microstructures for electrochemical CO_2 reduction

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The gas diffusion electrode (GDE) is a critical component in CO_2 electrolyzers, enabling current densities an order of magnitude higher than those achievable in H-type cell systems[1]. Commercial GDEs typically feature a hydrophobic dual-layer design - a porous carbon fiber-based substrate that facilitates gas, electrons, and heat transport, while providing mechanical support; and a microporous layer that improves contact with the catalyst layer and manages liquid transport[2]. While these electrodes - which have been repurposed from low temperature fuel cell technology - are functional, they may not fully meet the specific demands of CO_2 electrolysis possibly due to suboptimal microstructural characteristics, such as pore size distribution and surface wettability. Our group has recently developed a fabrication method based on non-solvent induced phase separation (NIPS) for synthesizing non-fibrous porous electrodes[3 4]. This technique, originally developed for membrane fabrication[5], enables precise control over electrode microstructure, enabling the manufacturing of structures (e.g. pore size gradients, multimodal pore size distributions) which are not attainable with traditional techniques. The process involves dissolving a carbon precursor polymer (e.g., polyacrylonitrile) and a pore-forming agent (e.g., polyvinylpyrrolidone) in a solvent (e.g., N,N-dimethylformamide), casting the solution onto a mold, and immersing it in a non-solvent (e.g., water) to initiate phase separation. The resulting polymer scaffold is then carbonized under an inert atmosphere to form a conductive carbon network. By adjusting the solvent/non-solvent exchange conditions, a range of microstructures can be achieved.

Here we investigate the influence of the GDE microstructure on the performance and stability of CO_2 electrolysis cells. To achieve this, we fabricate electrodes with distinct pore architectures and evaluate their performance with a copper catalyst layer in a zero-gap CO_2 electrolyzer. We hypothesize that overcoming multiphase transport limitations in CO_2 electrolysis requires electrodes with hierarchical pore architectures and well-controlled structural features. We aim to stablish a relationship between synthetic parameters and key microstructural attributes such as pore size distribution and porosity, as well as transport-related properties including permeability, capillarity, and diffusivity. These characteristics are systematically correlated with performance metrics obtained from electrochemical CO_2 reduction experiments, enabling the development of structure–property–performance relationships. Ultimately, by fine-tuning the pore architecture and surface characteristics of the GDEs, we aim to enhance mass transport, mitigate flooding, and enable higher current densities with improved operational stability.

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GDE design rules for a scalable zero-gap CO_2 electrolyzer

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Renewable electricity-driven electrochemical conversion of CO_2 into fuels and value-added chemicals holds significant promise for decarbonizing various sectors and providing a storage solution for intermittent renewable energy. Research indicates that low-temperature CO2 electrochemical conversion can be economically viable if operated efficiently, especially when the final product prices and market sizes are favorable [1-2]. Given that the aviation sector accounts for 2.5% of global CO_2 equivalent emissions and is projected to increase by 4% annually, the need for sustainable aviation fuel (SAF) is more pressing than ever. Airlines have committed to achieving net-zero CO_2 emissions by 2050, with SAF expected to contribute significantly to this goal. As such, the production of CO_2-derived fuels, particularly through pathways that convert CO_2 into liquid hydrocarbons suitable for jet fuels, is essential. The direct electrochemical reduction of CO_2 to C_2H_4 can serve as a key intermediate in these processes, highlighting the importance of developing efficient CO_2 conversion technologies to meet the growing demand for SAF and support the aviation industry's decarbonization efforts [3].

The viability of this electrochemical route, however, hinges on its competitiveness with established technologies. For instance, C_2H_4 production for jet fuel applications must compete with the more mature methanol synthesis pathway. Our Aspen Plus simulations, confirming the existing literature [3,4], demonstrate that a minimum energy efficiency of 28% is required for the electrochemical CO_2 to C_2H_4 conversion to be competitive. This energy efficiency is primarily determined by the electrolyzer's faradaic efficiency and cell potential, which are, in turn, dictated by factors such as catalyst activity, cathodic GDE design, membrane properties, anodic GDE design, and overall cell architecture. In this work, we specifically focus on the influence of cathodic GDE design and the GDE/membrane interface on achieving the necessary energy efficiency for competitive CO_2 electrolysis. We will present the results of our Aspen Plus process simulations, detailing the minimum energy efficiency requirements and the resulting GDE design rules (in-plane conductivity, through-plane conductivity, and ionic conductivity). Furthermore, we will show sensitivity analyses to illustrate the trade-offs between C_2H_4 faradaic efficiency and the allowable GDE design parameters.

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Resolving Voltage Losses in PEMWE via Operando Neutron Imaging and Advanced Electrochemical Characterization

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With the aim to decarbonize different economic sectors, coupling proton exchange membrane water electrolyzers (PEMWEs) and renewable energy resources has emerged as an appealing option: PEMWEs store electrical energy produced by renewables as green hydrogen that can be used as an eco-friendly fuel. However, decreasing the costs of PEMWEs and improving its durability is crucial for making green hydrogen compatible with fossil fuels [1]. This can be achieved only if an improved understanding of the intrinsic processes occurring in PEMWE is obtained.

In this work, we apply advanced methods to deconvolute the performance losses of the PEMWE. In-plane neutron imaging of the PEMWE is performed to investigate the two-phase transport of water and gases in the PEMWE. The influence of the operating conditions (current density and flow rate) and porous transport layer (PTL) porosity on the water thickness of the PEMWE was investigated. Furthermore, electrochemical characterization, consisting of the polarization curve and nonlinear frequency response method [2] measurements, was conducted with the goal of identifying the different voltage losses of the electrolyzer.

The synergy of different experimental approaches allowed further insights into the PEMWE performance. The voltage losses were found to be dependent on the water transport to the catalyst-coated membrane (CCM). The transport of the water determined the catalyst layer and membrane humidity, thus affecting both kinetics and ohmic resistance. While the current density only slightly affected the PTL and CCM humidity, the PTL structure influenced it significantly, with higher porosity PTL allowing higher CCM humidity and, therefore, lower voltage losses (Figure 1).

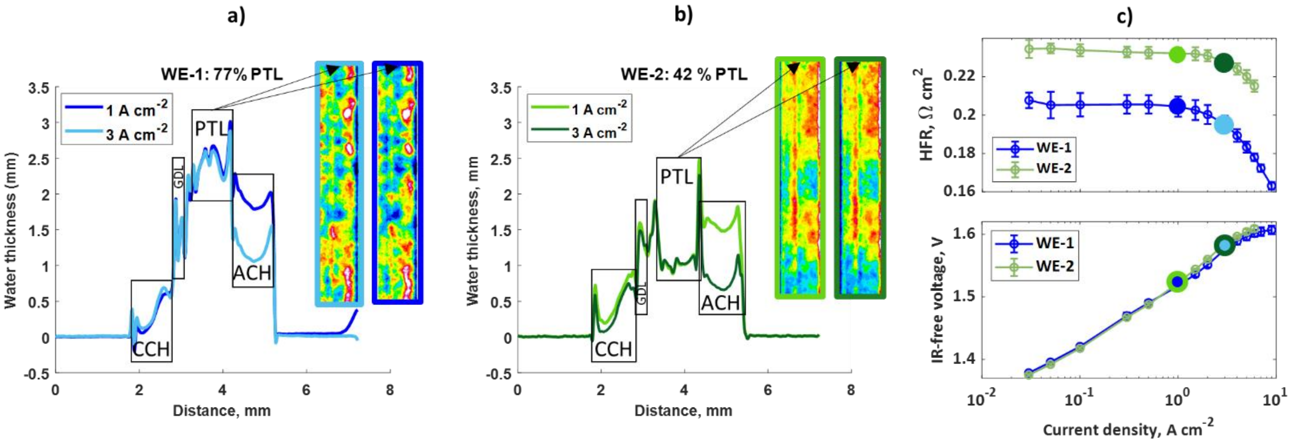

Figure 1: *Water thickness profiles across the sandwich coordinate of the PEMWE with a) 77 % porosity PTL and a) 42 % porosity PTL at current densities of 1 and 3 A cm^{-2}, and c) HFR and IR-free polarization curve of the PEWMEs with 77 and 42 % porosity PTL*

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Importance of Cation Management at the Cathode Gas Diffusion Electrode for CO_2 Electrolysis

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CO_2 electrolysis using gas diffusion electrodes (GDEs) has emerged as a promising technology for converting waste CO_2 into valuable chemicals. As this technology gains industrial traction, improving long-term durability is crucial to achieve the target lifetimes of tens of thousands of hours needed for economic viability.

One major performance degradation mechanism in anion exchange membrane-separated zero-gap CO_2 electrolyzers is carbonate precipitation formation and related flooding phenomena. This issue is closely linked to unintended cation crossover from the anolyte, due to membrane imperfections in permselectivity. To study this failure mode in detail, we developed a method to control the cation concentration at the cathode and investigated its effect on electrolyzer performance. We found that maintaining an optimal alkali cation concentration at the cathode GDE is essential for high electrolyzer performance. Below this optimal concentration, catalyst activity decreases, while excessively high cation concentrations activate the carbon gas diffusion layer towards hydrogen evolution. In extreme cases, this can block the CO_2 pathway to the catalyst, leading to cell failure.

Our research demonstrated that during continuous operation, the cation flux from the anolyte to the cathode can vary, resulting in fluctuating cation concentrations at the cathode. To stabilize electrolyzer performance, this effect can be counteracted by actively controlling the anolyte composition. Our method developed for active anolyte control is based on continuous monitoring of electrolyzer parameters, which allows characterization of the cells' state-of-health.

Impact of Different Electrode and Process Parameters on the Performance of a Hybrid Alkaline Water Electrolysis Setup Utilizing Nickel-Based Gas Diffusion Electrodes

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The use of green hydrogen can significantly lessen the environmental impact of processes like methanol production, ammonia synthesis or steel production. This shift could boost demand for green hydrogen across many industries. Presently, alkaline water electrolysis (AEL) is the most mature technology for hydrogen production. A novel hybrid cell design by Koj et al. [1] addresses a key AEL issue: cross-contamination of gases resulting from electrolyte remixing. This design employs nickel-based gas diffusion electrodes (GDEs), eliminating the need for anolyte and thereby avoiding remixing. Along with this, the design is simpler and more compact compared to standard AEL. The development of these GDEs builds on the work of Kaiser et al. [2]. They are manufactured by spray-coating a nickel mesh with a aqueous suspension containing all catalyst components. Previous studies explored various electrode parameters through physiochemical and electrochemical characterisation methods in a half-cell setup.

This research extends the analysis to a full cell setup, focusing on hydrogen cross-contamination assessed through online gas chromatography and overall performance measured via various electrochemical methods. Variations in electrode materials such as PTFE and iron content as well as electrode thickness are investigated. Furthermore, effects such as contact pressure, differential pressure, and operating temperature are studied. Results (figure 1) highlight the dependency hydrogen crossover from PTFE content. The final objective is minimizing the hydrogen crossover while maintaining a high level in performance. New findings from Barros et al. [3] reveal that a reduction in hydrogen crossover and slight improvement in performance can also be achieved by implementing a “minimal” gap to decrease supersaturation effects. The hybrid electrolysis setup, due to its unique GDE arrangement, may also benefit from these insights.

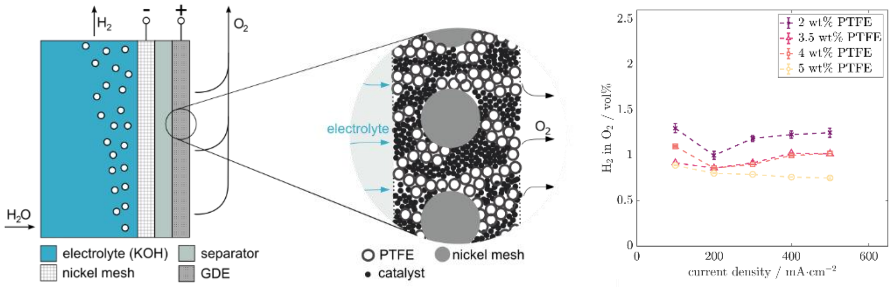

Figure 1: *a) Hybrid alkaline water electrolysis setup with nickel mesh cathode and GDE as anode. The electrodes in zero gap arrangement are separated by a porous Zirfon diaphragm. b) Hydrogen crossover measurements investigating GDEs with varying amounts of PTFE content.*

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Online Electrochemical Mass Spectrometry (OLEMS) for Screening of Gas Diffusion Electrodes for the Electrochemical CO_2 Reduction Reaction.

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The electrochemical reduction of CO_2 (CO_2RR) is important for the production of valuable chemicals employing sustainable energy and to ultimately lower the carbon footprint of chemical processes. However, the elucidation of catalysts, their selectivity, or stability is often limited by the slow throughput and low-sensitivity of conventionally used analytical methods such as e.g. online-coupled gas chromatography, HPLC or NMR spectroscopy. To address these challenges, differential electrochemical mass spectrometry (DEMS) and online electrochemical mass spectrometry (OLEMS) setups were developed, enabling the rapid and precise detection of major CO_2RR products, including both gaseous and volatile liquid species.[1]

In this work, we designed an OLEMS setup with an optimized 3D printed cell design that enables the detection of major gaseous CO_2RR products sequentially from multiple gas diffusion electrodes to screen features such as e.g. optimized catalyst composition, catalyst loading, variations in hydrophobicity, among others. The 3D printed cell architecture optimizes the hydrodynamic properties to minimize mass transfer limitations. This setup significantly accelerates CO_2RR catalyst evaluation by reducing experimental times and enhancing sensitivity, paving the way to more comprehensive and efficient studies of electrocatalyst performance.

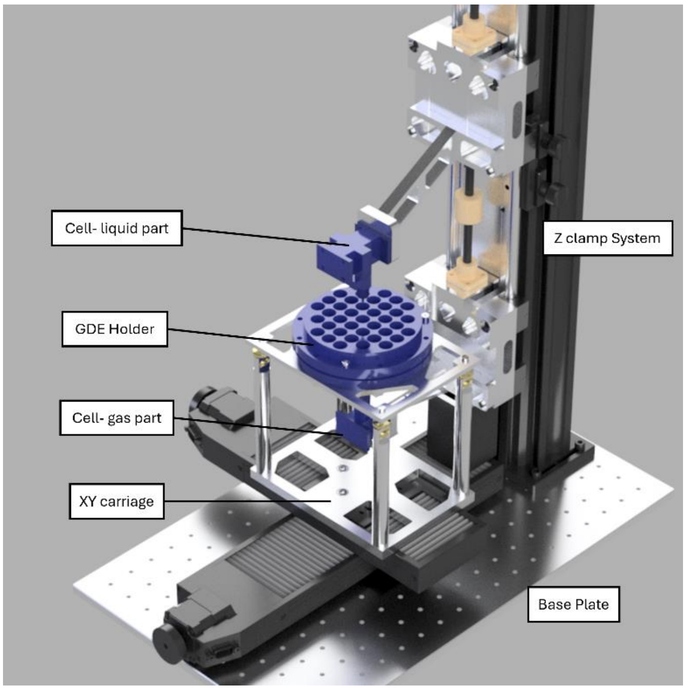

Figure 5. Rendered image of the designed GDE and cell positioning system for OLEMS measurements.

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Utilization of a gas diffusion electrode half-cell setup for anion exchange membrane water electrolysis and electrochemical ammonia synthesis

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Gas diffusion electrodes (GDE), which can be described as porous transport electrodes (PTE) in the terminology of water electrolysis, are central components in energy conversion and electrosynthesis technologies. Their composition of a catalyst layer on a porous substrate allow investigations with enhanced mass transport and high catalyst surface areas as well as application of industrial-relevant experimental parameters. Thus, the GDE approach was established in recent years as valuable bridging technology between fundamental electrochemical studies (e.g. rotating disk electrodes (RDE)) and more complex single cell measurements of membrane electrode assemblies (MEA).

In this contribution, we will highlight the application of a commercial GDE half-cell setup (FlexCell®, Gaskatel) in two research fields. Anion exchange membrane water electrolysis (AEMWE) combines the advantages of alkaline and proton exchange membrane water electrolysis by utilization of non-PGM catalysts and PFAS-free AEMs and ionomers. Investigations of the oxygen evolution reaction (OER) of AEMWE catalyst (layers) are currently mainly performed on RDE or MEA scale. Adoption of cell design and experimental parameters regarding electrode sizes, electrolyte flow rates and temperature enabled reliable testing in GDE half-cells under partially industrial-relevant conditions of up to 60 °C and 1 A cm^{-2} in 1 M KOH electrolyte, while at 80°C stability issues of the catalyst layer occurred (Figure 1). This novel testing procedure allows fast and early-stage optimization of catalyst layer structure and composition, while the transferability and comparability with RDE and MEA data are currently investigated.

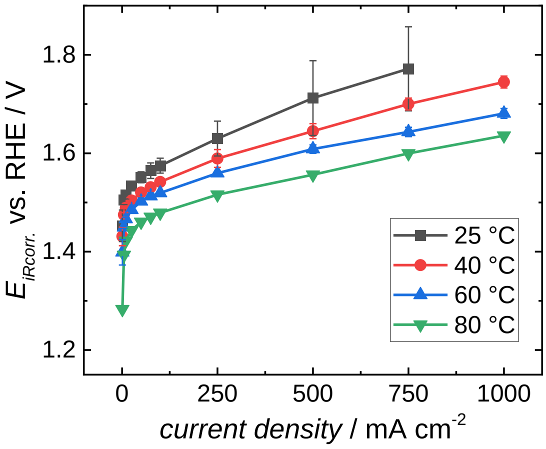

Figure 1. *OER polarization curves of $NiFe_2O_4$ (~ 3 mg cm^{-2}) in 1 M KOH with a flow rate of 60 mL min^{-1} without hot-pressed AEM membrane.*

On the other hand, GDEs based on transition metal nitride catalysts are investigated for the nitrogen reduction reaction (NRR) for the electrochemical ammonia synthesis. Among various transition metals, zirconium (oxy)nitrides were depicted as promising candidate, where the GDE half-cell setup overcomes limited N_2 solubility in aqueous electrolyte. Equipped with mass flow controller, recirculation unit and gas purifier, reliable NRR measurements can be performed. ZrN-based GDEs showed rather effects of contaminations and catalyst deactivation, while ZrN films deposited by chemical vapor deposition (CVD) hints towards some activity which has to be verified by isotope-labelled experiments.

Application of sample trapping gas chromatography for following the immediate electrolyser response to current density changes in electrochemical CO_2 reduction

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As renewable energy sources are well known for their weather and season induced fluctuation, dynamic operation is a typical development goal for Power-to-X technologies such as electrochemical CO_2 reduction (eCO2R), so they can double as grid stability services. Such load changes, however, may accelerate degradation and alter performance. It is therefore paramount to not only test devices using dynamic profiles and observing the effect, but to study the immediate response to such changes enabling knowledge-based development and on operation diagnosis.

The final step of CO_2 transport in a gas diffusion electrode (GDE) occurs though a thin wetting layer on the catalyst [1] requiring the electrode to form and maintain an equilibrium state to utilize the available catalyst surface without flooding the electrode. We were able to follow the products gas composition during this activation period and to combine the data with electrochemical data while varying catalyst layers and electrolytes [2].

We implemented a specialized gas chromatography method buffering 15 injections in fast succession before batch analysis using trapping selector valves [1] enabling us to analyze the product gas composition every 15 seconds over a 5-minute interval without giving up any accuracy or stability [2]. The data was compared and combined with online mass spectrometry data [4,5].

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Impurities in KOH and Their Impact on AEMWE: Evaluating Purification Protocols

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With the global push toward renewable energy, alkaline exchange membrane water electrolysis (AEMWE) stands out as a next-generation solution for green hydrogen production, combining the advantages of both alkaline water electrolysis (AWE) and proton exchange membrane (PEM) technologies.

While extensive efforts have focused on developing efficient catalysts and membranes, less attention has been paid to the quality of the electrolyte used – especially when working with noble-metal based electrocatalysts. Until now, only the impact of Fe impurities present in KOH has been widely discussed, mainly in the context of Ni-based electrodes used as electrocatalyst for the oxygen evolution reaction (OER) and hydrogen evolution reaction (HER) [1]. However, other cationic species present in the electrolyte may also interact with the catalyst surfaces or the anion exchange membrane, altering electrochemical performance. On the cathode, these species may adsorb onto active sites or promote side reactions; on the anode, residual metal ions can affect both catalyst activity and membrane stability [2]. These interactions highlight the importance of understanding and controlling the electrolyte composition.

In this study, we evaluate the effectiveness of several KOH purification protocols, already reported in the literature, in removing metal cations and how their removal impacts the AEMWE performance using non-noble metal catalysts. The tested methods are: (i) prolonged electrolysis of the electrolyte over Ni electrode [3]; (ii) contact of the electrolyte with $Ni(NO_3)_2$ solution followed by filtration [4]; (iii) prolonged contact of the electrolyte with Chelex® 100 resin [5]; and (iv) a fast protocol in which the electrolyte is passed over a Chelex®-packed column [6]. Inductively coupled plasma mass spectrometry (ICP-MS) was used to quantify the cations present in the electrolyte before and after purification. The AEMWE experiments using the purified electrolytes were performed in a 5 cm^2 electrolyzer, using Raney Ni and a PTL stainless-steel as cathode and anode, respectively, and a Sustainion X37-50 membrane as the alkaline exchange membrane.

In this communication, a correlation between the purification method, the electrolyte composition, and the stability of AEMWE will be presented. The different stabilities observed during AEMWE operated in different electrolytes was explained using Electrochemical Impedance Spectroscopy (EIS) and post-mortem cross-sectional analysis of the membrane and electrode layers.

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Electrochemical Regeneration of 1,4-NADH: The Role of Copper Electrode Morphology in Selectivity and Efficiency

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Bioelectrochemical systems are gaining increasing attention for their potential to revolutionize sustainable industrial processes by coupling enzymatic activity with electrochemical redox reactions [1]. A critical challenge in these systems is the reliance on expensive cofactors, such as nicotinamide adenine dinucleotide (1,4-NADH), which are consumed upon reduction, necessitating costly continuous supplementation [2]. To address this, researchers have explored in situ regeneration methods, with direct electrochemical regeneration emerging as a promising solution. However, key challenges remain, including poor selectivity and high overpotential, which depend heavily on catalyst properties and operating conditions. While prior studies have focused on catalyst material selection and reaction parameters, the structural and morphological characteristics of the electrode play a crucial yet underexplored role in enhancing efficiency and product selectivity.

In this work, we investigate copper-based electrodes, an environmentally and cost-effective material, for the electrochemical regeneration of 1,4-NADH. We developed a custom cell for the electrochemical deposition of copper on copper substrates, producing catalysts with varied grain sizes and morphologies. These engineered electrodes are systematically evaluated for their impact on 1,4-NADH regeneration efficiency and selectivity. Our results demonstrate that tailored copper electrode structures significantly influence reaction performance, with optimized conditions achieving high selectivity toward 1,4-NADH. Furthermore, the non-toxic and scalable nature of copper underscores its potential for sustainable electrochemical biocatalysis. This study highlights the critical role of electrode morphology in optimizing cofactor regeneration, offering a pathway toward more efficient and eco-friendly bioelectrochemical systems.

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Mitigating Phosphoric Acid Leaching in High-Temperature Proton Exchange Membrane Fuel Cells via Microporous Layer Modification of the Gas Diffusion Layer

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Phosphoric acid leaching is a major challenge in high-temperature proton exchange membrane fuel cells (HT-PEMFCs), where the acid electrolyte gradually escapes during operation [1]. Acid loss can proceed steadily through evaporation or accelerate exponentially via water replacement at high water activity levels [2]. As leaching progresses, membrane proton conductivity declines, reducing efficiency and accelerating fuel cell degradation [3]. Addressing this issue is critical for enhancing the durability and commercial viability of HT-PEMFCs. In HT-PEMFCs, microporous layers (MPLs) can serve as physical barriers to prevent liquid-phase phosphoric acid from migrating from the catalyst layer to the gas channels [4]. This study optimizes MPL configuration by varying PVDF binder content (0 – 20%) and loading (0 – 2 mg cm^{-2}) using Carbon Black XC-72, leveraging the layer's hydrophobic properties to improve acid retention and reduce leaching. Our evaluation shows that a 10% PVDF composition delivers the best performance, with an ohmic resistance increase rate of 57.2 $\mu\Omega \cdot cm^2\ h^{-1}$, as shown in Fig 1(a)—six times lower than that of commercial gas diffusion layers (GDLs). This optimal formulation also achieves a voltage degradation rate of 60 $\mu V\ h^{-1}$, as shown in Fig. 1(b), representing a fivefold improvement over conventional GDLs. These results highlight the critical role of MPL optimization in controlling acid leaching, thereby enhancing HT-PEMFC performance and extending operational lifespan.

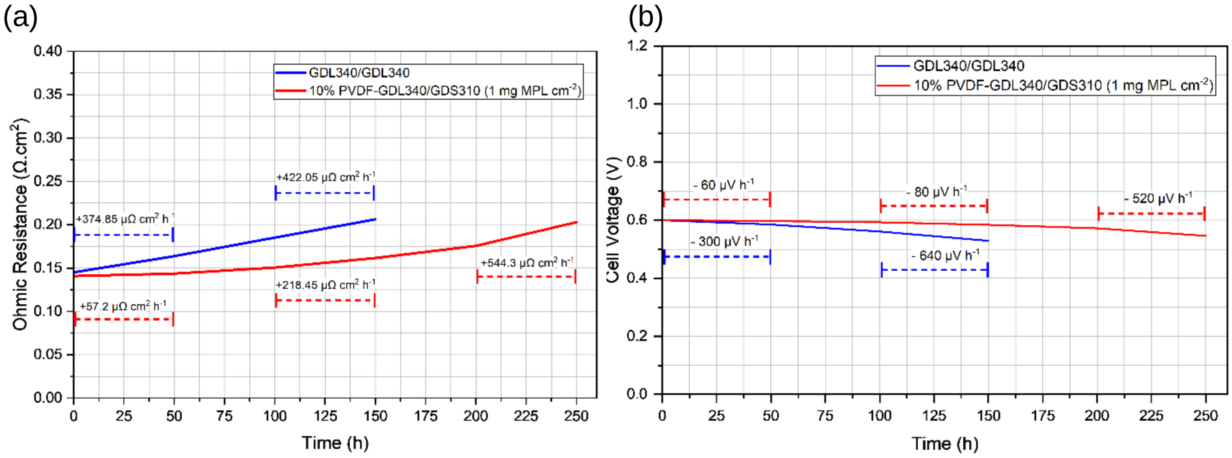

***Figure 1**: Comparison between commercial GDLs and modified GDLs: (a) Ohmic resistance; (b) Cell voltage.*

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Replacing per- and polyfluoroalkyl substances in high temperature proton exchange membrane fuel cell electrodes

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Polytetrafluoroethylene (PTFE) as one of the most prominent per- and polyfluoroalkyl substances (PFAS) is used in the high temperature proton exchange membrane fuel cells (HT-PEMFC). As a binder it supports water removal in the catalyst layer and the microporous layer in significant amounts of up to 40 wt%. In HT-PEMFCs, phosphoric acid is used as the proton conductor, and its wettability of the catalyst can be inhibited by too much PTFE.[1] The European Chemical Agency (ECHA) plans to restrict the use of PFAS in the European Union because of evidence of adverse health effects in humans and their persistence.[2] As of March 2025 ECHAs scientific committees for Risk Assessment (RAC) and for Socio-Economic Analysis (SEAC) are still evaluating the potential impacts of restricting PFAS in the energy sector[3].

The HT-PEM compatible polymers polyether ether ketone (PEEK) and poly(pentafluorostyrene) (PPFS) are being tested for their viability as a PTFE replacement. One main point of attention is the comparison of the hydrophobicity regarding the catalyst layer containing various amounts of one polymer to a catalyst layer containing PTFE. The values of the PTFE reference GDEs are shown in Figure 1, the trend shows a higher contact angle with higher PTFE loadings because of higher hydrophobicity.

Gas diffusion electrodes (GDE) containing the gas diffusion and microporous layer as well as the catalyst layer are fabricated using doctor blading. To test for proper coating the catalyst layer is validated with inductively coupled mass spectrometry (ICP-MS) for correct catalyst loading and with X-ray fluorescence spectroscopy (µ-XRF) to confirm a homogenous coating and distribution of the binder. Computed tomography (CT) measurements provide information about the layer height, the structure of the catalyst layer and the pores. To evaluate the electrochemical performance of the oxygen reduction reaction (ORR) of the GDE with alternative binders a three-electrode half-cell setup is used (Gaskatel Flexcell®, Gaskatel Hydroflex® reference electrode, platinum counter electrode 160 °C, concentrated phosphoric acid).

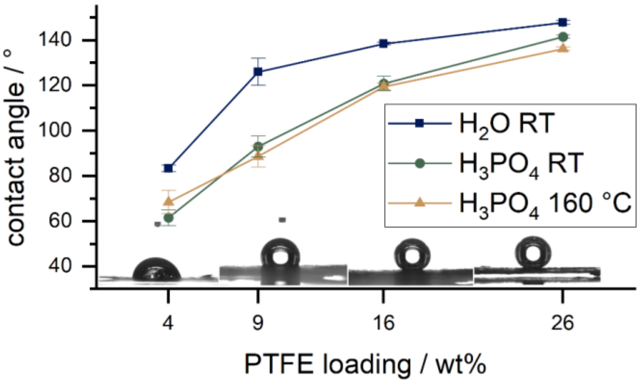

***Figure 1**: Contact angle measurements of GDEs with different PTFE loadings by using water, phosphoric acid and phosphoric acid at the temperature for electrochemical measurements (160 °C).*

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Modelling the Impact of Surface Chemistry on Catalyst Degradation in PEM Fuel Cells

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Modern proton exchange membrane fuel cells (PEMFCs) exhibit high power performance and low environmental impact. However, large-scale commercialization remains limited due to irreversible degradation processes, with the major portion of performance losses originating in the cathode catalyst layer (CCL). Disintegration or loss of catalyst particles, carbon substrate, and ionomer electrolyte compromise both efficiency and durability. Prior research [1,2] has underscored the impact of platinum dissolution on the CCL microstructure. Especially under dynamic conditions that are typical of automotive applications, cyclic oxidation and reduction of platinum significantly accelerate the dissolution. Accurate predictive degradation models must therefore account for the interplay of platinum dissolution and oxide formation. We expect our multiscale modeling framework, depicted in Fig.1, that we have developed and exploited in past contributions to be outfitted with predictive capabilities. The framework integrates particle-level degradation mechanisms, encompassing detachment, coagulation, and dissolution, with electrode-level models, including particle population balance and porous electrode theory. These components capture how microstructure evolves over time and influences transport behavior and local reaction kinetics. We present two extensions to the existing framework, shown as red boxes in Fig. 1, which relax assumptions or empirical correlations employed in previous works [3,4]. The first extension concerns the influence of ionomer coverage of the catalyst particles on particle dissolution and microstructural and materials properties, especially wettability and water transport properties of the CCL. The second extension entails a detailed microkinetic model for platinum oxidation and reduction to capture peculiar details of Pt dissolution during potential cycling [5]. Given the complexity and nonlinearity of the model and its extensions, we integrate machine learning surrogates into the framework. This hybrid approach enhances predictive accuracy and computational performance, while preserving physical interpretability, supporting real-time diagnostics and lifetime forecasting of PEMFCs under practical load profiles.

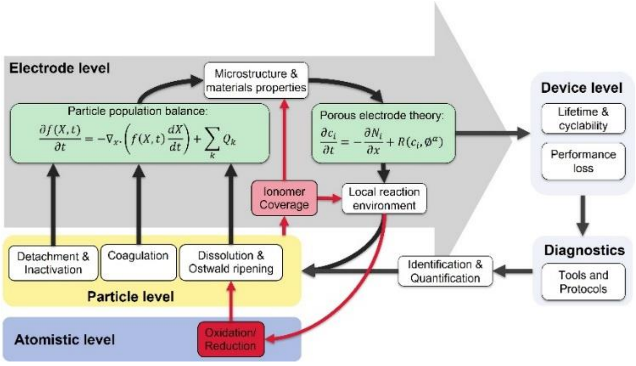

Figure 1: Hierarchical modelling framework.

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Fundamental Understanding of the Reversible Platinum Oxidation Formation of Catalyst-Coated Membranes for PEM Fuel Cells

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Long-term durability remains a significant challenge for PEM fuel cells and many of its underlying degradation mechanisms are still unclear. For instance, platinum oxide formation is closely linked to platinum dissolution, leading to a loss in electrochemically active surface area (ECSA). In last decades, simple Pt oxidation models were replaced by multiscale degradation models, which depict catalyst aging processes such as the formation and reduction of platinum oxides, platinum dissolution, particle growth due to Ostwald ripening, platinum ion transport through the ionomer and platinum band formation in the membrane.[1] However, there is still improvement necessary to include a multitude of factors like humidity, different voltages and holding times to reliably and accurately predict the degradation behaviour of catalyst materials using one model.

In this work, 5 cm^2 single cell experiments were carried out on a customized G60 PEM fuel cell test station (Greenlight Innovation Corp.). For the catalyst-coated membrane (CCM) preparation, ~2 nm platinum nanoparticles supported on high surface area carbon (TEC10E50E, Tanaka) with a loading of 0.1 $mgPt/cm^2$ was used on the cathode side. The anode catalyst material (TEC10V30E) and Pt loading (0.2 $mgPt/cm^2$) remained unchanged in all experiments. The experiments were carried out at 80 °C and 100% relative humidity under H_2/N_2 atmosphere. Each CCM measurement was repeated five times. CV and EIS methods were used to monitor the changes in the Pt oxide layer formation before, during and after the chronoamperometric measurements at different holding times.

The charge of the Pt oxide reduction peaks was correlated with the ECSA to determine the platinum oxide coverage. Based on experimental data, we evaluated the logarithmical growth of platinum oxide on the cathode catalyst layer and simulated this behaviour using different modelling approaches based on the works of Jahnke et al. [1], Darling and Meyers [2] and Redmond et al. [3]. Our results indicate a logarithmic correlation between holding times and platinum oxide coverage. But depending on the voltage, holding times up to 100.000 s can lead to a steady-state growth of the Pt-O coverage. Interestingly, only a coverage of ~0.6 (equivalent for 2 e- reactions) was estimated by holding the voltage at 0.85 V for 10,000 s. A decrease in humidity from 100% to 60% significantly reduces the Pt-O coverage up to 70%. Lowering the voltage from 0.95 V to 0.75 V also results in a reduction of Pt-O coverage, which can be predicted by the selected simulation models with different degrees of accuracy.

In summary, we offer key insights into the reversible oxidation formation mechanisms of platinum nanoparticles by combining experiments and simulations. This helps to identify the main key parameters for improving the long-term durability of PEM fuel cells.

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Bridging GDE Half-Cell and MEA Testing for Improved Electrode Characterization

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One factor slowing the development of polymer electrolyte fuel cells (PEFCs) as sustainable energy converters is the need for high-throughput testing for advanced fundamental understanding of the properties of membrane electrode assemblies (MEAs). The gas diffusion electrode (GDE) half-cell setup has gained attention as an intermediate method for catalyst (layer, CL) characterization for the oxygen reduction reaction [1]. It bridges the gap between basic-research oriented rotating disk electrode (RDE) testing and application-focused full-cell MEA analysis, combining advantages like fast, inexpensive catalyst screening (RDE) and technically relevant current densities up to 3 A cm^{-2} of realistic CLs (MEA) [2-4]. However, traditional GDE half-cell setups cannot assess the limitation of ion (proton) transport, like in a real PEFC, due to direct contact of electrode (= CL) and liquid electrolyte.

This study therefore aims to take the GDE half-cell to the next level to better mimic realistic PEFC conditions by integrating a membrane between the CL and the electrolyte. Carbon paper was used as gas diffusion layer (GDL) and Nafion™ as membrane material. The catalyst layer composed of Pt on C as catalyst and various ionomers and was manufactured either on the GDL or the membrane by ultrasonic spray coating. The membrane was attached to the GDL by hot-pressing. Tests were performed in a commercial test cell (Flexcell® PTFE, Gaskatel GmbH).

Adding the membrane to the GDE half-cell system changes mass transport properties of the CL completely: protons have to travel through the membrane and water cannot diffuse in the electrolyte but must be released through the gas diffusion layer. Key challenges like membrane swelling and detachment at high currents due to water production were resolved by optimizing pressing and handling parameters as well as careful choice of GDL. To gain a better understanding of the advanced setup, electrochemical impedance spectroscopy with an adapted equivalent circuit derived from distribution of relaxation times analysis was employed. Results under various test conditions show that two out of four capacitive processes, which are ongoing, plus an additional ohmic resistance can be attributed to the test cell and conditions. Therefore, they are summed up as total cell resistance which is used for iR-correction.

We have successfully demonstrated a proof of concept for integrating a membrane into a traditional GDE half-cell setup. This advanced setup perfectly mimics the cathode of a PEFC while still being a considerably faster and more reliable tool for electrode testing.

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From Active Material to Functional Gas Diffusion Electrodes: Comparative Assessment for Alkaline and Near-neutral Zn-Air Batteries

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The state-of-the-art rechargeable Zn-Air battery (RZAB) technology mainly focuses on the use of strongly alkaline electrolytes (mainly KOH-based).[1,2] However, carbonation of the electrolyte, carbon corrosion, clogging and/or flooding of the gas diffusion electrodes (GDE), as well as restructuring/dissolution of the catalyst(s), result in the failure of the oxygen reduction reaction (ORR) catalytic function for the discharge of the RZAB.[1-4] To overcome some of these limitations impeding the widespread adoption of RZAB, switching to a near-neutral electrolyte is appealing. However, due to the different properties of the electrolyte (e.g., pH, viscosity, etc.), not only the ORR rate itself is strongly influenced, but also the wetting of the catalytic layer of the GDE is affected tremendously.

Within in this presentation, we will at the beginning mainly report on a screening of catalyst materials and electrolyte compositions under well-defined and controllable conditions by means of rotating disk electrode (RDE) investigations. Hereby, a comparison between alkaline and near-neutral electrolytes will be discussed, revealing some trends on the catalytic materials activity. Then, we will depict the performance and endurance of GDEs embedding these catalyst materials, which were manufactured via a scalable calendering technique, when in contact with selected electrolytes. From comparing alkaline electrolyte with near-neutral electrolytes, we will discuss parameters and factors influencing GDEs activity, and their durability, together with results gained from RDE investigations. A special focus will be given on critical (and often neglected) parameters governing the performance and endurance of near-neutral RZAB recorded experimentally.)

Figure 1. *Schematic illustration from **(left)** the catalyst characterization via RDE-measurement (0.5 M KOH), over **(middle)** the cross-section of a GDE and **(right)** subsequent cycling of the GDE in a near-neutral electrolyte (1Z5K20TMP).*

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Optimization of the wettability of gas diffusion electrodes for zinc-air batteries (ZAB) by exploring new manufacturing methods

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For an electricity supply with high degree of renewable energy share, mid- to long-term storage technology is needed to bridge days and weeks with low supply. These kind of storage systems don't need a high-power output, but have to store the electrical energy as cheap and efficient as possible to be economically competitive. Existing technologies don't match the needed cost profile, i.e. costs per stored energy and costs per power [1]. New technologies with different cost profiles are therefore needed.

One promising technology is the class of Metal-Air-Batteries. Especially Zinc-Air-Batteries (ZAB) are a good candidate as they are safe and based on abundant and cheap materials. The main issues that needs to be solved for the realization of a rechargeable ZAB are the loss of structure in the zinc-electrode during cycling, carbonate precipitation due to CO_2 absorption and its low round-trip-efficiency around 60% caused by sluggish oxygen reactions.

Besides an improved catalyst for the oxygen reactions, the bifunctional gas diffusion electrodes for use in a ZAB must be optimized for the different demands on the transport processes of the oxygen reduction reaction and the oxygen evolution reaction. Wettability plays a critical role here.

Typically, wettability of metal-based gas diffusion electrodes can't be adjusted without changing the stability of the electrode as the binder PTFE also is responsible for the hydrophobic parts of the electrode. Decoupling stability from wettability therefore gives more freedom to finetune the wettability. In this work we present several ways to first produce a pure metal GDE and then adjust the wettability by adding a hydrophobic coating for an optimal performance.

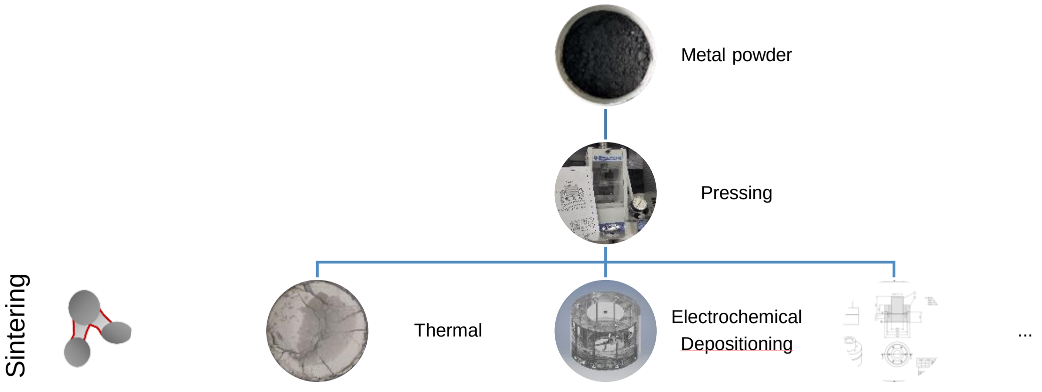

***Figure 1:** Different possibilities to sinter µ-porous metal electrodes*.

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Advanced α-MnO_2 Electrodes for Enhanced Oxygen Reduction in Aluminum-Air Batteries

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The transition to sustainable energy sources is rapidly advancing. Still, the intermittent nature of solar and wind energy requires efficient and large energy storage systems to bridge periods of low electricity generation. One promising technology is the aluminum-air (Al-air) battery, which offers several advantages, such as the abundance of aluminum, safety, low cost, and the ease of storing and transporting aluminum. This primary battery works similarly to a fuel cell, requiring aluminum to be replaced after use. However, aluminum products can be efficiently extracted from the electrolyte through seed-mediated precipitation, and aluminum production can occur externally via the established inert-electrode smelting process, ensuring a complete cycle [1].

A key challenge for Al-air batteries is improving their performance and efficiency. This study focuses on optimizing the cathode, where the oxygen reduction reaction (ORR) occurs [2,3]. In particular, we investigate α-MnO_2 as a catalyst for the ORR, exploiting its well-established catalytic properties. However, α-MnO_2 suffers from low electronic conductivity, which limits its performance. To address this issue, we examine a series of α-MnO_2-to-Vulcan ratios as catalyst layers incorporated into gas diffusion electrodes (GDEs). The GDEs are evaluated in a novel GDE half-cell setup, demonstrating that the addition of conductive Vulcan significantly enhances ORR activity, allowing the catalyst to reach its full intrinsic potential. The optimized electrodes are also tested in an Al-air demonstrator cell and exhibit performance close to conventional Pt catalysts.

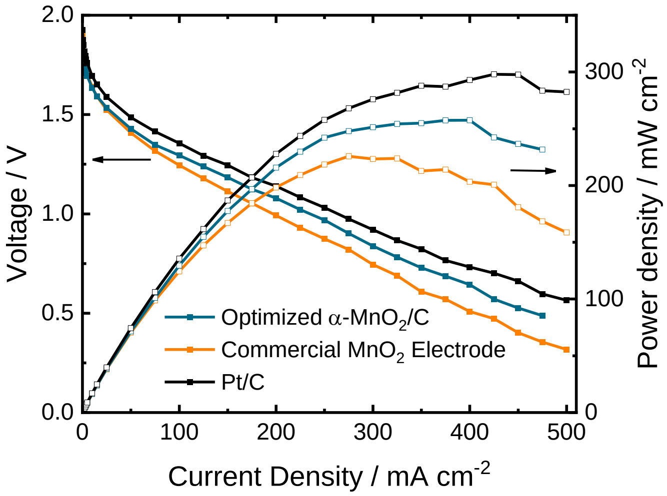

Figure 1: *Polarization and power density curves of Al-air demonstrator cell using different cathodes.*

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All-Carbon Gas Diffusion Electrodes for the Cathodic Electrosynthesis of Hydrogen Peroxide. Results from the Project Power2HyPe

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Hydrogen peroxide (H_2O_2) is a very important chemical (oxidant) used across a wide spectrum of industries ranging from disinfection, pulp bleaching to propellants. The anthrachinone process which is well established in industrial peroxide production for decades is characterized by a large carbon footprint caused by a large energy demand. *Power2HyPe* (**www.power2hype.eu**) is a collaborative research project dedicated to an industrial-scale demonstration of an electrochemical synthesis route enabling energy savings of around 30% per kg hydrogen peroxide. The cathodic, 2-electron reduction process at the cathode, which is a core element in the Power2HyPe process, is envisaged to be operated using gas diffusion electrodes which only employ low-cost, PGM-free catalyst. A systematic study has been carried out using gas diffusion electrodes containing carbon catalysts with engineered surface chemistry (carbon nanofibers CNFs, graphene nanoplatelets GNPs) as well as carbon black materials from industrially established grades.

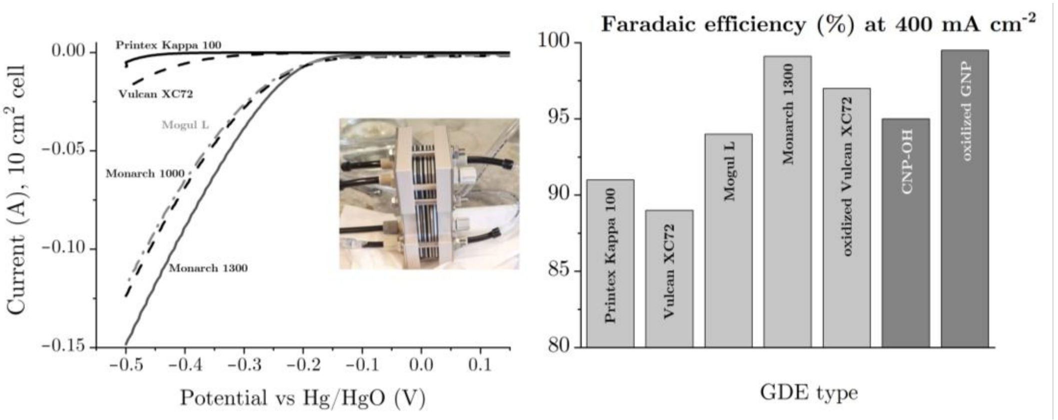

Figure 1. *Performance of different all-carbon GDEs in flow cells: Linear scan voltammograms vs Hg/HgO (left) and faradaic efficiency (H_2O_2 selectivity) at 400 mA/cm^2 (right).*

Summary

Flow cell measurements have demonstrated that the cathodic electrosynthesis of peroxide in alkaline solutions can be operated at industrially relevant current densities of 400 mA cm^{-2} and higher with a stable faradaic efficiency of more than 95% (prolonged test are underway) using low cost, all-carbon gas diffusion electrodes.

Acknowledgement

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Poster Contributions

Towards a Model Gas Diffusion Electrode for Electrochemical CO_2 Reduction

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Global reliance on fossil fuels for transportation and energy generation continues to contribute to climate change. Furthermore, as fossil fuels are finite resources, the need for renewable fuel and energy alternatives is becoming increasingly important. Electrochemical reactions offer a pathway to harness renewable energy sources such as solar, wind, and hydro for producing sustainable chemicals and fuels.[1] For example, CO_2 can be converted into a variety of value-added chemicals and fuels, including methane (CH_4), the primary component of natural gas, and ethylene (C_2H_4), a critical precursor for polyethylene, one of the most pervasive plastics in the electrochemical CO_2 reduction reaction (eCO_2RR).[2]

Despite extensive study since the 1980s, the commercialisation of the reaction remains limited by poor performance (selectivity and activity). Gas diffusion electrodes (GDEs) address some of these issues by delivering CO_2 gas directly to the catalyst, overcoming the solubility and diffusion limitations of traditional aqueous-phase cells and enabling operation at higher, commercially relevant activities (current densities), though selectivity still remains a problem.[3] Aqueous phase H-cells are still commonly used for screening new catalysts due to the complexity of GDEs with multiple length scales at different interfaces where phenomena occur during the reaction.[4] For an electrochemical reaction to take place on a GDE, the relevant species (in the case of eCO_2RR: CO_2, e^- and H^+) must all meet at the triple phase boundary, i.e. where the electrocatalyst, gas and electrolyte meet. Therefore, porosity, pore structure, pore path length and wettability must all play a role in the reaction and thus govern electrode performance, i.e. how the gas, electrolyte and electrocatalyst meet. Such optimisation of these factors is difficult to achieve in traditional carbon paper-based GDEs due to the random distribution of the carbon fibres.

In our research, we employ surface laser texturing to create a model GDE with periodically patterned and well-defined pores, thus simplifying the triple phase boundary. We compare the performance of these model GDEs electrodes by varying the pore size, spacing and path length. By studying these model GDEs, we gain insights into optimising electrodes for the commercialisation of ECO_2RR.

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Development of PFAS-free silver-based Gas Diffusion Electrodes for electrochemical CO_2 reduction

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The chemical industry is responsible for a significant percentage of global greenhouse gas emissions, as fossil feedstock still plays a central role in most production processes [1]. A promising solution to reduce these emissions is the electrochemical reduction of CO_2, which allows for the production of key platform chemicals such as carbon monoxide (CO), ethylene or formic acid from CO_2, water, and renewable electricity. Nevertheless, there are still technical challenges that need to be solved before commercial implementation of the technology, especially regarding efficiency and the long-term stability of the electrolysis components. One of the critical elements in the electrolysis is the porous gas diffusion electrode (GDE). GDEs are commonly rendered hydrophobic by using polytetrafluoroethylene (PTFE) as the binder. The hydrophobicity is essential to maintain an extended reaction zone within the electrode, as it prevents the electrolyte from blocking the pores and ensures efficient gas transport to the catalyst [2]. However, due to the negative environmental impact of fluorinated compounds and a possible ban in the near future, there is a growing need to substitute PTFE with fluorine-free binders that provide both mechanical stability and adequate hydrophobicity to prevent liquid electrolyte breakthrough.

This work focuses on the application and optimization of the GDE manufacturing processes for alternative binder systems in silver-based GDEs for the production of CO. For that purpose, different methods and parameters are investigated to find suitable processing conditions for the novel binders and obtain mechanically stable electrodes with suitable performance in terms of achievable current density and durability during long term electrolysis.

The performance of the GDEs is analyzed in a flow-through electrolysis setup under alkaline conditions with the target product CO. Physical characterization and advanced morphology studies are performed on pristine and used GDEs. Besides developing more sustainable electrodes, the goal of the investigation is to reveal the interplay between manufacturing processes and electrode properties, providing a deeper understanding of their impact on overall performance and electrochemical behavior during electrolysis.

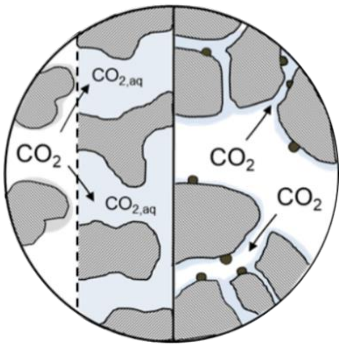

Figure 6: *Schematic illustration of the wetting behavior within the GDE with (right) and without (left) hydrophobic properties.*

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CO_2 Electrolysis Using Silver Gas Diffusion Electrode in Zero-Gap MEA Configuration

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The electrochemical reduction of CO_2 to CO in gas-fed membrane electrode assembly (MEA) systems offers a promising pathway for sustainable chemical production. Achieving high current densities with prolonged operational stability remains a critical challenge, primarily due to cathode flooding and electrolyte management issues. Here, we demonstrate a zero-gap MEA CO_2 electrolyzer employing silver-coated carbon paper gas diffusion electrodes (GDEs) and CsOH as the electrolyte. Our system achieves a CO Faradaic efficiency (FE_CO) of ~85% at a current density of 200 mA cm^{-2}, sustained over 100 hours of continuous operation. The use of CsOH as an anolyte enhances carbonate ion conductivity and lowers ohmic resistance, enabling efficient charge transport and high current densities. Cesium cations modulate the electric double layer, suppressing HER and promoting CO_2 reduction, which supports high Faradaic efficiency and stable long-term operation without performance degradation.[1] Furthermore, the silver coating provides robust catalytic performance and mitigates catalyst degradation under high cathodic overpotentials.[2, 3] Operando studies highlight the importance of water and cation management across the membrane to prevent gas diffusion layer (GDL) flooding and ensure stable CO_2 mass transport.[4] This work underscores the potential of integrating Ag-based GDEs with CsOH electrolyte in scalable zero-gap configurations for durable and efficient CO_2-to-CO conversion. This research has received funding from the European Research Executive Agency (REA) under the powers delegated by the European Commission, MSCA European Postdoctoral fellowships grant agreement No. 101106114, Project FOTOCER.

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Understanding the Influence of Cations in Zero-Gap CO(2) Electrolyzers

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The development of efficient electrochemical CO_2 reduction is of interest as a strategy to reduce rising atmospheric CO_2 levels and promote a circular carbon economy and valuable products. CO_2 electroreduction (CO2RR) in zero-gap membrane electrode assemblies (MEAs) allows industrially relevant reaction rates, making it a scalable approach. Achieving the desired high and stable rates of C2+ product formation in those cell configurations remains a challenge due to failure mechanisms like gas diffusion electrode (GDE) flooding and salt formation caused by cation crossover. [1],[2],[3] Since electrolyte ions have also been shown to be crucial in enabling certain desired reaction pathways, a deeper understanding of ion movement and the resulting implications is needed in order to balance the beneficial and detrimental effects.

The cation effect, which is known to enhance C2+ product formation with increasing cation radius and with increasing concentration, has been widely studied in CO2RR. However, its influence in the carbon monoxide reduction reaction (CORR), particularly with different cations and anolyte concentrations in zero-gap electrolyzers, remains less explored. In a previous study we highlighted that AEM are not 100% permselective and unintended cation crossover can occur and alter the selectivity based on the anolyte concentration used.[1] This study further examines these effects, highlighting how electrolyte composition impacts CO(2)R selectivity.
In addition, we introduce an adapted zero-gap MEA cell designed for practical CO(2) reduction and operando studies. This setup allows real-time analysis of CO(2)RR products while monitoring structural and chemical changes during electrocatalysis which can help us better understand the CO(2)RR mechanism. Our findings reveal the role of electrolyte ions and concentrations, including unintended cation crossover through anion exchange membranes, affecting selectivity and stability. This approach addresses limitations of conventional in situ and operando methods, which often rely on specially designed cell that differ from those used in standard lab operations.

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From ants to plants: Upscaling electrochemical CO_2-to-formate/formic acid production and the implications on relevant conditions

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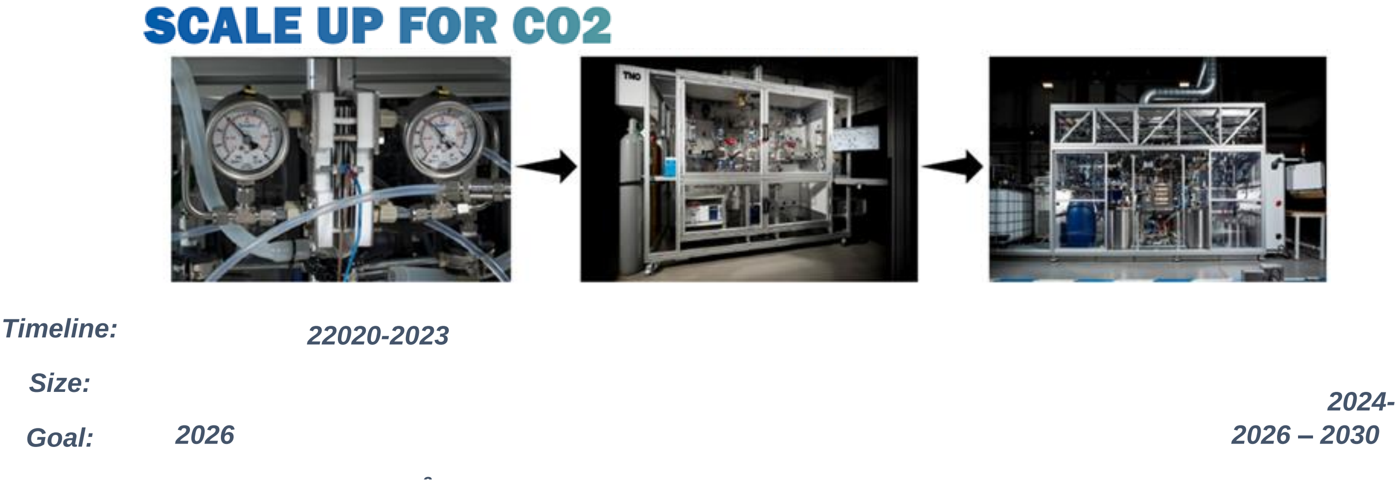
The potential of low-temperature electrolysis for direct conversion of CO_2 into fuels and commodities (CO, formate, alcohols, C_2+) has been demonstrated at laboratory scale. However, despite a fast-growing research field and a wide industrial interest, CO_2 electrolysis is still at TRL~3-5, and several fundamental knowledge gaps still remain at the level of a single electrochemical cell, as well as unknowns on reactor and process design. At TNO-SPES we aim to identify upscaling hurdles in an early stage, and therefore contribute to advance the CO_2 electrolysis technology towards industrial implementation in the near future. Integrating R&D at all levels (electrodes>cell> reactor>process>system) is urgently needed to bring the technology towards industrial implementation. In particular, the electrochemical conversion of CO_2 to formic acid over a gas diffusion electrode (GDE) can be challenging due to the complex interplay of various parameters.

During academic research favorable conditions are often chosen in order to optimize the activity, selectivity and stability of the reaction. Various studies in which parameter screening gives greater insight into the working mechanisms are beneficial for theoretical understanding, however, practical conditions leading from technoeconomic evaluations imply that certain criteria are more restricted. One specific example here is the alkalinity of the system that leads from the need to have viable conversion rates. Regardless of the chosen electrolyte, the local reaction environment around the cathode becomes more alkaline with increasing current densities, and implies the need for stable catalyst materials under such conditions. In addition, the desired conversion from CO_2 to formic acid (HCOOH) becomes restricted by the pH exceeding the formic acid/formate pKa of 3.75, leading to formate production instead. Alternatively a 3-compartment system can be deployed in which the so-called 'middlelyte' can be used to regenerate the formic acid directly.

During this lecture we will explore the pros and cons of the 2- and 3-compartment systems for CO_2 reduction systems , discuss the influence of the gas diffusion electrode and assess how academia and industry can guide one another in testing relevant electrochemical parameters.



Process development for the Electrochemical CO_2 Reduction to Formate in Bio-compatible electrolytes

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The high CO_2 concentration in the atmosphere and its adverse impacts on global climate require innovative solutions to mitigate climate change effects. Electrochemical CO_2 reduction (eCO_2R) is a promising pathway for converting CO_2 into value-added compounds at mild temperatures and pressures. Among the products from eCO_2R, formate can be produced at high yields at high current densities using Sn-, In-, and Bi-based catalysts[1]. This study investigates catalyst materials, electrolyte composition, and temperature for CO_2 conversion to formate in biologically compatible conditions. The electrolyte used for the electrochemical CO_2 reduction reaction was optimized for maximum formate production and direct transfer to a bio-reactor. CO_2 electrolysis was conducted in a phosphate-based electrolyte in a flow cell setup[2]. Investigation of In, Sn, and Bi-based catalyst materials revealed the highest formate production with indium oxide catalyst. The development of a compatible electrolyte included the optimization of buffer concentration and temperature. We observed a counter-intuitive decline in formate production upon increasing the concentration of phosphate in the buffer. Varying the temperature also showed decreased efficiency upon increasing the temperature from 30 to 40 °C. The optimized electrolyte and temperature conditions led to formate concentrations up to 1.81 mol L^{-1}. Furthermore, we evaluated catalyst stability in a zero-gap double-membrane CO_2 electrolyzer configuration and the effect of anion exchange membrane. We observed a similar performance between Sustainion and PiperION AEMs, yielding pure formic acid solution with 250 mmol L^{-1} and FE of 80 %, but decreased efficiency over time when FUMASEP AEM was employed. Finally, the application of electrochemically derived formate was demonstrated using *Methylorubrum extorquens* TK001 strain cultivated in fed-batch process [2]. This work demonstrates process development for electrochemical CO2 reduction reaction in different cell configurations and how tunning process conditions can satisfy different applications. The integrated electro-biochemical reaction cascade highlights the potential of using CO_2-based formate as a sustainable carbon source for microbial synthesis.

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Multilayer GDEs for long-term stable acidic CO_2 reduction to formic acid

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Employing green electricity and shifting from fossil to renewable feedstock to reduce anthropogenic CO_2 emissions is a sustainable method contributing to cracking down the challenges for achieving greenhouse gas neutrality. Recently, electrochemical reduction methods using gas diffusion electrodes (GDEs) have become a viable approach for converting CO_2 from sources like waste incineration, cement industry, or biogenic sources to valuable platform chemicals like CO, ethylene and formic acid. Previous research from our group has demonstrated the feasibility of bismuth (Bi) based GDEs in acidic CO_2 electrolysis to produce formic acid which comes with the advantage of a significantly simplified upstream processing and reduced carbonate precipitation within the electrode. However, the process still faces severe challenges associated with long-term stability and the accumulation of product which is essential for reaching a commercially competitive process. The reason for the negative impact of increasing formic acid concentration in acidic environment is still not fully understood [1].

Recent studies have shown that systematic GDE engineering and applying a protective layer (PL) for the catalyst can improve stability by regulating the ion transport and shield the catalyst from harsh conditions. Meanwhile, the hydrophobic gas diffusion layer prevents electrode flooding, ensuring continuous CO_2 diffusion to the catalyst layer during operation [2]. The presented work aims at the fabrication and optimization of such multilayer substrates for GDEs for formic acid production starting from the already established single layer architecture which reaches current densities of several 100 mA/cm² but lacks sufficient long-term stability. Based on this, the beneficial effect of electrodeposited Bismuth catalyst to produce a very thin and well-defined catalyst layer is demonstrated. Through optimization the electrode properties, we try to get a better understanding of how the electrode needs to be tailored in order to reach the required performance metrics for industrial operation with high durability. To this end, physical characterizations and advanced microstructural studies to understand the complex electrode framework before and after electrolysis complement the electrochemical characterization and give insights into electrode degradation.

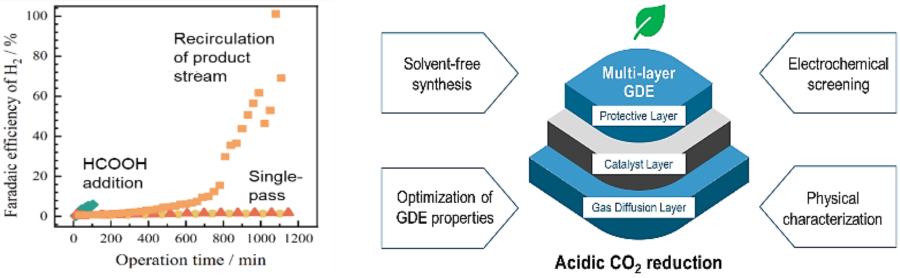

Figure 1: Stability of long-term acidic electrolysis at 200 mA/cm² during accumulation of formic acid [1] (left) Overview of concept and methodology for multi-layer GDE development (right)

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Innovation of oxygen-depolarized cathodes as alternative gas diffusion electrodes for CO_2 electrolysis for scalable CO_2 electrolysis.

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The oxygen depolarised cathode (ODC) is a very stable gas diffusion electrodes (GDEs) currently used to make multiple kilotonnes of NaOH and Cl_2.[1]. In the chlor-alkali electrochemical industry, various issues, also encountered in the development of stable CO_2 reduction GDEs, have already been tackled. [2,3]

We identify the similarities between chlor-alkali ODC electrodes and CO2R GDE and introduce an innovative approach by utilizing conventional ODC electrodes for CO_2 electrolysis in three-compartment flow cells, enabling CO production under industrially relevant conditions. Notably, the GDE shows a high CO selectivity at different current density during the initial electrolysis stage, shown in Figure 1. Even operating at 2.0 kA/m², the overall cell voltage remains within a moderate range.

Additionally, scalability of the process was evaluated by upscaling a single cell up to 100 cm². The system demonstrated a broad operational window capable of triggering CO production in both flow-through and flow-by modes. The CO concentration in the outlet gas reached approximately 20 mol.%, with a production rate of 11.4 g/hour, facilitating efficient separation. We have optimized the operation to maximize the CO production, including flow field design, pressure difference regulation, gas flow rate, and electrolyte flow rate.

Finally, we conducted stability experiments for up to 60 hours of accumulated batch testing to identify key parameters governing stable CO production in long-term electrolysis. This effort aims to advance robust CO_2 electrolysis technologies toward large-scale demonstration

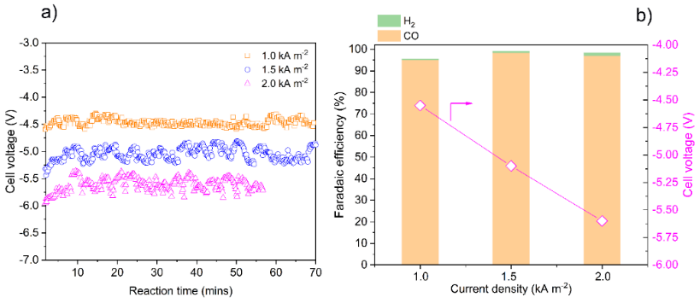

Figure 1 *Performance of oxygen depolarization cathode at different current density for CO_2 electrolysis: a) cell voltage over time, b) faradaic efficiency along with cell voltage. Reaction conditions: 0.5 M $KHCO_3$ in cathodic compartment and 1 M KOH in anodic compartment.*

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Advancement of gas diffusion electrodes for electrochemical CO_2 reduction: Transitioning from laboratory to industrial implementation

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Gas Diffusion Layers (GDLs) are essential components in electrochemical CO_2 reduction systems, ensuring efficient gas transport and playing a critical role in overall cell performance [1]. At Avantium (AVT), we traditionally relied on a wet carbon dough and coin press method for GDL production. While effective at lab scale, this method poses limitations in scalability, process flexibility, and structural reproducibility – key factors for advancing electrochemical technologies toward industrial adoption [2].

To address these challenges, AVT in collaboration with Coatema (COA) and under the EU-funded WaterProof project, is developing a roll-to-roll (R2R) manufacturing process aimed at continuous, scalable GDL production. Initial lab-scale trials using a sheet-to-sheet (S2S) system enabled optimization of process parameters and successful adaptation of the coin press recipe. This work formed the basis for prototype development at COA, culminating in a full R2R GDL production line integrating material application, lamination, drying, and calendering units [3].

Laboratory studies explored key boundary conditions, including lamination quality and thermal behavior at temperatures up to 200°C. These findings were transferred to a calendaring unit, where continuous production of mechanically durable and uniform GDLs was demonstrated.

By translating lab-scale knowledge into industrially viable production protocols, this work marks a significant step in upscaling GDE technology. It supports the broader goal of realizing scalable, cost-effective electrochemical CO_2 conversion systems, aligned with ongoing pre-pilot demonstrations at commercial current densities. The R2R method developed under WaterProof brings us closer to large-scale manufacturing of high-performance electrodes, helping bridge the gap between research and industrial implementation.

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Electrochemical Investigation of CO Reduction with Assistance of Proton Pumping

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The electrochemical reduction of carbon dioxide (CO_2) and subsequently carbon monoxide (CO) has been demonstrated to offer a promising route towards sustainable chemical production, utilizing renewable electricity to convert CO_2 into valuable C_{2+} products.[1] In this study, investigation focused on the design of a fully gaseous membrane electrode assembly-based (MEA) electrolyzer system, in which the conventional oxygen evolution reaction (OER) was replaced by the hydrogen oxidation reaction (HOR) on the anode. This approach was motivated by designing simplified systems that would eliminate the need for liquid electrolyte and allow system operation at lower cell potential. Particularly, this study focuses on elucidating the impact of the various membranes utilized in MEAs, including both commercial and self-made bipolar membranes (BPMs), on the electrochemical performance of the electrolyzer in CO electrolysis and proton pump experiments.

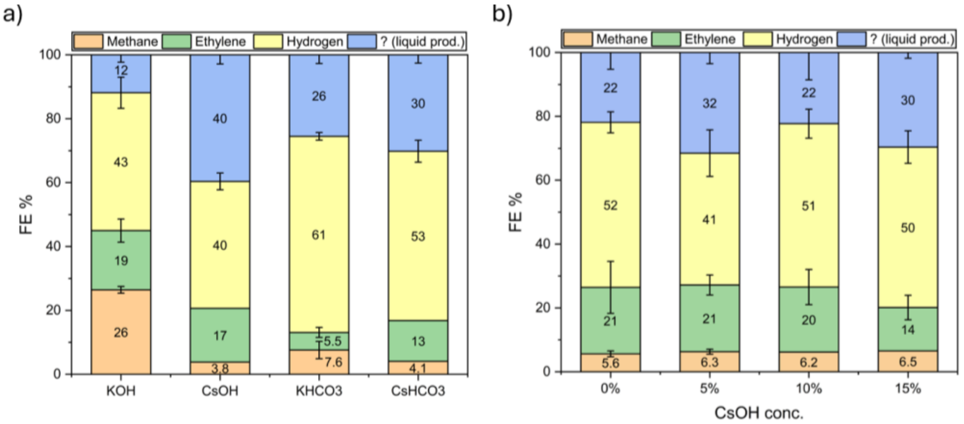

***Figure 1**: a) FE graph for the CORR with Nafion 117 + Piperion BPM soaked in different solutions: KOH, CsOH, $KHCO_3$, and $CsHCO_3$ at 100 mA/cm^{-2}. b) FE graph for the CORR with Fumatech BPM with increasing CsOH concentration from 0 % up to 15 % at 50 mA/cm^{-2}*

Proton pump experiments have demonstrated that BPMs composed of Nafion 117 and Piperion, which are self-made, exhibit superior stability and ion transport properties when compared to commercially available BPMs. This enhancement in performance results in higher current densities, thereby establishing them as a viable alternative for forward bias operation. The operation of the established BPMs was achieved at a maximum ethylene FE of 20%, which is approximately half of the benchmark performance typically observed in standard zero-gap liquid-phase electrolyzers.[3] Challenges, particularly with CO diffusion through the membrane, lead to anode CO poisoning and performance degradation of Pt-based HOR catalysts, which was mitigated using Pt-Ru alloys. While CO tolerance improved significantly, potential oscillations between -1.8 V and -2.4 V are highlighted, which undermine long-term stability. Furthermore, attempts to utilize the cation effect to improve selectivity toward ethylene production did not yield significant improvements, with hydrogen evolution remaining dominant in the process.[2]

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Pore-scale Simulation of Water Management in Gas Diffusion Electrodes for CO_2 Reduction

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Electrochemical reduction of CO2 (CO2R) into useful chemicals represents a solution to mitigating climate change by closing carbon emissions cycles. CO2R electrolyzer technology, however, needs to be improved before it can be widely deployed at an industrial scale.(1)
Gas-diffusion electrodes (GDEs) can overcome mass-transfer limitations related to the diffusion of CO2 feed gas into the aqueous electrolyte. GDEs are typically composed of a gas diffusion layer (GDL) through which CO2 can easily diffuse, and a catalyst layer (CL) where CO2 dissolves into liquid electrolyte before reacting on solid catalyst surfaces. Flooding refers to the complete filling of CL and GDL pores with electrolyte, and represents a significant challenge for the scale up of CO2R.(1) Indeed, it is understood that there exists an optimal partial filling of CL pores which maximizes reaction and product selectivity.(2) Furthermore, GDL flooding prevents feed gas diffusion and leads to cell failure.(3)
Electrolyte wetting in the CL and eventual flooding are affected by many factors beyond overall porosity or pore geometry,(4,5) such as electrowetting due to electrode potential,(6,7) local pH conditions and concentrations,(8,9) and salt precipitation.(1) Local conditions, and therefore CL micrometer-scale characteristics, which favor optimal electrolyte filling are not well understood, and cannot be easily measured. Therefore, 3D simulations can be used to give new insights where hydrodynamic, electrokinetic, and advection-diffusion-reaction phenomena are coupled.
A framework for such simulations was developed, carefully considering how larger simulations would be able to scale, and allowing for flexibility in boundary conditions to support variable wetting and electrochemical double layer (EDL) modeling. Particular attention is given to the coupling between the hydrodynamic and advection-diffusion-reaction systems, especially as entrapped gas bubbles may affect species diffusion in nearby electrolyte.

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Hierarchically Structured catalyst layers for PEM fuel cell

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Hydrogen as a potential energy carrier plays a pivotal role in clean energy conversion. Polymer electrolyte membrane fuel cells (PEMFC) are considered efficient for future sustainable mobility devices. However, the major challenges in commercializing PEMFC are the high cost of Pt-based c atalysts as well as implementing efficient methods in fabricating gas diffusion electrodes to attain higher performance.

To make PEMFCs more cost competitive, its essential to fabricate efficient catalyst layers (CL) to enhance catalyst utilization beyond the right choice of material.
In the conventional process the catalyst ink if formed through the evaporaton of solvents from the colloidal catalyst inks. These can lead to multiple scenarios like catalyst agglomeration, CL inhomogenity, flooding and fuel starvation. To bridge these gaps we are adapting a new strategy of a special thermal treatment of the catalyst layer to form a hierarchial CL structure. This can be characterised by high interfacial area and low interfacial energy to facilitate the mass transport. Comparative studies by considering various factores including ionomer carbon ratio, solid content and drying conditions are being carried out.

On the device level, the electrodes for PEMFC can be optimized by tuning catalyst-ionomer-solvent-ratios. This can lead to better mass transfer, charge transfer, catalyst utilization and water dissipation. Additional structural engineering can be achieved with optimizing doctor blade coating for decal process followed by spray coating for catayst coated membranes. Our work includes a comprehensive understanding of the catalyst performance in device level by the feedback generated from electrochemical and surface spectroscopic techniques. The enhancement in the mass transport and charge transport will be studied extensively in comparison with conventional process.

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Evaluating Performance of Hybrid Pt-Catalysts Under Realistic Conditions for Fuel Cell Application

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Hydrogen stands at the forefront of our transition to net-zero emissions. The increasing demand for green hydrogen necessitates innovative methods for its transport. In this regard, hydrogen carriers, e.g. ammonia and methanol, are very promising for long distance transport. However, the hydrogen obtained from those carriers may contain impurities such as NH_3, CO and CO_2, which can impair anode performance in fuel cells.[1] Having impurity tolerant anodes is thus crucial for fuel cells in a new hydrogen economy based on such hydrogen carriers. Minimizing the cost is also crucial for widespread adoption of these technologies which can be achieved by employing non-noble metal catalyst for the oxygen reduction side, although, these catalysts often show poor stability for long term operations. In this context, hybrid catalysts with low platinum (Pt) loading, combining stability for the oxygen reduction reaction (ORR) and tolerance to anode-side impurities, emerges as an attractive solution.

In this work, we evaluate the performance of Pt-FeNC hybrid catalysts and benchmark it against the commercial FeNC and Pt/C catalyst with similar Pt content. Noble metal-based electrocatalysts, especially Pt, are highly active for both hydrogen oxidation reaction (HOR) and oxygen reduction reaction (ORR) but are sensitive to poisoning. The hybrid Pt-FeNC catalyst, with a Pt-loading up to 2 wt.%, has been also found to be active for HOR, but was not prone to adsorption of CO or CH3OH.[2] It was suggested that the Pt was covered by a thin shell of Fe-oxide, making it promising as a poison-tolerant catalyst. [3]

Rotating disk electrode (RDE) and gas diffusion electrode (GDE) and polymer electrolyte membrane fuel cell (PEMFC) setups are employed to characterize the catalyst. GDEs are promising as they allow to study realistic catalyst layers at elevated current densities along with realistic mass transport.[4] The transferrability of the testing protocols and performance are evaluated by testing the optimized electrodes in fuel cell. We investigate the influence of mass loading, binder concentration, and different gas diffusion layers.CO-stripping is utilized to analyse noble-metal accessibility or CO-adsorption sensitivity before and after HOR and ORR, along with structural analysis to characterize the catalyst layers.

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Investigation of Catalytic Ink Formulations for use in Slot-die Coating for Proton Exchange Membrane Fuel Cell Electrodes

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The structure and properties of the catalyst layer (CL) play a crucial role in the performance, cost, and durability of proton exchange membrane fuel cells (PEMFCs) [1,2]. To address these challenges, several approaches have been explored to optimize catalyst composition, ink formulation, and coating techniques, aiming to enhance mass transport and overall efficiency [2,3,4]. However, scaling these methods for industrial application remains challenging, requiring further collaboration between academic and industrial researchers.

This study investigates the properties of various platinum-based, carbon-supported catalyst inks for slot-die coating applications, targeting scalable catalyst-coated membrane (CCM) production. Catalyst inks with varying solid contents and ionomer-to-carbon (I/C) ratios were prepared and analysed. Empirical studies were conducted on different dispersion methods, and ink slurries were characterized using particle size distribution, zeta potential, and rheological analysis to assess flow behaviour—an essential factor in slot-die coating [2]. Additionally, a three-interval thixotropic test was employed to simulate ink behaviour during slot-die extrusion.

Various substrates, including gas diffusion layers, proton exchange membranes, polytetrafluoroethylene (PTFE) foils, and polypropylene (PP) foils, were evaluated for their suitability in achieving uniform CL coatings. The decal transfer method, coupled with hot pressing, was employed to form homogeneous and well-defined catalytic layers with low interfacial resistance on commercially available membranes. Structural integrity was examined via scanning electron microscopy (SEM), laser scanning microscopy (LSM), and optical microscopy to assess uniformity and defect density. Performance validation was conducted by integrating CCMs with gas diffusion layers (GDLs) and proton exchange membranes in a half-cell setup. Inks with a solid content of 5% and an I/C ratio of 1.0 was found to yield uniform coating thickness and Pt loading on the decal substrate, in sheet-to-sheet coating.

Future research is being focused on optimizing the CCM-GDL interface and benchmark CCMs for single cell LT-PEMFCs aimed at heavy-duty applications, with a focus on improving performance and durability under targeted operational conditions. These advancements will contribute to the development of high-performance CCMs with enhanced durability and scalability.

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Alternative Gas Diffusion Electrode Designs: Influence of Porosity Gradients on the Electrochemical Activity

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Gas diffusion electrodes (GDEs) are essential in various electrochemical applications aimed at reducing CO_2 emissions and combating climate change, such as fuel cells, electrolyzers and metal-air batteries. [1-3] However, carbon-based GDEs often face issues with corrosion in alkaline environments during OER and ORR, necessitating metal-based materials as viable alternatives. [4] Nickel foam is an attractive solution due to its high OER electrochemical activity and stability in alkaline media. This study explores carbon-free gas diffusion electrodes (CF-GDEs) with distinct porosity gradients, made from MnO_x-coated macroporous substrates and hydrophobized with PTFE. The research demonstrates that these CF-GDEs significantly outperform a commercial carbon-based GDE (GDE_{ref}) by showing reduced overpotentials and enhanced electrochemical stability. In particular, the layered design Fop|Fop*|Fle* exhibited a one-third reduction in ηOER (0.24 V) compared to GDE_{ref} at 10 mA cm^{-2}. Noteworthy, this CF-GDE also shows excellent long-term stability without degradation, which is a common issue in carbon-based GDEs due to carbon corrosion. Stability tests revealed the formation of electrochemically active NiO_x, Ni_6MnO_8, and NiMn layered double hydroxides, resulting in a doubling of current densities. These findings highlight the potential of CF-GDEs with optimized porosity gradients for advanced applications in sustainable energy technologies.

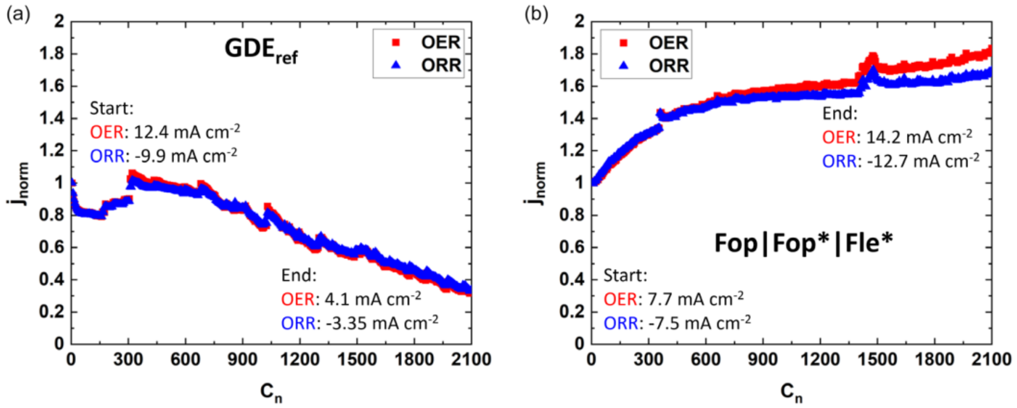

***Figure 1**: Long-term stability measurement of a) GDE_{ref} and b) Fop|Fop*|Fle* at 1.5 and -0.75 V versus Hg/HgO 1M NaOH for 2100 cycles and each working mode (OER and ORR) lasted 20 s.*

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Numerical Modelling and Fabrication of Enzymatic Gas Diffusion Electrodes for Oxygen Reduction

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Due to the structural simplicity and high energy density, small gas molecules such as H_2, O_2, CO, CO_2 are frequently regarded as reactants or intermediates in various sustainability and alternative energy strategies. However, the high bond symmetry and stability of these molecules typically require the use of catalysts and/or elevated temperatures and pressures to overcome high activation energy barriers in the reactions. As a promising alternative to expensive abiotic catalysts, redox enzymes have the ability to catalyse reactions involving these gases with remarkable catalytic efficiency and affinity under mild conditions. Nevertheless, the low solubility of these gases and the inability of enzymes to function in dry environments pose significant challenges for their integration into bioelectrodes. This limitation can be addressed by the use of gas diffusion electrodes (GDEs) [1], where enzymes are immobilized on the electrode surface close to a three-phase boundary. However, enzyme immobilization concentrates biocatalysts within a confined space, introducing conditions of molecular crowding and restricted substrate access. Additionally, at high reaction rates, the redox processes themselves can alter the local microenvironment, for instance, by generating or consuming protons, leading to local pH fluctuations that further impact enzymatic activity [2].

Altogether, predicting the behaviour of such bioelectrodes under varying conditions is inherently complex due to the numerous variables involved in electroenzymatic systems, including electrode structure and enzymatic reaction kinetics. In this context, numerical modelling emerges as a valuable tool to elucidate how these factors influence enzyme performance and to direct the rational design of enzymatic bioelectrodes and GDEs [3].

In this study, we fabricated an enzymatic GDE for oxygen reduction, incorporating functionalized carbon nanotubes and bilirubin oxidase from *Myrothecium verrucaria* (*Mv*BOD). We employed Comsol Multiphysics® to model the electrocatalytic behaviour of the electrode, with particular focus on the local pH changes induced by the enzymatic reaction. We notably introduced the buffer equilibrium in the near-neutral pH-range by accounting for species activity coefficients, and the pH-dependence of enzymatic activity. This approach enabled us to predict the shape of experimental electrocatalytic curves under varying buffer concentrations. To validate the model, we used confocal fluorescence microscopy, which provided spatial and temporal resolution of pH changes occurring near the electrode during potential cycling [4]. The developed model can serve as a foundation for the rational design of more efficient and stable enzymatic GDEs.

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Effective Reaction Rates for Oxygen Reduction Reaction in Silver Gas Diffusion Electrodes from intrinsic Kinetics

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The oxygen reduction reaction (ORR) in porous gas diffusion electrodes (GDEs) plays a key role in many technical processes, such as chlor-alkali electrolysis. This study aims to improve ORR performance by developing a predictive model that links integral reaction rates to specific electrode geometries. On this basis, we intend to derive an optimised electrode design.

In this contribution we focus on the effective reaction rates in single pores. A spatially resolved geometry of the pore is taken from FIB-SEM measurements at HZB [1]. Based on the pore geometry the distribution of the electrolyte and the shape of the free surface is calculated using the method smoothed particles hydrodynamics (SPH). The intrinsic kinetics of the ORR reaction is taken from measurements in a rotating disc setup by Živković et al. [2].

A reaction-diffusion model is applied to different pore geometries to examine the influence of overpotential and geometric parameters on reaction rates. Key geometric parameters are identified, and a correlation could be derived for the integral reaction rate at high overpotentials (|η| > 0.3 V), using this parameters in dimensional analysis.

This study indicates a restriction of the reaction to a region close to the triple phase boundary at elevated overpotentials. As plotted in Figure 1, the concentration of oxygen (left) and reaction rate (right) both rapidly decline at the pores surface, in regions where the electrolyte is in contact to silver. Thus, we find a strong correlation of the integral reaction rate to the length of the contact line between silver and electrolyte at high overpotentials (|η| > 0.3 V).

The application of the correlation to real pores demonstrates a high degree of predictability. In the present study, a sample of pores was analysed. For an overpotential of η=-0.5V the integral reaction rate of these pores can be predicted with over 90% accuracy.

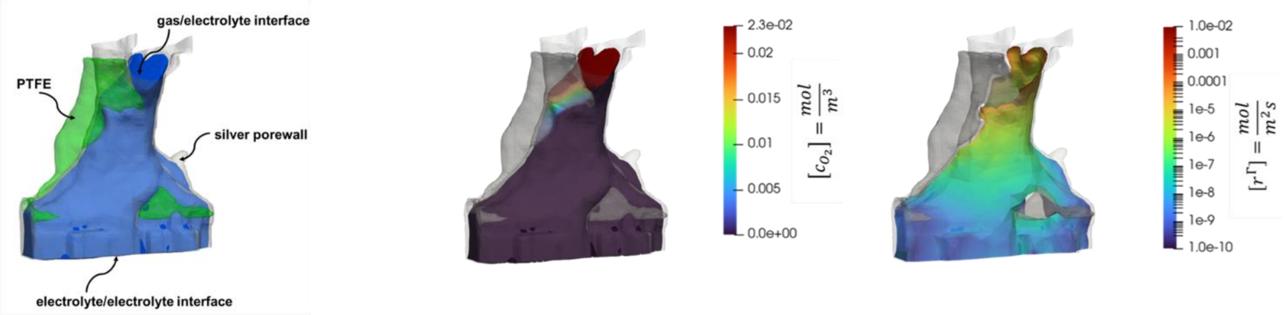

Figure 7: Electrolyte distribution and free surface at the gas/electrolyte interface in a real pore example (left). Oxygen distribution (middle) and reaction rate (right) modelled within the electrolyte of a pore.

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Laplacian pressure-controlled gas diffusion electrodes for organic electrosynthesis

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In synthetic organic chemistry, the utilization of gases like CO_2 as C1 building blocks or for hydrocarbon functionalization presents a major challenge [1]. Electrochemistry offers a broad potential window, circumventing high activation barriers and overpotentials, thus providing a sustainable alternative to conventional large-scale processes. However, mass transport limitations of gaseous reactants result in low yields and shifted selectivity [2]. The use of gas diffusion electrodes (GDEs) addresses this issue in aqueous electrolytes by establishing a liquid-gas phase interface on a thin catalyst layer, enabling short transport distances and preventing reactant depletion [3]. For organic electrolytes, conventional carbon/Teflon-based GDEs fail due to inadequate wetting or electrode flooding, as their mixed polar/non-polar nature hinders phase separation. Studies by Lazouski et al. in DMF have demonstrated that tuning the pressure balance between gas and liquid at a GDE allows precise control over catalyst wetting and electrolyte penetration [4]. This is achieved by a liquid column at the gas outlet, generating a counteracting Laplace pressure, which can be adjusted by varying the column height, enabling the use of metal mesh-based GDEs.

The collaborative project GDE4OES, part of the BMBF-funded Cluster4Future ETOS, aims to develop electrochemically stable, high-performance gas diffusion electrodes based on Ni-, Ag- and Pt-coated steel gauzes for cathodic limited organic syntheses such as CO2 reduction (CO2RR). The electro-carboxylation of acetonitrile to cyanoacetic acid serves as a representative model reaction. The concept of a Laplace pressure controlled GDE potentially offers an alternative to conventional large-scale processes that can be rapidly scaled up to an industrial production and transferred to further electrosynthesis for olefins, alkynes, aldehydes, ketones and imines [1].

Therefore, a continuous flow test setup is being developed that enables precise control of the gas-liquid interface position. An optical access window allows real-time imaging of the GDE surface to capture interfacial phenomena. This setup is used to investigate the wetting behavior, gas-liquid separation and permeation at various GDEs of different materials and structures. Additionally, the influence of the interface position on the electrochemical reaction is analyzed, with a focus on activity, selectivity and the reaction mechanism.

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Exploring membrane designs and reaction conditions towards selective formaldehyde formation in electrochemical methanol oxidation

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The electrochemical methanol oxidation to formaldehyde (FA) offers several advantages over established thermal production routes, including the potential for simultaneous hydrogen production and the utilization of renewable energy sources.[1,2] Another advantage of the electrochemical pathway is the potential to produce FA with low water content, potentially eliminating the need for energy-intensive separation of FA from water, which are formed in a 1:1 ratio in the established Formax and silver contact processes.

The first example of a gas-phase electrolyser for the selective oxidation of methanol on Pt functionalized Nafion membranes was already presented in 1992 by Fedkiw et al.[1], highlighting the strong dependence of product selectivity and water content in the product stream on the reaction conditions. Recently Mechler et al.[3] reported a faradaic efficiency of more than 80 % towards anhydrous FA at 100 mA/cm² over polycrystalline Pt from anhydrous methanol. However, similar experimental procedures by Jiao et al.[4] and Nam et al.[5] suggest that DMM is formed in the electrolyser and subsequent acid-catalysed hydrolysis is required to release FA, resulting in the undesirable addition of high amounts of water to FA.

Considering the controversy in the literature, we report here our recent understanding of FA formation by electrochemical methanol oxidation using a gas-phase electrolyser operated at 100 °C following the approach of Fedkiw et al.[1] We highlight the trade-offs in selective FA production depending on the water content in the gas-phase effluent stream. For instance, a decline in the effluent water content from 0.71% to 0.32% led to a decrease in FA selectivity from 40% to 24%, while the dimethoxymethane (DMM) was significantly favoured with an increase from 7% to 40%. The addition of gaseous water to the anode gas stream resulted in a shift in the FA/DMM equilibrium, favouring the FA selectivity again (42%). In addition, we will present our recent endeavours in MEA preparation using sputtered Pt films on GDL supports and membrane casting, as well as process parameter variations including catholyte variations to facilitate the formation of a Pt catalyst in the appropriate microenvironment to favor FA over DMM formation at low water contents.

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Powdered Catalyst Design for Ammonia Oxidation in Electrochemical Watersplitting

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Research on water electrolysis for the production of green hydrogen from renewable energy sources is essential for our future energy economy. A key component in this research is the improvement of the oxygen evolution reaction (OER), which represents the bottleneck in electrochemical water splitting.[1] While the main focus of OER research has been on reducing the overpotential by developing catalysts and improving cell setups, alternative reactions to bypass the OER are also increasingly coming into focus.[2,3]

By exploring alternative anodic reactions, it is possible to reduce the anodic half-cell potential of electrolysis while producing industrially relevant oxidation products compared to the oxygen of the OER. One such alternative anodic reaction is the ammonia oxidation reaction (AmOR), in which ammonia is anodically converted to industrially relevant nitrogen oxides, such as nitrate.[2,3]

Our work focuses on the design and development of nitrate-free synthesized layered double hydroxide (LDH)-based catalysts for AmOR. Due to the nitrate-free nature of our synthesized catalysts, we reduce the risk of false positive nitrate detections due to reactant residues in the investigation of possible AmOR catalysts. First tests on synthesized binary CuFe-, NiFe- and tertiary CuNiFe-LDHs showed clear AmOR activities without detection of nitrogen oxides coming from reactant residues. Besides the catalyst synthesis, our contribution will include AmOR tests of the entire substitution series from CuFe- over a wide compositional range of CuNiFe- to NiFe-LDHs as well as a detailed analysis of the post-electrolysis electrolyte.

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Multiscale Framework for Electrode–Electrolyte Simulations

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Computer simulations of electrochemistry systems help explore relevant microscopic details that are difficult to observe experimentally but are not free from artefacts resulting from finite-size effects. For example, an electrochemical cell includes two electrodes separated by a neutral bulk solution. In simulation, the two interfaces are separated by a few nm, a distance at least comparable to the correlation length of the solution. To overcome this problem, we propose a multiscale framework to simulate macroscopic separations between electrodes based on the H-AdResS+PI protocol [1, 2] and the Constant Potential Method (CPM) [3, 4]. The electrodes and their neighbouring molecules are described at a fully atomistic level and are coupled at constant chemical, thermal, and electric equilibrium to an infinite non-interacting particle reservoir. As an electrolyte, we employ a coarse-grained model of 1-butyl-3-methylimidazolium (BMI^+) and hexafluorophosphate (PF_6^-) [5, 6] in combination with graphite-based electrodes. The chosen continuous switching between models allows the resulting external potential in the interfacial region to explicitly include the long-range electric coupling between the electrodes and the electrolyte. This multiscale approach provides an efficient and precise tool for probing the structural and dynamic properties of electrolyte bulk and electrode-electrolyte interfaces in complex electrochemical systems.

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De Nora R&D in Gas Diffusion Electrodes – Solving Key Technology Challenges Together

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Catalyst Coated Substrates (CCS), including Gas Diffusion Electrodes (GDEs) as well as its close, complementary technology, the Porous Transport Layers (PTLs), are critical components of the Membrane Electrode Assembly (MEA) applied in electrochemical systems, enabling efficient gas-liquid-solid interactions essential for reactions such as hydrogen evolution or oxidation, oxygen evolution or reduction, and CO_2 reduction. State-of-the-art GDEs integrate advanced catalyst layers with engineered porous structures to optimize mass transport, electrical conductivity, and durability. Recent innovations include the use of conductive polymers on PTFE membranes to enhance stability and mitigate electrolyte flooding in gas-fed electrolyzers, controlled manufacture of GDE structure in carbon-free GDEs, application of complex structures as PTL components to control mass transport properties and non-aqueous GDEs for ammonia synthesis [1-4]. These developments are paving the way for scalable, industrially relevant systems in energy conversion and carbon utilization [5].

De Nora Group, a global leader in electrochemical technologies, offers a comprehensive portfolio of GDE products under its E-TEK® brand, which include anode and cathode for PEM, Alkaline and Phosphoric Acid (or high temperature) Fuel Cells, Oxygen Depolarized Cathodes (ODC) for brine electrolysis and hydrochloric acid recovery and complete electrode packages for alkaline water electrolysis [6-7]. De Nora's GDEs are designed and engineered for high system performance and aimed at reducing energy consumption. They feature optimized porosity, hydrophobicity, and catalytic coatings tailored to specific industrial needs and are backed by decades of R&D and global manufacturing capabilities, ensuring consistent quality and innovation.

With new innovations, De Nora is also venturing outside the classical technology segments, bringing our know-how and expertise to the fields of CO_2 electrolysis and PEM-Electrolysis. Since 2020, De Nora has actively taken part in several European funded R&D projects focused on advancing GDE and PTL technologies:

- **ISEHM**: Development and implementation of economical production of non-woven GDEs for HT-PEM Fuel Cells, where De Nora led the development and scalable manufacture of GDE on non-woven substrates. Attention was given to innovations in GDE manufacturing, including coating application method, thermal treatment and quality control, which could enhance production capacity of these GDEs without significant capital investment [8].

- **ANEMEL**: Electrolysis of low-grade water into green hydrogen, with focus on using abundant materials for membrane, catalyst and electrode development. De Nora role includes catalyst development and ink formulation, electrode and MEA production, support stack development and sustainable sourcing of materials [9].

- **ECO2Fuel**: Design, manufacture, operate and validate the worldwide first low-temperature, 1 MW direct, electrochemical CO2 conversion system to produce economic and sustainable e-fuels and chemicals. De Nora will develop and scale-up manufacture both anodes and cathode (based on GDE/PTL technology) using electrocatalysts and ionomers from project partners that converts CO_2 and water into synthetic fuels [10].

- **PROMISERS**: Advancement of new, non-PFAS components for PEM Fuel Cell and PEM Electrolysers, utilizing materials based on hydrocarbons and cellulose. De Nora's role is to develop coating inks and electrodes in the form of CCS/GDE or CCM and manufacture the MEA using non-PFAS ionomers and membranes developed within the project [11].

- **HYPRAEM**: Aims to develop an Anion Exchange Membrane Electrolyser stack and a layout capable of producing hydrogen at unprecedented high gauge pressures, ensuring direct integration into various processes used by the thermochemical industry. De Nora will support the project from two directions: 1. Development and Manufacture of novel Electrodes (based on GDE and PTL) and MEA, and 2. Design and Manufacture of Cells for the high pressure AEM electrolysis [12].

These collaborative efforts underscore De Nora's commitment to solving key technological challenges in electrochemical systems and driving the transition toward sustainable energy and chemical production.

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