

Design and Application of a Gas Diffusion Electrode (GDE) Cell for *Operando* and *In Situ* Studies

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Abstract: Presented here is an electrochemical three-electrode Gas Diffusion Electrode (GDE) cell tailored for *operando* and *in situ* investigations of electrocatalytic processes, with a particular focus on X-ray scattering studies. The optimized cell is engineered to accommodate the minimal sample-detector distances requisite for comprehensive X-ray total scattering investigations. An in-depth understanding of catalytic processes requires their study under ‘working’ conditions. Configured as a flow-cell, the setup therefore enables the examination of electrocatalysts under high current densities and associated gas evolution phenomena, particularly pertinent for reactions like the oxygen evolution reaction (OER). Notably, its transparency simplifies cell alignment, troubleshooting, and facilitates scans through the catalyst layer, crucial for background corrections. Demonstrating its versatility, we showcase its utility through Small Angle X-ray Scattering (SAXS), X-ray Diffraction (XRD), and X-ray Pair Distribution Function (PDF) analyses of total scattering data.

Keywords: Electrocatalysis · Gas diffusion electrode · High current density · *Operando* studies



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studies.



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1. Introduction

The transition towards renewable energy and abundant feedstocks requires, among others, the advancement of electrocatalysts. A notable example lies in the large-scale production of green hydrogen derived from water splitting through renewable energy sources. Green hydrogen can be used as a chemical feedstock but also for long-term energy storage. Even though the precious and scarce metals Ir and Pt are required for (acidic) water splitting, the cost of large-scale green hydrogen production predominantly depends on electric energy prices and not those of the metal precursors. On the other hand, their abundance may limit scalability of the technology.^[1] Equally important, however, the catalysts employed in this process, particularly the Ir-based OER catalysts, significantly determine its energy efficiency and consequently the cost of green hydrogen. Hence, there is a crucial demand for highly active electrocatalysts that also exhibit reasonable abundance and stability. It is only when all three criteria are met that we can effectively address the so-called terawatt challenge of large-scale renewable energy use.^[2,3]

Improved electrocatalysts can be developed by trial-and-error approaches, or by design through fundamental and applied research. The typical workflow involves synthesizing the catalysts, evaluating their performance, and characterizing them. In fundamental research, significant efforts have been directed toward achieving performance evaluations that can be effectively translated to ‘real-world devices’. Notably, this led to the development of gas diffusion electrode (GDE) setups in fuel cell research,^[4-7] which serve to bridge the gap between rotating disk electrode (RDE) measurements^[8] and the characterization of membrane electrode assemblies (MEAs)^[9] – the fundamental units of fuel cells or electrolyzers. In GDE setups thick catalyst films instead of thin films are studied and high current densities are achieved. Some designs interface the catalyst and liquid-electrolyte reservoir with a membrane electrolyte and even allow the operation at elevated temperature and pressure as is typical for MEAs.^[10] Furthermore, recent efforts in our group led to their application in electrolysis reactions such as the OER.^[11,12]

Most catalyst characterization, however, is still performed on the as-synthesized material under ambient or inert conditions. The thus determined catalyst structure is then compared with the catalyst performance, *i.e.* under electrocatalytic conditions. This approach neglects the fact that the catalyst often exhibits a substantially different structure during the reaction than when observed under ambient or inert conditions. In other words, the ‘real catalyst’ only exists under reactive conditions. Such derived structure-activity relationships therefore must be critically assessed for their limitations. For this reason, *in situ* and *operando* studies become increasingly popular. They typically require substantially more effort than the above-mentioned *ex situ* studies, and often the use of large-scale facilities such as synchrotrons. Nevertheless, they are essential for developing an in-depth understanding of electrocatalysis.

One distinguishes between ‘*in situ*’ study when the electrocatalyst is investigated in its ‘natural’ environment, and ‘*operando*’ study when in addition the reaction rate is monitored. Several studies discuss and demonstrate the merits of *in situ* and *operando* approaches in electrocatalysis, and here we only highlight the recently published review by Magnussen and Drnec *et al.*^[13] In the current work, we present the results of our efforts to translate the design of GDE setups to allow *in situ* and *operando* studies with a focus on the application of scattering techniques. First, we discuss the design criteria of our cell and present its layout, and second, we demonstrate its first application for two selected examples, *i.e.* the study of an oxygen reduction reaction (ORR) catalyst and the study of an OER catalyst.

1.1 Electrochemical Cells for Operando and In Situ Scattering Studies

In literature, several cell designs for *operando* and *in situ* measurements have been introduced. Two examples of designs that have been used by our research team are the ‘PSI design’ introduced by Binninger *et al.*^[14] and the ‘ESRF design’ introduced by Martens *et al.*^[15] These cells are suitable for *operando* and *in situ* X-ray absorption as well as X-ray scattering. However, they are not optimized for extremely close sample-detector distances required for total scattering for Pair Distribution Function analysis. In addition, in the ‘PSI design’ gas bubble formation can lead to an interruption of the cell contact due to the thin layer design. Nevertheless, the new cell design presented here is based on our extensive experience working with these cells^[16-20] and is understood as an advancement of these designs. Further cell designs for *operando* and *in situ* studies in electrocatalysis can be found in refs. [13,21-24] and the references therein.

1.2 Cell Design Criteria

The primary objectives of our new cell design were to address the following critical considerations for *operando* and *in situ* scattering studies:

- **Sample complementarity with ‘standard GDE measurements’:** In the new design, it is ensured that identical samples as used in our conventional GDE setup can be implemented in the cell. The catalyst loading is the same as the one typically used in GDE measurements, and the sample can be easily cut from the prepared GDE layers.
- **Achieving high current densities for gas-consuming and gas-evolving reactions:** we designed a three-electrode cell with a large electrolyte volume (Fig. 1), clear separation of the working electrode (WE), reference electrode (RE), and counter electrode (CE) to minimize cross-contamination and disturbances from gas bubble formation (note that at high current densities, typically gas bubble formation at the CE becomes a problem even though gas consuming reactions are investigated at the WE). Furthermore, the *iR* and cell resistances are minimized due to an integrated Luggin capillary and compact design. Additionally, the horizontal geometry facilitates the release of gas bubbles at the WE and CE.
- **Optimized X-ray geometry:** We employed an in-plane geometry for X-ray analysis to maximize the cross-section and facilitate the monitoring of the catalyst layer at various depths with respect to the catalyst-electrolyte interface. Furthermore, we reduced the beam pathway travelled through the electrolyte and cell body materials to minimize interference and optimize background subtraction.
- **Optimized X-ray detection:** The angular opening was maximized, and the cell-detector distance minimized to maximize the Q-space range for total scattering and improve data quality.
- **Transparent and resilient materials:** Utilizing a cell body material that is as transparent to X-rays as possible. Transparency to visible light facilitates trouble shooting, *e.g.* the

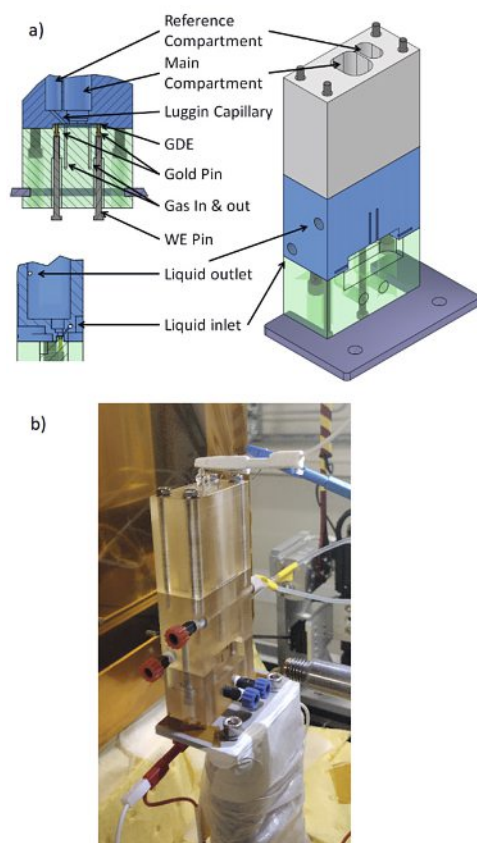


Fig. 1. a) Schematic of the 3-electrode GDE cell for X-ray scattering. b) Photo of the realized cell mounted in preparation for X-ray scattering measurements at DanMAX beamline at MAX IV synchrotron in Lund, Sweden.

identification of gas bubbles in the cell. We used polysulfone (PSU) as cell material as it is also resistant to acidic and alkaline environments.

- **Ease of assembly and mounting:** The cell is designed to facilitate assembly and mounting under challenging circumstances, such as working late nights as is common at synchrotron beamtimes.

2. Examples of Experiments Performed with the New Cell Design

To demonstrate the applicability of the newly designed cell, we have performed electrochemical *operando* X-ray scattering experiments of both gas-consuming and gas-evolving reactions. In gas-consuming reactions such as the ORR, high current densities can only be achieved if the gas transport through the liquid, *e.g.* the electrolyte, is avoided or limited. In classic ORR RDE measurements, the achieved current densities are only a few $\text{mA cm}^{-2}_{\text{geo}}$ before the reaction rate becomes mass transport limited. In contrast, up to a few $\text{A cm}^{-2}_{\text{geo}}$ can be achieved in GDE and MEA configurations. As the catalyst_{geo} structure might also change with current density/applied electrode potential, achieving high current densities in *operando* measurements is desirable. Gas-evolving reactions such as the OER typically face the problem of gas bubble removal. High current densities inevitably lead to massive gas evolution which can lead to a loss in electrical contact in the cell.

2.1 Experimental Details

The electrochemical cell is run in a three-electrode set-up with the working electrode (WE), a platinum counter electrode (CE), and a leakless Ag/AgCl (eDAQ) reference electrode (RE). The working electrodes were prepared by vacuum filtration of the re-

spective catalyst inks onto a gas diffusion layer (GDL) coated with a C-based microporous layer (MPL) (Freudenberg H23C8, Fuel Cell Store). The electrode of the Pt/C commercial catalyst (Tanaka, TEC10E50E-HT) was prepared with a nominal loading of $200 \mu\text{g cm}^{-2}$ *via* vacuum filtration and was measured with $0.1 \text{ M H}_2\text{SO}_4$ (Suprapur) in the upper compartment. No membrane electrolyte was separating the catalyst layer and liquid electrolyte. The NiFe LDH catalyst was prepared following a literature procedure.^[25] The NiFe LDH working electrode was prepared with a nominal loading of $400 \mu\text{g cm}^{-2}$ and was measured in 0.5 M KOH (Suprapur).

X-ray scattering data was collected in two experiments at two different beamlines: at ID31 at the European Synchrotron Radiation Facility (ESRF) in Grenoble, France, and at the DanMAX beamline at the MAX IV synchrotron in Lund, Sweden. At the ESRF the X-ray energy was 75 keV while at MAX IV it was 35 keV . In all experiments, the beam was microfocused to achieve a vertical beam size of less than $10 \mu\text{m}$. This allows height scans (z-scans) through the catalyst layer, to obtain scattering data of both the electrochemical cell, the electrolyte, and the carbon support, to allow for background subtraction.^[20] The scattering data was collected as 2D images on Dectris Pilatus3 X CdTe 2M detectors. The 2D data were azimuthally integrated with the PyFAI software.^[26] All X-ray scattering data were collected during potential holds.

Rietveld refinement of the X-ray diffraction data was performed with the GSAS-II software package.^[27] A model of a Pt *fcc* phase (Fd-3m, 225) was fitted to the experimental data. Isotropic models were used for the domain size, microstrain, and thermal displacement parameter U_{iso} . The instrumental resolution parameters for peak broadening were determined from a CeO_2 reference standard.

For SAXS data analysis, the cell and the carbon background were subtracted to isolate the Pt scattering signal from the other contributions. The background-subtracted SAXS data were fitted over a q -range of $0.03\text{--}0.5 \text{ \AA}^{-1}$ with a polydisperse spherical model. The size distribution is lognormal and number-weighted. SAXS data were analyzed with the SAS View software package (*SasView*, <https://sasview.github.io/>).

The collected X-ray total scattering data was Fourier transformed to obtain the PDFs using xPDFsuite.^[28] For the data reduction $Q_{\text{min}} = 2 \text{ \AA}^{-1}$, $Q_{\text{max}} = 19.5 \text{ \AA}^{-1}$, $Q_{\text{maxinst}} = 26 \text{ \AA}^{-1}$ and $r_{\text{poly}} = 0.9 \text{ \AA}$ were used. A Pt model was fitted to the PDF data using the PDFgui software.^[29] The background signal from the cell, the electrolyte, and the carbon background were subtracted from the data in Q -space. The parameters Q_{damp} and Q_{broad} were determined from the CeO_2 reference. A Pt *fcc* structural model was fitted to the data to determine the lattice parameter a , the atomic displacement parameter U_{iso} , and a nanoparticle size using a spherical shape factor (Sp diameter).

2.2 Studying Oxygen Reduction Reaction Catalysts

We have used a commercial Pt/C catalyst, with an average particle size of $4\text{--}5 \text{ nm}$, as a reference catalyst to conduct *operando* X-ray scattering experiments during the ORR. The catalyst was deposited on a GDL/MPL to function as a GDE, and the cell was operated in self-breathing mode (oxygen reaches the catalyst from the atmosphere *via* diffusion) with $0.5 \text{ M H}_2\text{SO}_4$ as the electrolyte in the upper reservoir. The electrolyte reservoir was continuously refreshed using a two-channel peristaltic pump. Scattering data was collected at different potential holds, starting from OCP, holding at 0.9 V vs RHE and 0.4 V vs RHE . At each potential, we acquired SAXS, XRD, and PDF data. The catalyst layer was scanned across in height with a microfocused beam, and in Fig. 2 the diffraction data collected at different z-locations in the cell are shown. With this set of scattering data, background subtraction of the carbon and electrolyte components is achieved. Background subtraction of the individual components is especially important

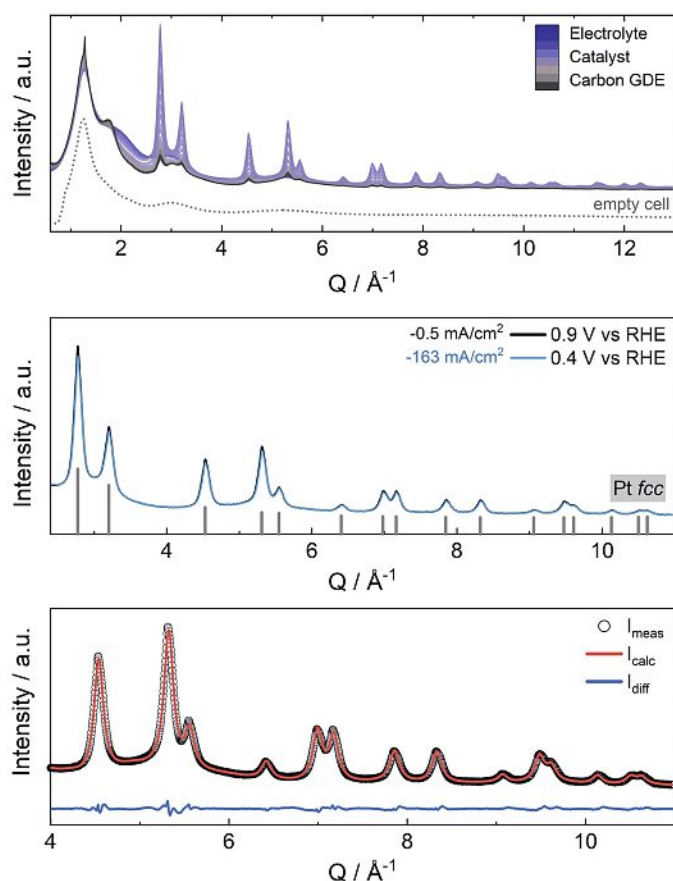


Fig. 2. Top panel: XRD scattering patterns obtained from a vertical scan through the cell from the GDL through the Pt/C layer (loading $200 \mu\text{g cm}^{-2}$) up to the electrolyte. The diffraction pattern from the PSU polymer collected for the empty cell is shown with an offset. Middle panel: XRD patterns collected of the Pt/C catalyst during potential holds at 0.9 V vs RHE and 0.4 V vs RHE, plotted together with a reference of an *fcc* Pt crystal structure (lattice constant $a = 9.25 \text{ \AA}$). The steady-state current densities recorded at the respective potential hold are given in the figure legend. Bottom panel: Rietveld refinement of the diffraction pattern collected at 0.4 V vs RHE.

for the analysis of SAXS and PDF data. While the empty cell and the other background components show scattering features at low Q ranges, the diffraction pattern of the Pt nanoparticles dominates at higher Q , and we can assign all Bragg peaks to the Pt reference.

The diffraction data collected during ORR at 0.4 V vs RHE differs only slightly from the pattern collected at 0.9 V vs RHE (middle panel Fig. 2). However, current densities of $> 0.15 \text{ A cm}^{-2}$ could be maintained, which is extremely high for *operando* scattering conditions.

The XRD patterns collected in the cell are of a high data quality, which is required for Rietveld analysis. A Pt *fcc* model was fitted to the XRD data and allowed to determine the lattice parameter of the NPs under potential control as well as crystalline domain sizes and microstrain. A decrease in lattice parameter and an increase in microstrain is observed when stepping the potential from 0.9 V to 0.4 V vs RHE. This agrees with *operando* diffraction studies on Pt/C ORR catalysts performed in an RDE-type electrochemical setup at low current densities.^[30]

Furthermore, with the newly developed cell, also SAXS and PDF data were obtained in very good quality that allow it to fit structural models to the data, and thus extract additional quantitative structural information of the catalyst under high current density *operando* conditions in the GDE cell. In Fig. 3, SAXS and PDF fits of experimental data obtained for the Pt/C catalyst during ORR (0.4 V vs RHE) in the GDE cell are shown. The fit parameters are included in Table S1.

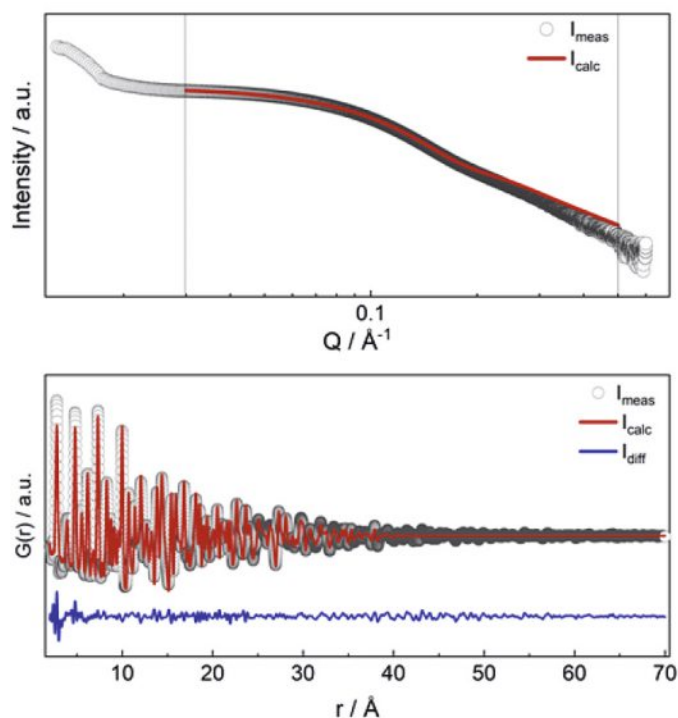


Fig. 3. Top panel: Fit of a model of polydisperse Pt spheres to the SAXS data of the catalysts obtained at a potential hold at 0.4 V vs RHE. Bottom panel: Fit of a Pt structural model constructed of spherical particles to the PDF data $G(r)$ obtained at a potential of 0.4 V vs RHE.

2.3 Studying Oxygen Evolution Reaction Catalysts

To demonstrate the capabilities of the cell for gas-evolving reactions, we have collected data from a NiFe-layered double hydroxide (LDH) sample as an OER catalyst in the newly developed cell. NiFe LDH is a state-of-the-art catalyst for OER in alkaline media and is known to undergo structural changes under reaction conditions.^[31] We applied a series of potential holds (1.4 V to 1.7 V vs RHE) and collected X-ray diffraction data at each potential step. Total scattering data for PDF analysis were collected separately at different experiments. The obtained total scattering data and PDFs are shown in Fig. 4. Both in the XRD and PDF, structural changes are apparent when the potential and hence the OER current is increased.

An advantage of the cell design with the catalyst layer in horizontal orientation is that bubbles from evolving gases can easily escape in an upward direction. This allows for much higher current densities in the cell compared to cells that use thin electrolyte films with the working electrode in a vertical direction. In the example shown below OER current densities of 35 mA cm^{-2} could be maintained over the prolonged measurement time required for total scattering.

3. Conclusions

We introduced here a new electrochemical cell design tailored for *operando* X-ray scattering experiments implementing a 3-electrode GDE design. The cell underwent rigorous testing under *operando* conditions at two different synchrotrons, enabling the comprehensive characterization of ORR and OER catalysts. The design yielded high-quality XRD, SAXS, and PDF data even under conditions of high current densities. The horizontal layout of the WE effectively managed vigorous bubble formation without compromising data quality. Additionally, the in-plane X-ray geometry probed with a microfocused beam enabled the recording of essential background data, facilitating comprehensive structural analysis.

Computer files from the cell drawings can be obtained from the authors upon request. Supplementary information is available here.

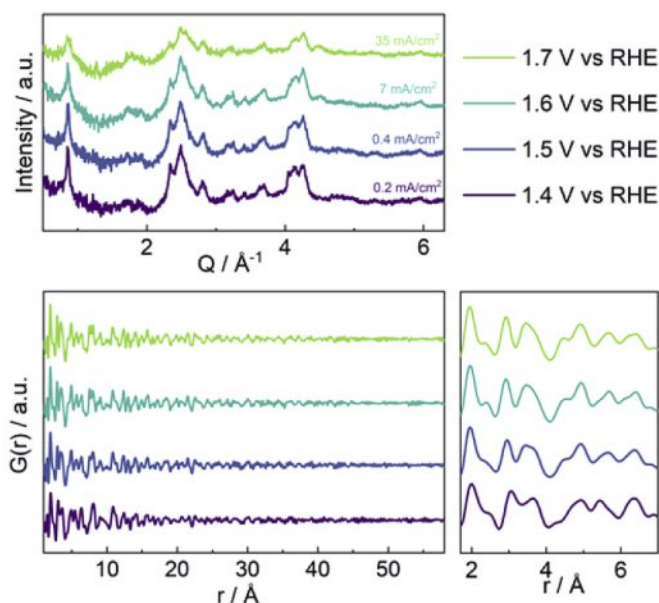


Fig. 4. Top panel: Total scattering data from the NiFe LDH catalyst collected during potential holds, going from 1.4 V vs RHE to 1.7 V vs RHE. Current densities measured for the respective potentials are included in the figure. Bottom panel: PDF data obtained from X-ray total scattering data for the NiFe LDH catalysts collected during the potential hold series with a zoom-in (right) into the local range of the PDF.

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Use of AI in Writing Process

The authors used AI tools in word (auto suggestion of phrases) and ChatGTP 3.5 to check for language and individual phrases. No complete sections were written nor was any reference added with the help of AI tools.

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