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Supplementary Information for:

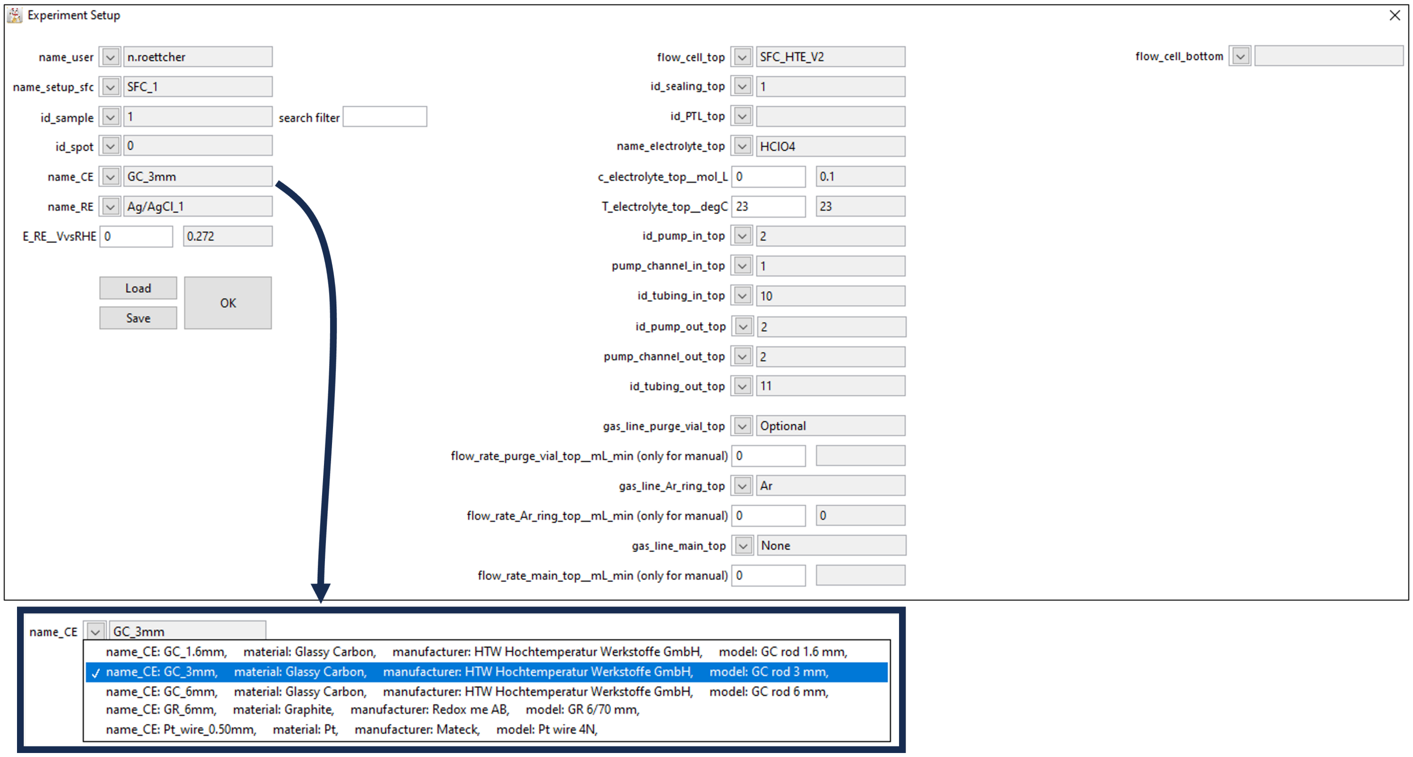
Accelerating materials research with a comprehensive data management tool: A case study on an electrochemical laboratory

Nico C. Röttcher,*[a,b] Gun D. Akkoç,[a,b] Selina Finger,[a,b] Birk Fritsch,[a] Jonas Möller,[a] Karl J. J. Mayrhofer,[a,b] and Dominik Dworschak*[a]

1 Database Schema

The database schema used in our group for SFC-ICPMS measurements is attached as a separate file to the Supplementary Information. Furthermore, in the data repository, the database schema is available as MySQL file.[1] Experimental metadata related to the SFC setup is stored in the table *exp_sfc* and corresponding tables, i.e., *flow_cell_assemblies*, *flow_gas*, and *flow_electrolyte* for details on the used cell, electrolyte flow, and gases used. Similarly, electrochemical metadata is stored in table *exp_ec*. All metadata information is combined in the view *exp_ec_expanded*. Electrochemical and EIS data is stored in *data_ec* and *data_eis*, respectively. In the same way, for ICP-MS experiments *exp_icpms* and *data_icpms* stores metadata and experimental data. Selected analyte-internal standard couples are defined and stored in the table *exp_icpms_analyte_internalstandard*. Analysis parameters and results, for instance, for integration analysis on ICP-MS and electrochemical data is stored in the table *ana_integrations*.

2 Experiment – Database Communication



name_user	n.roettcher	
name_setup_sfc	SFC_1	
id_sample	1	
id_spot	0	
name_CE	GC_3mm	
name_RE	Ag/AgCl_1	
E_RE__VvsRHE	0	0.272
flow_cell_top	SFC_HTE_V2	
id_sealing_top	1	
id_PTL_top		
name_electrolyte_top	HClO4	
c_electrolyte_top__mol_L	0	0.1
T_electrolyte_top__degC	23	23
id_pump_in_top	2	
pump_channel_in_top	1	
id_tubing_in_top	10	
id_pump_out_top	2	
pump_channel_out_top	2	
id_tubing_out_top	11	
gas_line_purge_vial_top	Optional	
flow_rate_purge_vial_top__mL_min (only for manual)	0	
gas_line_Ar_ring_top	Ar	
flow_rate_Ar_ring_top__mL_min (only for manual)	0	0
gas_line_main_top	None	
flow_rate_main_top__mL_min (only for manual)	0	
flow_cell_bottom		

name_CE GC_3mm

name_CE: GC_1.6mm,	material: Glassy Carbon,	manufacturer: HTW Hochtemperatur Werkstoffe GmbH,	model: GC rod 1.6 mm,
✓ name_CE: GC_3mm,	material: Glassy Carbon,	manufacturer: HTW Hochtemperatur Werkstoffe GmbH,	model: GC rod 3 mm,
name_CE: GC_6mm,	material: Glassy Carbon,	manufacturer: HTW Hochtemperatur Werkstoffe GmbH,	model: GC rod 6 mm,
name_CE: GR_6mm,	material: Graphite,	manufacturer: Redox me AB,	model: GR 6/70 mm,
name_CE: Pt_wire_0.50mm,	material: Pt,	manufacturer: Mateck,	model: Pt wire 4N,

Figure S 1 Screenshot of user interface to specify metadata for the electrochemical experiment prompted before each experiment to ensure keeping track of any changes in metadata throughout an experiment day. The parameters are always prefilled from former experiment to have faster and better usability.

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[a] Forschungszentrum Jülich GmbH, Helmholtz Institute for Renewable Energy Erlangen-Nürnberg, Cauerstr. 1, 91058 Erlangen, Germany.
[b] Department of Chemical and Biological Engineering, Friedrich-Alexander-Universität Erlangen-Nürnberg, Egerlandstr. 3, 91058 Erlangen, Germany.
* E-mail: n.roettcher@fz-juelich.de, d.dworschak@fz-juelich.de.

3 Processing & Analysis

3.1 Experimental details to SFC-ICPMS experiments

The measurements were performed using a scanning flow cell (SFC) coupled with an inductively coupled plasma-mass spectrometry (ICP-MS) (Agilent 7900, Agilent Technologies). A polycrystalline Pt foil (99.99%, MaTecK) was used as the working electrode. The area of the electrode exposed to the electrolyte was 3.6 mm^2. A glassy carbon rod (diameter of 1.6 mm, HTW Hochtemperatur-Werkstoffe) was used as counter electrode, Ag|AgCl (Metrohm) as the reference electrode. The potential of the reference electrode against reversible hydrogen electrode (RHE) was determined using Pt wire (99.99%, MaTecK) and H_2 (H2 99.999%, Air Liquide). 0.1 M $HClO_4$ was used as electrolyte freshly prepared from perchloric acid (Suprapur® Merck) and Milli-Q water (>18 MΩ, Merck) and is continuously de-aerated by an Ar stream (Ar, 99.998%, Air Liquide). Electrochemical measurements are recorded by a Gamry Ref 620™ potentiostat (Gamry Instruments).

The Pt concentration in the electrolyte stream was measured by the ICP-MS following the intensities of ^{195}Pt with respect to the intensity of ^{185}Re as internal standard. Calibration was performed with freshly prepared standard solutions containining 0.5, 1.0 and 5.0 µg L^{-1} of Pt in 0.1 M $HClO_4$ (Certipur ICP-MS Standard, Merck). Before entering the ICP-MS, the sample was mixed with an internal standard solution containing 3.0 µg L^{-1} Re in 0.1 M $HClO_4$ (Certipur ICP-MS Standard, Merck). The flow rate of the electrolyte from the SFC to the ICP-MS was determined on 0.33 mL min^{-1}.

The measurement signal of the added internal standard is smoothed by means of linear fit. With the filtered signal, the calibration result, and the flow rate, the Pt dissolution profile is normalized to the mass flow rate. This is further normalized by the ECSA as determined by H_{upd} charge extracted from CV and assuming a surface specific charge of and 210 µC cm^{-2}. Electrochemical potentials are post-compensated by the high-frequency resistance determined by EIS spectroscopy. Electrochemical currents are normalized to the area of the electrode in contact with electrolyte.

3.2 Use-case: Modeling of Nyquist plot

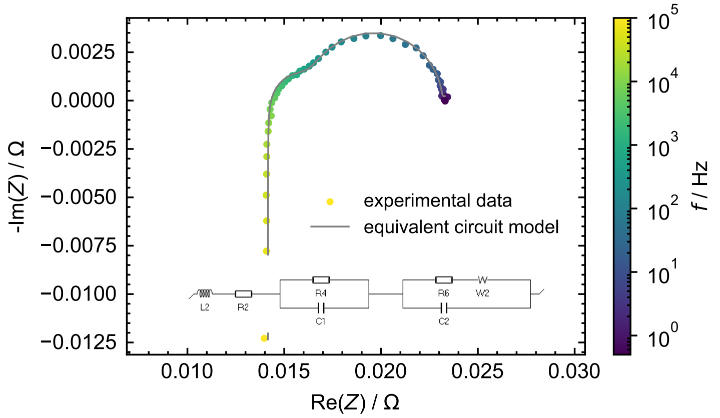

Figure S 2 Modeling of galvanostatic electrochemical impedance spectroscopy data using an equivalent circuit model depicted in the inset.

Electrochemical impedance data displayed in Figure S2 are recorded from a proton exchange membrane water electrolysis cell. The cell is assembled using an in-house designed cell fixture and comprises of an anode with a catalyst loading of 1.17 mg$_{\mathrm{Ir}}$ cm^{-2} (Umicore Elyst75 0480) and a Ti-fiber PTL 250 µm (Bekaert), a Nafion 212 membrane with 50 µm thickness and cathode with a catalyst loading of 0.18 mg$_{\mathrm{Pt}}$ cm^{-2} (Tanaka Pt/C, TEC10V40E and a carbon PTL with MPL (Freudenberg H24C5). Electrodes are manufactured via Mayer rodding on decal and hot pressing with membrane. Electrode and MEA preparation described elswhere.[2] Electrochemical experiments are performed using a Biologic VSP 300 equipped with a Booster at 80 °C with anodic DI water flow rate of 100 mL min^{-1}. Galvanostatic electrochemical impedance spectroscopy experiments arer recorded from 500 mHz to 100 kHz at an offset current density of 0.6 mA cm^{-2} and a perturbation of 0.1 A.

Modeling of the Nyquist plot is performed using the Python module *impedance.py* with 'L0+R0+(R1/C1)+(R2+W)/C2' as equivalent circuit model.[3]

3.3 Use-case: Feature extraction from ICP-MS and correlation to electrochemical potential

To showcase the power of correlating multiple experiment, in the main text, the coupling of electrochemical experiments to dissolution data as recorded by ICP-MS is described. Fiedler *et al.* recently introduced a feature extraction algorithm of ICP-MS dissolution profiles.[4] Thereby, a Python-based routine was applied to fit the dissolution data with a set of lognormal functions in order to extract the peak maxima (modes) in the spectra. The timestamp of the ICP-MS signal can be correlated with the potential of the CV as annotated in **??**c in the main text. Linear interpolation of the measurement points was exploited to correlate the fitted time point to the accurate potential. Subsequent error propagation allowed to gauge an uncertainty of this potential (see Fiedler *et al.*[4] for details). This analysis routine is applied on data illustrated in **??**c in the main text with the results being depicted in Figure S3. Such an analysis procedure on correlated experiments can be seamlessly performed due to the database-integrated data workflow.

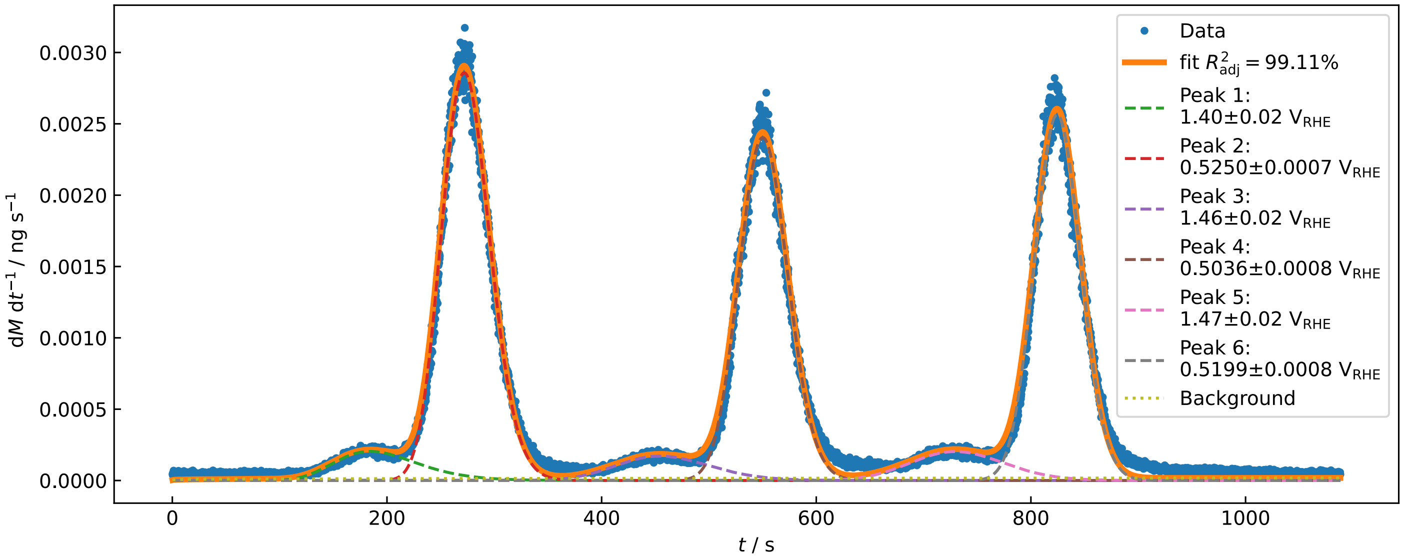

Figure S 3 Application of a feature extraction algorithm on SFC-ICP-MS data of electrocatalyst ($M = m_{\mathrm{Pt}}$) dissolution as introduced by Fiedler *et al.*[4] on data illustrated in **??**c in the main text, clearly separating anodic and cathodic dissolution peaks[5]. The legend data refers to the peak mode.

4 Visualization of data

In [ ]:

```
# query EC experiments based on metadata
exp_ec = pd.read_sql('''SELECT  *
                        FROM hte_data.exp_ec_expanded
                        WHERE name_user="n.roettcher"
                            AND name_setup_sfc = "SFC_1"
                            AND DATE(t_start__timestamp) = "2023-10-06"
                            AND fe_top_id_pump_out_device = 'SFC'  # not connected to ICP-MS
                            AND fe_top_pump_rate_out__rpm = 0  # no electrolyte flow
                            AND ec_name_technique="exp_ec_cv"
                            AND ROUND(cv_E_apex1__VvsRE + ec_E_RE__VvsRHE, 1) = 1.2  # or 1.6
                        ;
                        ''', con=con, index_col='id_exp_sfc')
```

In [ ]:

```
# query EC data based on selected experiments
data_ec = exp_ec.dataset.get_data(con, 'data_ec', add_cond = 'cycle IN (2)')

# apply HFR from EIS experiment linked to the selected experiments
exp_ec = tools_ec.update_R_u__ohm(exp_ec)
# calculate post-compensated potentials
data_ec = tools_ec.compensate_R_u(exp_ec, data_ec)
# calculate geometric current density
data_ec = tools_ec.geometric_current(exp_ec, data_ec, geo_cols='fc_top_name_flow_cell_A_opening_ideal__mm2')
# load samples composition from database and calculate gravimetric current density if applicable
exp_ec, data_ec = tools_ec.gravimetric_current(exp_ec, data_ec)
# Derive ECSA applying Hupd method for Pt, fitting a horizontal line for the capacitive current
exp_ec, data_ec = tools_ec.derive_ECSA(exp_ec, data_ec,
                                       method='Pt_Hupd_horizontal',
                                       geo_cols='fc_top_name_flow_cell_A_opening_ideal__mm2')
```

In [ ]:

```
# Apply our research group style template to the following matplotlib figure
with plt.rc_context(plot.get_style(style='singleColumn',)):
    fig = plt.figure()
    ax1 = fig.add_subplot(111)
    exp_ec.dataset\
          .add_column(# Define the legend label for the experiments based on the scanrate of the experiment
                      'label', values=exp_ec.cv_scanrate__mV_s.astype(str)+' mV s$^{-1}$')\
          .add_column(# Apply a colormap on the different experiments
                      'color', values='pub_blues', cmap_min=0.3, cmap_max=0.8)\
          .plot(# Plotting geometric current density vs compensated potential for all experiments
                x_col='E_WE__VvsRHE',
                y_col='j__mA_cm2geo_fc_top_cell_Aideal',
                data=data_ec,
                ax=ax1,
                )\
          .fill_between(# Fill the area between total current and capacitive current
                        # as derived from ECSA tool indicating the faradaic charge
                        x_col='E_WE__VvsRHE',
                        y_col= 'j_capacitive__mA_cm2geo_fc_top_cell_Aideal',
                        y2_col = 'j__mA_cm2geo_fc_top_cell_Aideal',
                        data=data_ec,
                        ax=ax1,
                        label='',
                        alpha=0.6,
                        axlabel_auto=False,
                        )
    ax1.legend()
    ax1.set_ylim([-0.5, 0.35])
    ax1.set_xlim([-0.05, 1.3])
    fig.savefig('20231013_visualization.svg')
    plt.show()
```

Figure S 4 Exemplary script to visualize database-stored experiments.

As outlined in Figure S4, experiments are selected by means of SQL query referring to the database view *exp_ec_expanded*, which combines metadata information from all linked tables. Thus, complete metadata is joined into a single table and thus can be used to flexibly filter experiments.

As the experimental data is linked to the experiment by identifier, it can be retrieved by with no additional user input. Normalization of the current, referencing of the electrode potential to the RHE, as well as post-potential compensation is performed afterwards. While this could also be performed within the previous step when retrieving the electrochemical data, it is specifically mentioned here, to clarify data lineage.

The visualization of the experiments is built on Pythons libraries, i.e., Matplotlib and Pandas.[6,7] Panda's DataFrame class is expanded to allow plotting of all experiments within a single command. In the backend, for each entry in the experiment DataFrame a matplotlib object is added to a figure also considering any style specifications in that entry. By this the label, color, linestyle, as well as the subfigure where to place the object can be specified in the experiment DataFrame and, thus, easily be linked to the metadata of the experiment. As depicted in Figure S5, the label is directly linked to the scan rate of the CV.

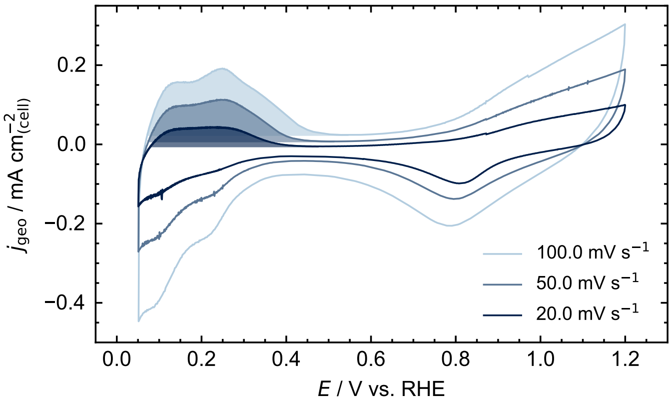

Figure S 5 Result figure of the script illustrated in Figure S4 displaying a CV on polycrystalline Pt at varying scan rate.

5 Server infrastructure

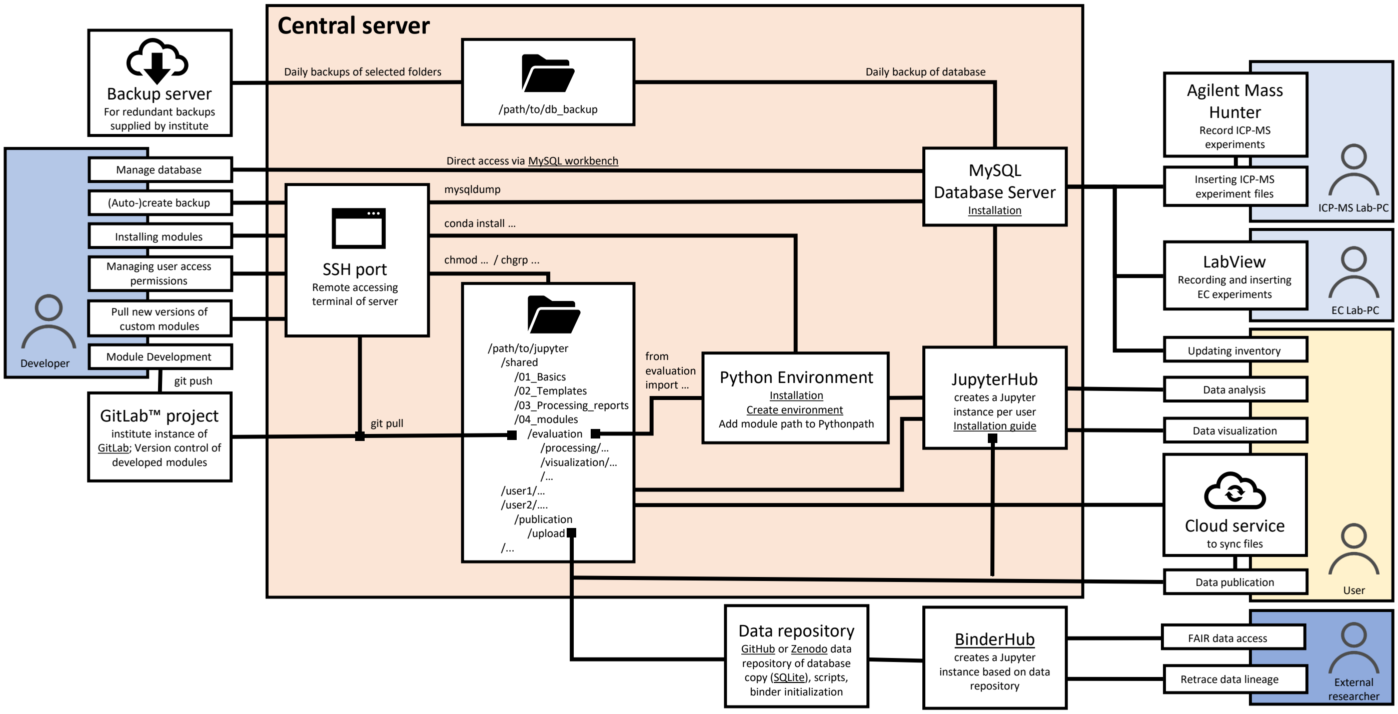

Figure S 6 Illustration of the server infrastructure and the software used in the presented data management tool.

References

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