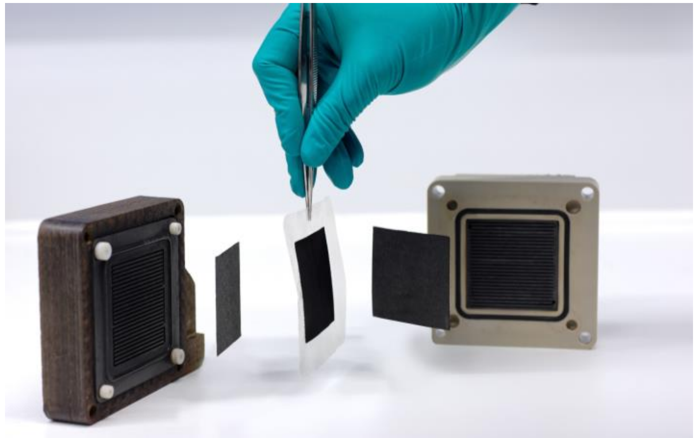
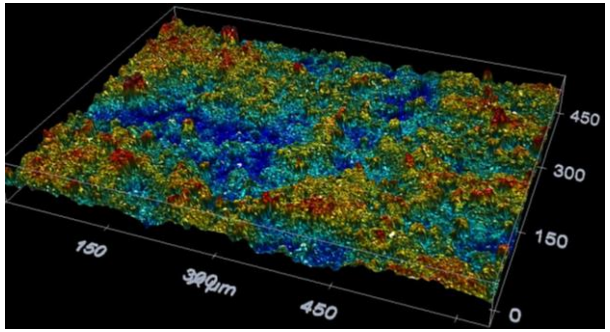
DEVELOPMENT OF MEMBRANE AND ELECTRODES FOR THE HT-PEM FUEL CELL TECHNOLOGY

J. Müller-Hülstede, D. Schonvogel, H. Niehoff, J. Buschermöhle, T. Zierdt, A. Schechterle, P. Wagner, K. A. Friedrich
Institute of Engineering Thermodynamics, German Aerospace Center (DLR)
25.09.2025

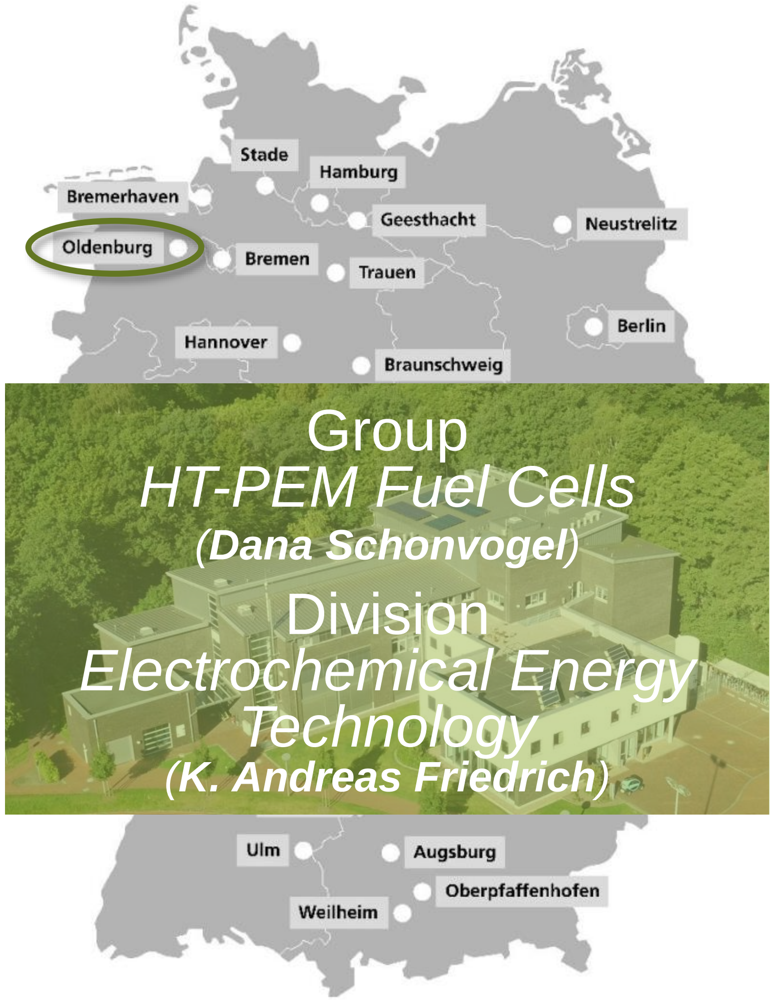
DLR Institute of Engineering Thermodynamics



Material Development
Long-term stable, efficient components for HT-PEM fuel cells
Catalysts, membranes, electrodes, membrane-electrode-assemblies

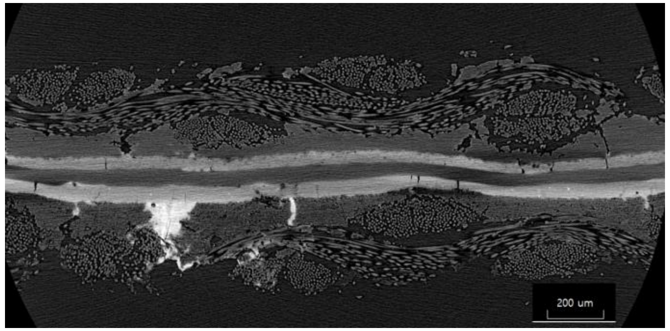


Analytics and Quality Control
On gas diffusion layers and bipolar plates after fabrication
Electrochemical, physico-chemical, imaging methods



Group
HT-PEM Fuel Cells
*(**Dana Schonvogel**)*
Division
Electrochemical Energy Technology
*(**K. Andreas Friedrich**)*

Performance Studies
From thin-film analysis to PEM fuel cells
Activity and degradation, contamination effects, accelerated stress tests

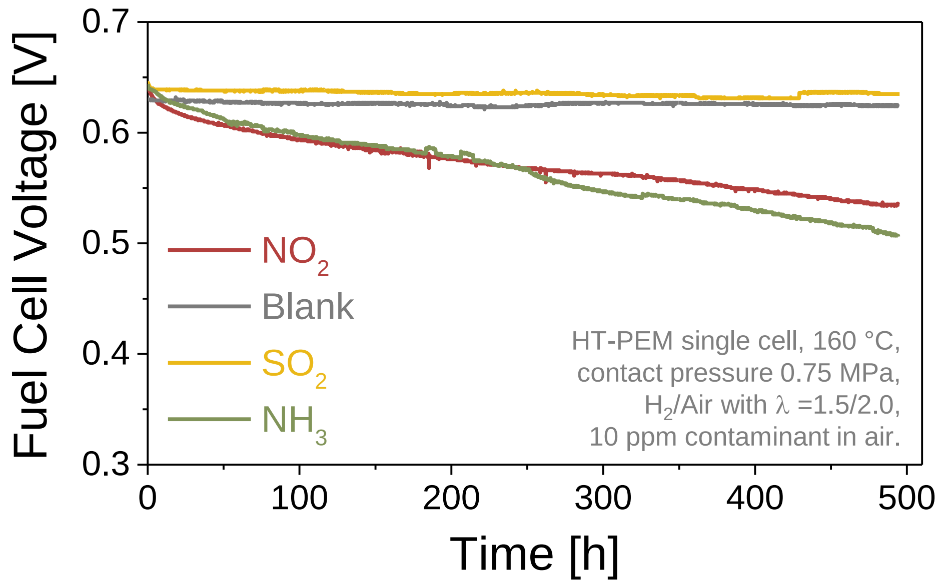


Cost-efficient Electrodes
for PEM fuel cells
Reduced PGM-contents, use of M-N-C catalysts

HT-PEM Fuel Cells

Advantages

- Increased tolerance towards contaminants like CO or H_2S due to **160 °C**
- Direct use of industrial quality H_2 or reformates
  → **Application flexibility**



Challenges

- Larger stack sizes compared to LT-PEMFCs
- Limited lifetime due to membrane creeping and corrosion of components
- Phosphate poisoning of catalytic active sites
  → Higher Pt loading of **up to 1 $mg_{Pt}cm^{-2}$ per electrode**
- PTFE contents of 10-40 wt.% (CL), 20-40 wt.% (MPL), 0-30 wt.% (GDL)

Increasing membrane stability
Reducing PGM-contents

D. Schonvogel et al., J. Power Sources **2021**, *High temperature polymer electrolyte membrane fuel cell degradation provoked by ammonia as ambient air contaminant*, 109, 401.
D. Schonvogel et al., Int. J. Hydrog. Energy **2021**, *Impact of air contamination by NOx on the performance of high temperature PEM fuel cells*, 46, 33934.
D. Schonvogel et al., Int. J. Hydrog. Energy **2021**, *Effect of air contamination by sulfur dioxide on the high temperature PEM fuel cell*, 46, 6751.

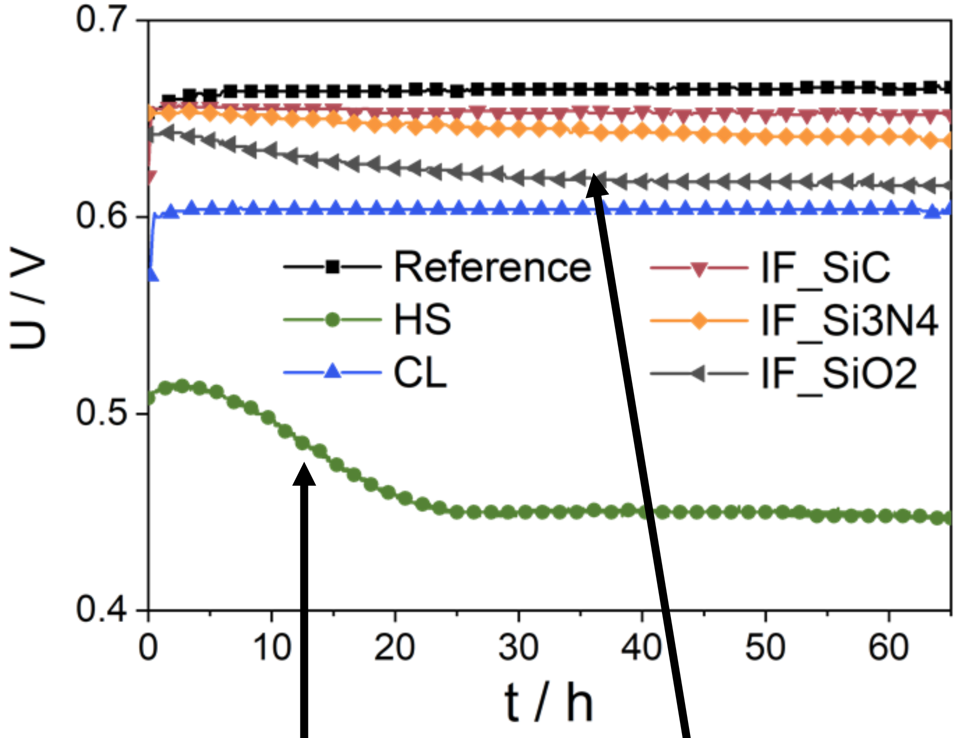
HT-PEM FC – MEMBRANES

Effect of Membrane Modification on Performance

- Collaboration with BASF → Celtec®-based study
  - Standard Celtec®-P as reference
    - Use of Celtec®-based fabrication process
    - Membranes with 2 wt% inorganic filler, crosslinked or increased solid-content

Membrane

Membrane	Solid content
Celtec®-P	5 wt%
SiO_2 Filler	6-8 wt%
Si_3N_4 Filler	6-8 wt%
SiC Filler	6-8 wt%
Crosslinked	6-8 wt%
High-solid	18 wt%



HT-PEM single cell testing: 160 °C, contact pressure 0.75 MPa, dry H_2/Air (1.5/2.0).

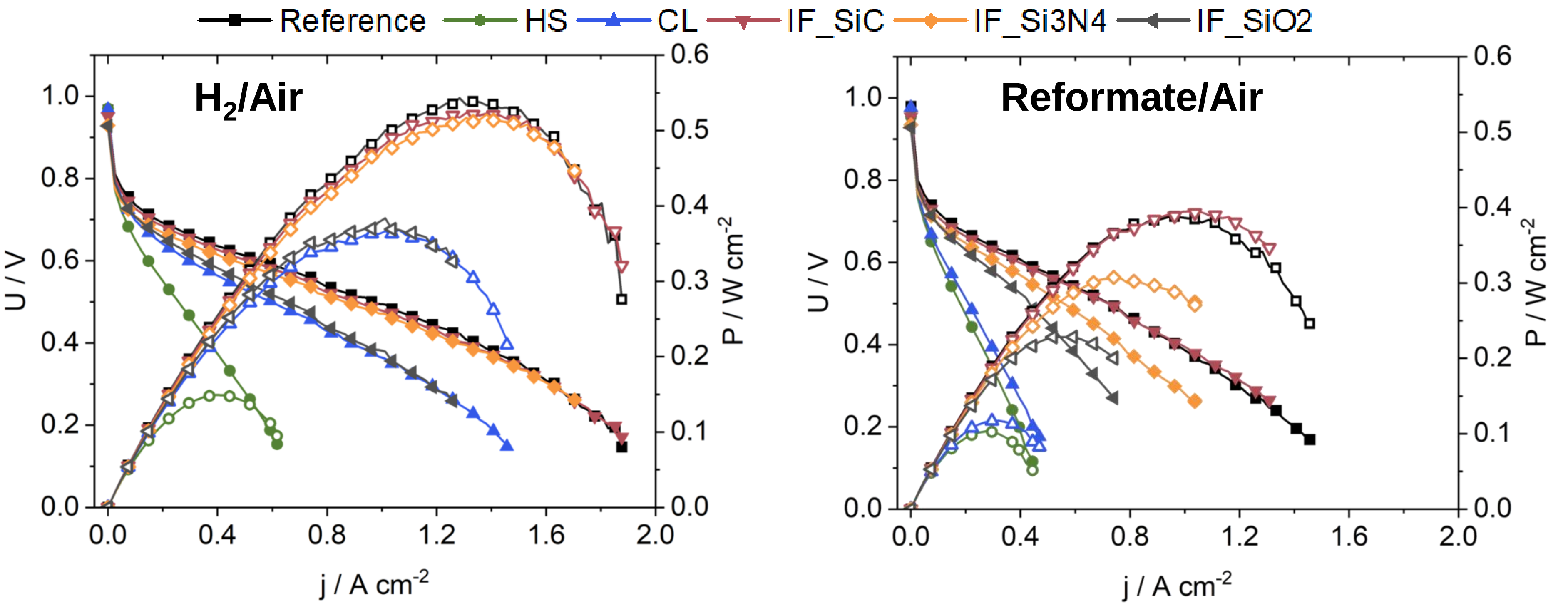
Irreversible acid leaching from membrane

Hydrophilic character of SiO_2 limits acid distribution

J. Müller-Hülstede et al., ChemSusChem **2025**, *Performance of High Temperature PEM Fuel Cells as a Function of PBI Membrane Modification*, under revision.

Effect of Membrane Modification on Performance

- Best performance for silicon carbide as filler in HT-PEM
- Lowest performances for high-solid and crosslinked membranes



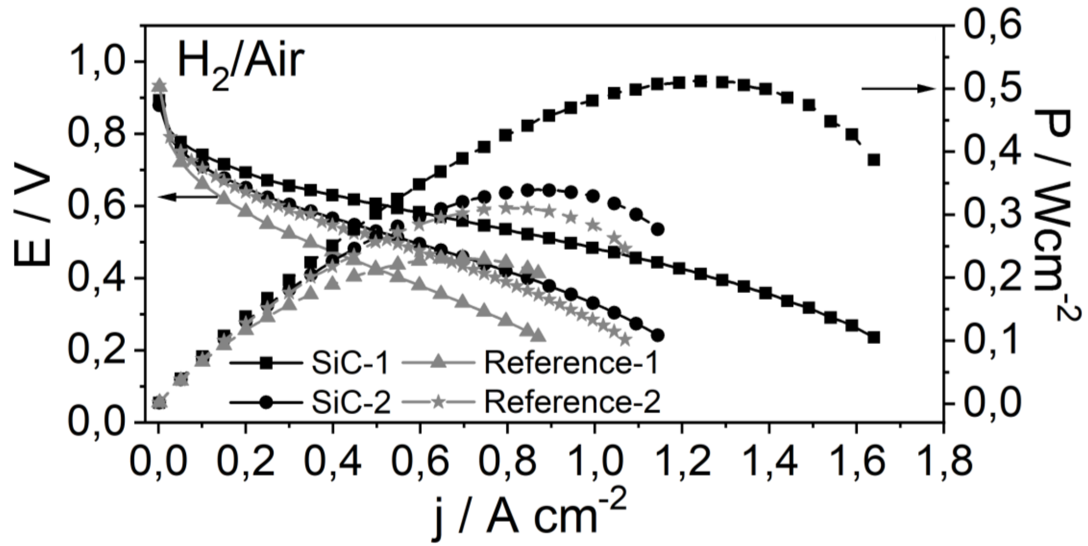
HT-PEM single cell testing: 160 °C, contact pressure 0.75 MPa, dry gases.

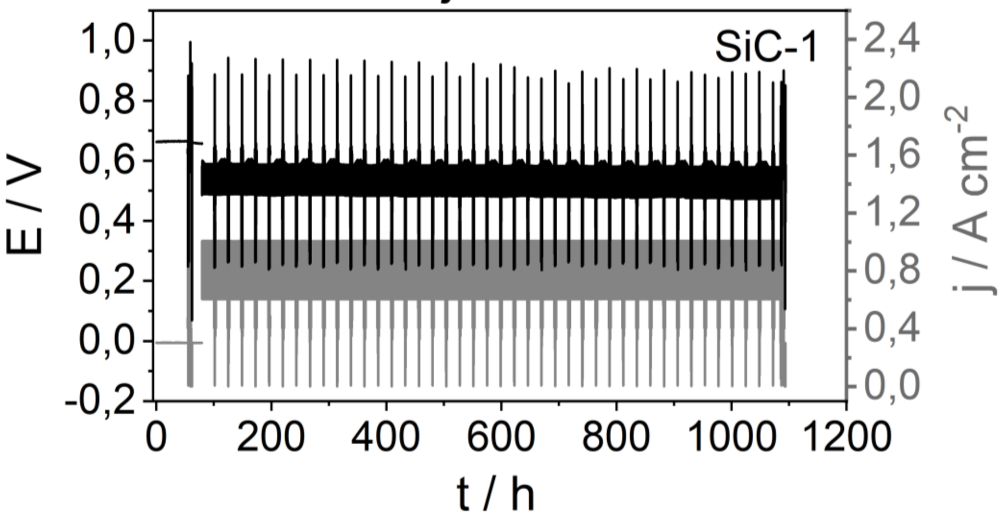
→ Next step: Long-term stability of SiC membrane

J. Müller-Hülstede et al., ChemSusChem **2025**, *Performance of High Temperature PEM Fuel Cells as a Function of PBI Membrane Modification*, under revision.

Silicon Carbide based HT-PEM Membranes
Degradation over Time

- Test of two particle diameters
  - SiC-1: 0.1–1.0 µm
  - SiC-2: 2.0 µm
- 1,000 h of load cycling
  - 4 min at 0.6 A cm^{-2} & 16 min at 1.0 A cm^{-2}
  - SiC-based lower degradation rates (<65 µV h^{-1} for SiC, >100 µV h^{-1} for Celtec)
  - <u>But</u>: Lower OCVs in case of SiC
- Lower acid losses in case of SiC



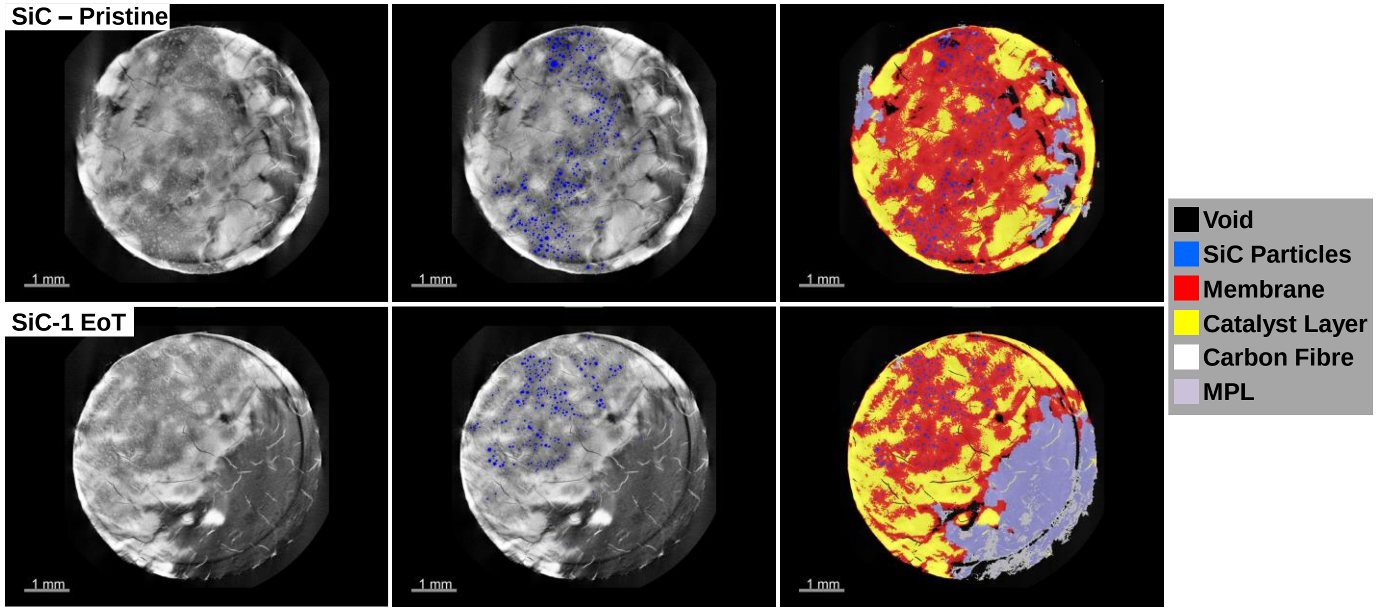


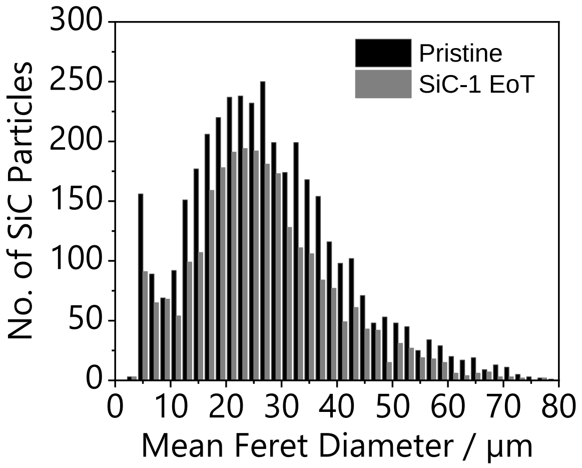
HT-PEM single cell testing: 160 °C, contact pressure 0.75 MPa, dry H_2/Air (1.5/2.0).

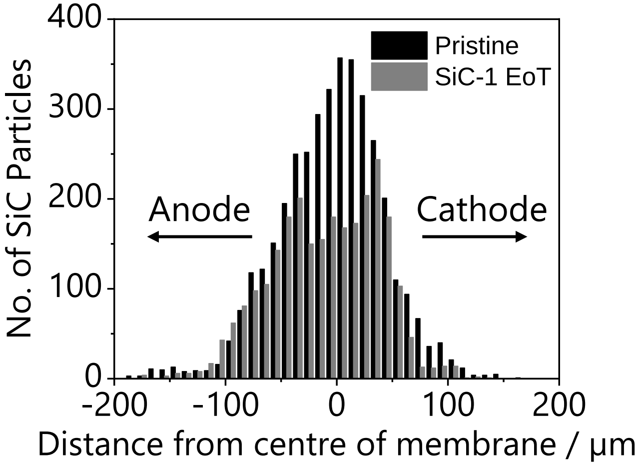
D. Schonvogel et al., J. Power Sources **2024**, *Performance and durability of high temperature proton exchange membrane fuel cells with silicon carbide filled polybenzimidazole composite membranes*, 591, 233835.

Silicon Carbide based HT-PEM Membranes
Computer Tomography with Machine Learning

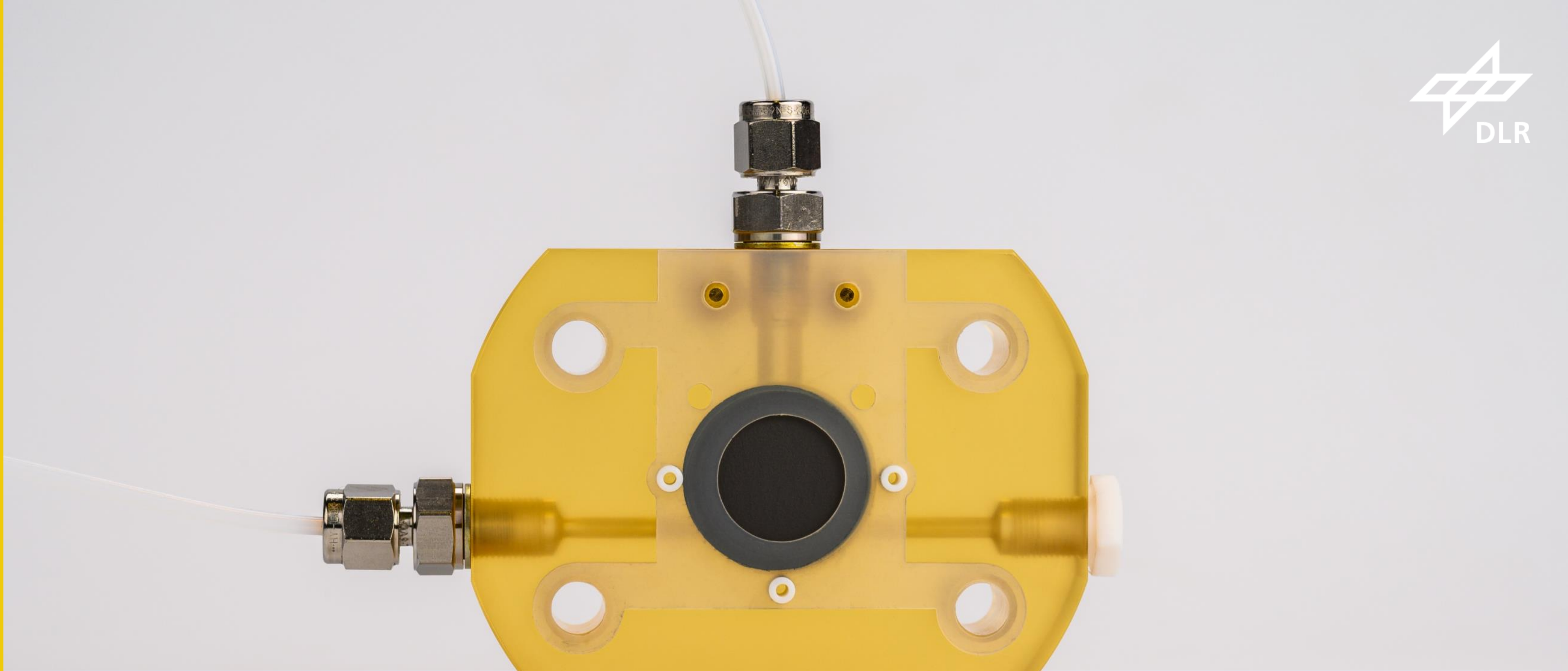
- Cooperation with UNSW Sidney
- Lower membrane thinning using SiC
- Evidence of mobility and redistribution of SiC particles







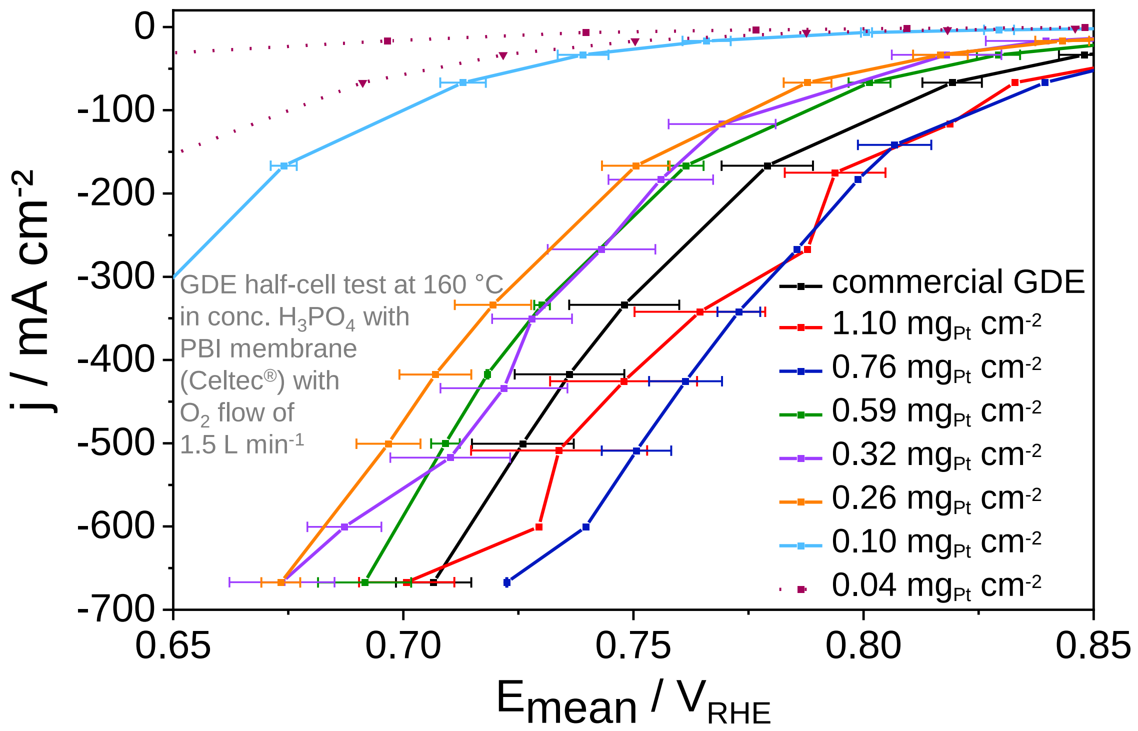
D. Schonvogel et al., J. Power Sources **2024**, *Performance and durability of high temperature proton exchange membrane fuel cells with silicon carbide filled polybenzimidazole composite membranes*, 591, 233835.

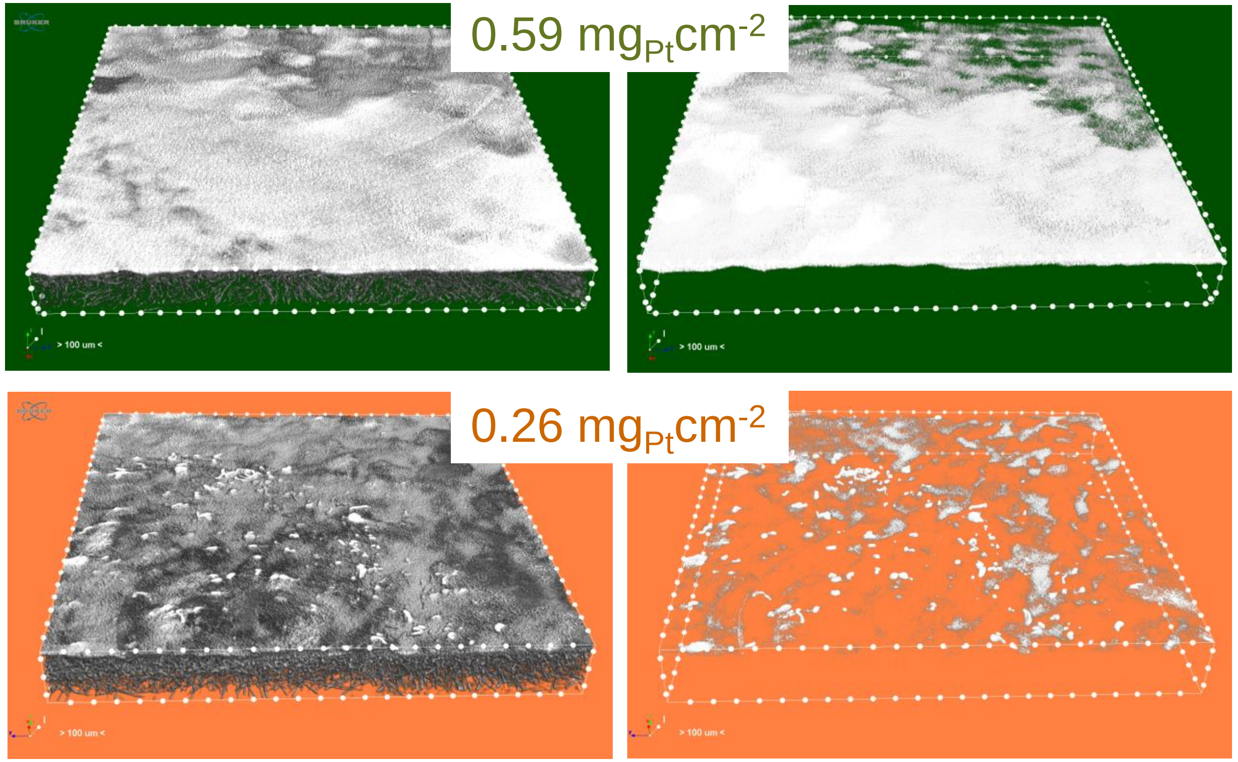


HT-PEM FC – ELECTRODES

HALF-CELL AND SINGLE CELL STUDIES

Reduction of Platinum Contents in HT-PEM Electrodes





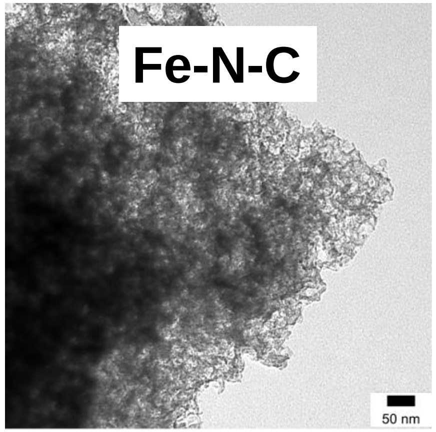
µ-Computed tomographic images of the whole GDEs (left) and catalyst layers only (right).

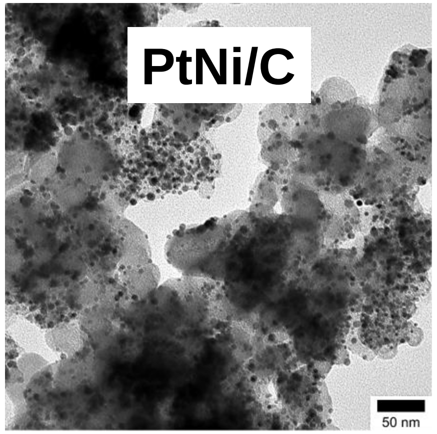
→ **Simple reduction of Pt-loading limited**
→ **Next step: Hybrid HT-PEM electrodes**

H. Schmies et al., J. Power Sources **2022**, *Reduction of platinum loading in gas diffusion electrodes for high temperature proton exchange membrane fuel cell application: Characterization and effect on oxygen reduction reaction performance*, 529, 231276.

Reduction of Platinum Contents in HT-PEM Electrodes
Hybrid HT-PEM Electrodes

- Pt content reduction through incorporation of catalytic active filler
  → Studying the effect of Fe-N-Cs in Pt-alloy cathodes



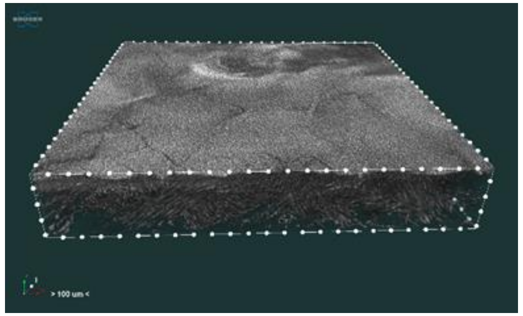


10 wt%

+ 40 wt% PTFE

50 wt%

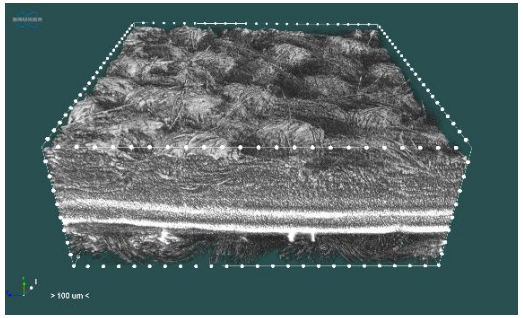
Hybrid Gas Diffusion Electrode (GDE)



Ultrasonic spray coating of cathode on woven Celtec®-based gas diffusion layers followed by sintering at 350 °C for 10 minutes in N_2 atmosphere

Standard: 0.85 mg_{Pt} cm^{-2}
GDE 1: 0.40 mg_{Pt} cm^{-2}
GDE 2: 0.65 mg_{Pt} cm^{-2}

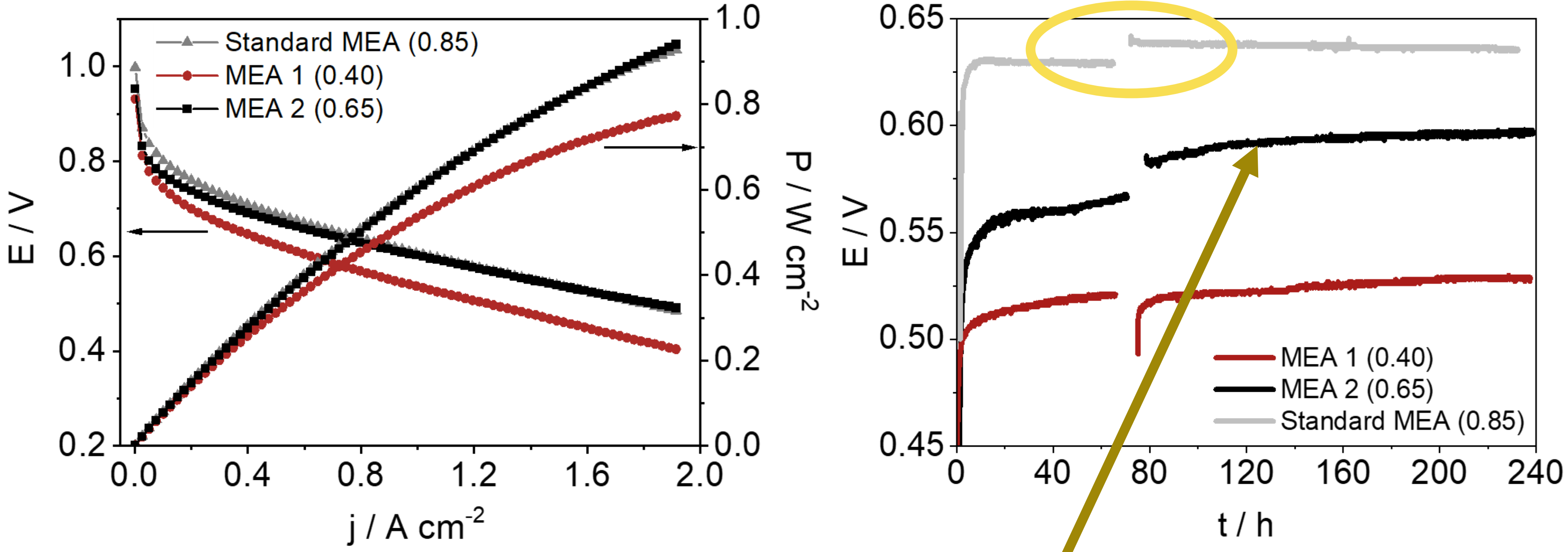
Membrane-Electrode-Assembly (MEA)



MEA-assembling analogous to Celtec®-technology with active area of 20.25 cm² and reduced Pt-content on cathode site

J. Müller-Hülstede et al., ChemSusChem **2023**, *Towards the Reduction of Pt Loading in High Temperature Proton Exchange Membrane Fuel Cells – Effect of Fe-N-Cs in Pt-alloy Cathodes*, e202202046.

Reduction of Platinum Contents in HT-PEM Electrodes
Hybrid HT-PEM Electrodes



HT-PEM single cell testing: 160 °C, contact pressure 0.75 MPa, Dry H_2/Air (1.5/2.0); polarization curves: H_2/O_2 (1.5/9.5)

Voltage increase in case of conventional Pt-based MEA immediately after electrochemical measurements

Slow voltage increase caused by electrolyte redistribution in presence of Fe-N-C

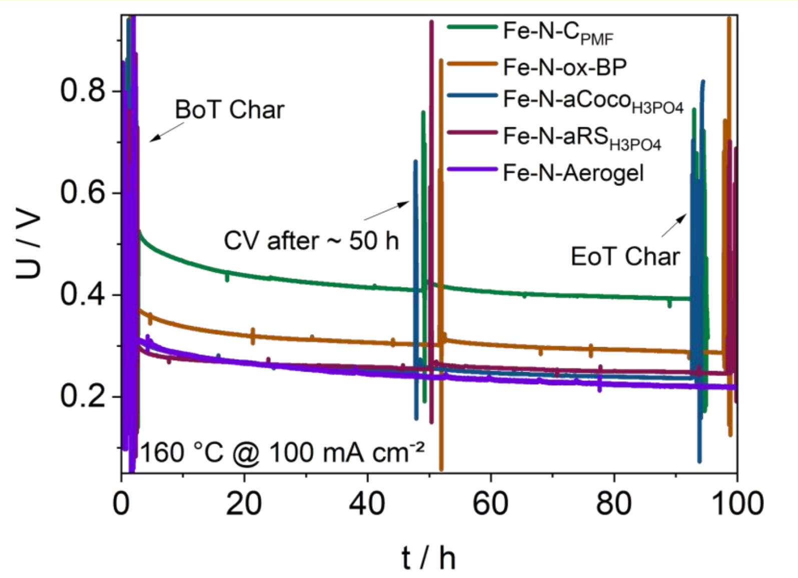
→ Improvement of catalyst layer needed

J. Müller-Hülstede et al., ChemSusChem **2023**, *Towards the Reduction of Pt Loading in High Temperature Proton Exchange Membrane Fuel Cells – Effect of Fe-N-Cs in Pt-alloy Cathodes*, e202202046.

PGM-Free Electrodes
Metal-Nitrogen-Carbon Catalysts (M-N-C)

Fe-N-C-based electrodes first studies

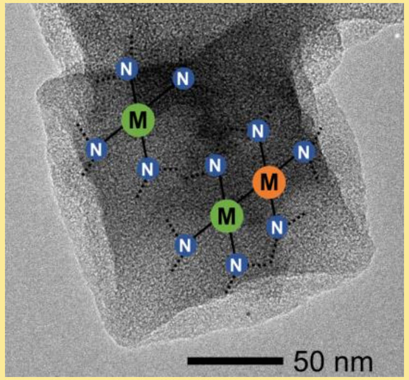
- Investigation of C-supports for Fe-N_x[1-4, 5]



➢ Performance limitations of support-based Fe-N-Cs in HT-PEMFC
➢ Insufficient stability in HT-PEMFC
➢ Inhomogeneous catalyst layer

Fe-N-C-based electrodes current studies

- Catalyst optimization
  - Bimetallic FeCo[6] and FeSn-N-Cs[7]

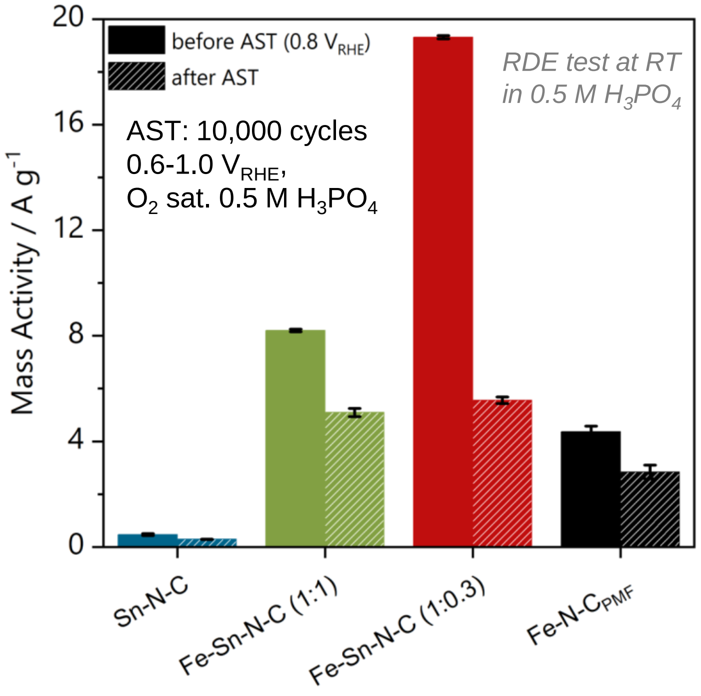


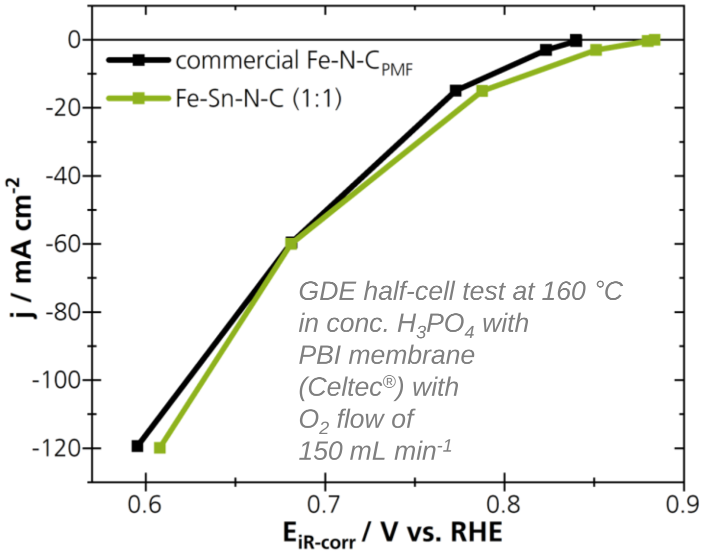
- Analysis of CL fabrication
  - Binder content[8]
  - Additives[9]
  - Drying method[10]

[1] J. Hülstede et al., Materials 2021,14, 1, 45.
[2] J. Müller-Hülstede et al., ACS Appl. Energy Mater. 2021, 4, 7, 6912.
[3] J. Müller-Hülstede et al., J. Power Sources 2022, 537, 231529.
[4] J. Müller-Hülstede et al., Int. Journal Hydrogen Energy 2024, 50, 921-930.
[5] T. Zierdt et al., ChemSusChem 2025, 18, e202401843.
[6] M. Mooste et al., Electrochimica Acta 2025, 514, 145620.
[7] J. G. Buschermöhle et al., ACS Catal. 2025, 15, 6, 4477–4488.
[8] T. Zierdt et al., ChemElectroChem 2024, 11, e202300583.
[9] T. Zierdt et al., ECS Trans. 2024, 114, 325.
[10] T. Zierdt et al., ChemSusChem. 2025, 18, e202500905.

Bimetallic Fe-Sn-N-C Catalysts

- MOF-based synthesis with one pyrolysis step
  - Two metal ratios Fe:Sn (1:1 and 1:0.3)
  - Fe-loadings between 0.7 and 1.0 wt.%
- Activity and stability
  - In diluted H_3PO_4 enhanced mass activity for Fe-Sn-N-Cs
    - Beneficial effect of small amounts of Sn
  - In HT-PEM GDE half-cell higher OCV and comparable performance to commercial Fe-N-C
    - Catalyst layer fabrication has strong impact on performance



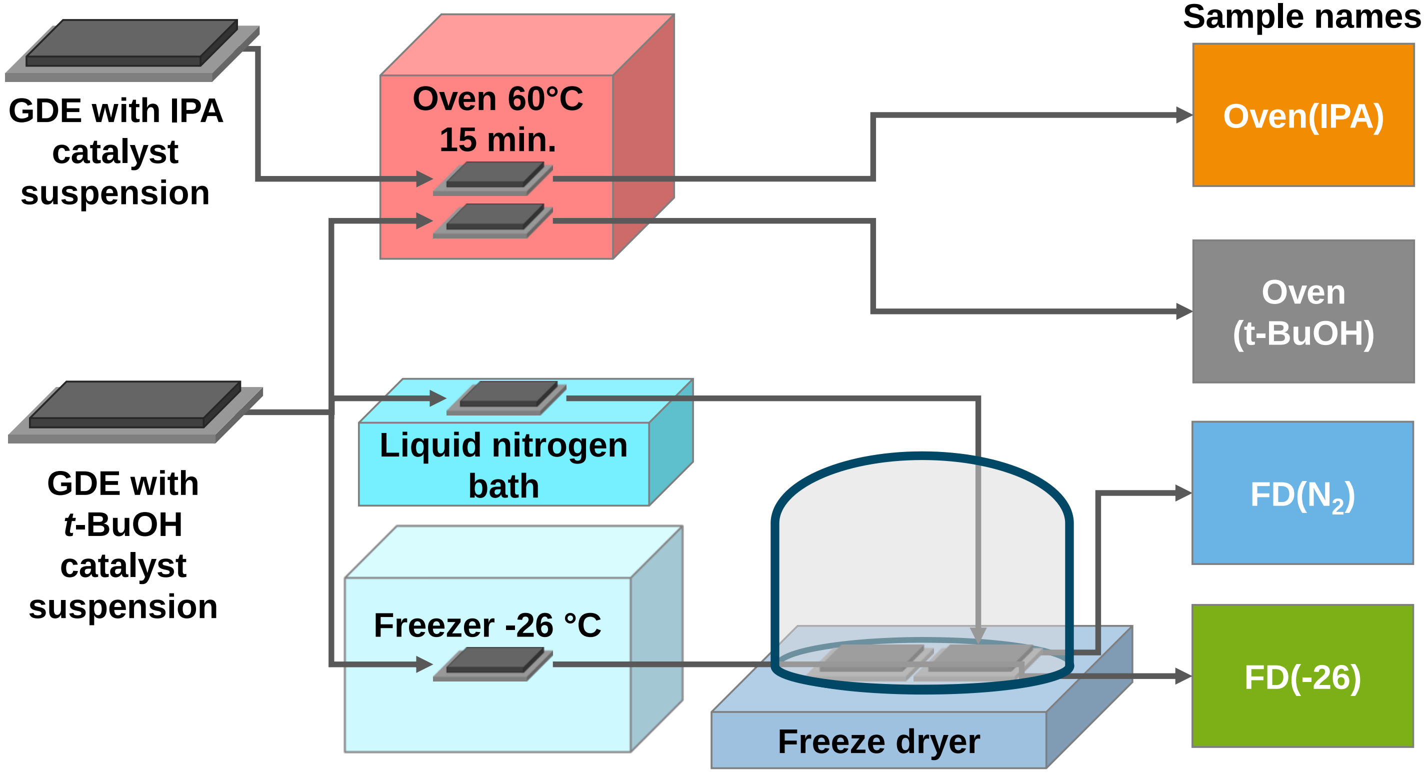


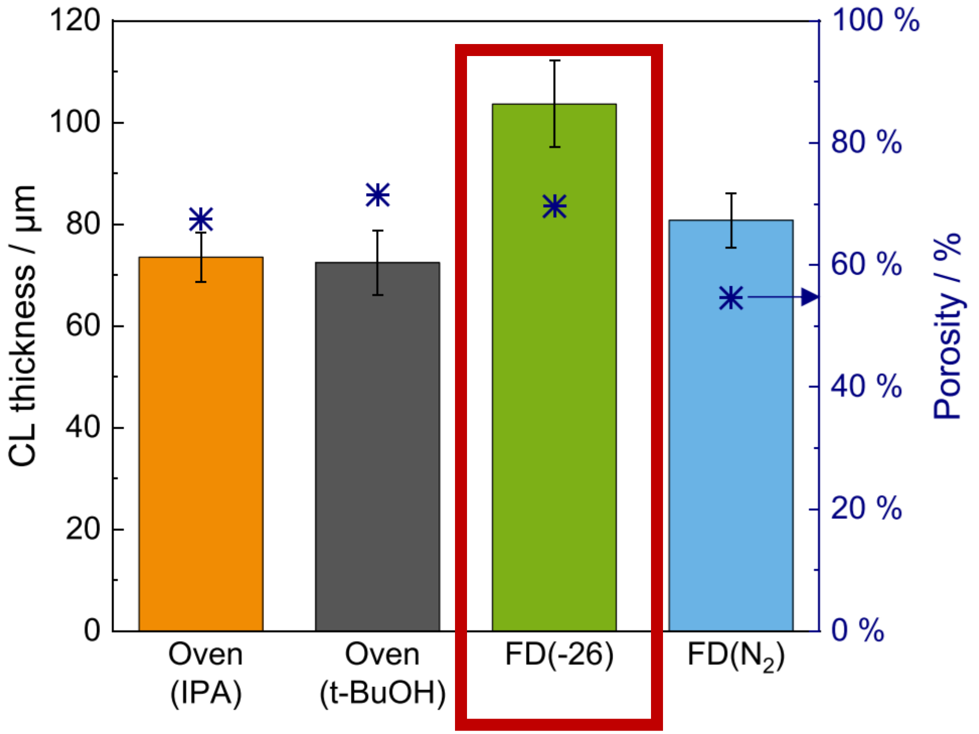
→ Further structural analysis of catalyst (*in-situ*) needed
→ Optimization of catalyst layer fabrication required

J. G. Buschermöhle et al., ACS Catal. **2025**, *Fe–Sn–N–C Catalysts: Advancing Oxygen Reduction Reaction Performance,* 15, 6, 4477–4488.

Optimization of Fe-N-C-based Catalyst Layers
Freeze-Drying vs. Oven-Drying

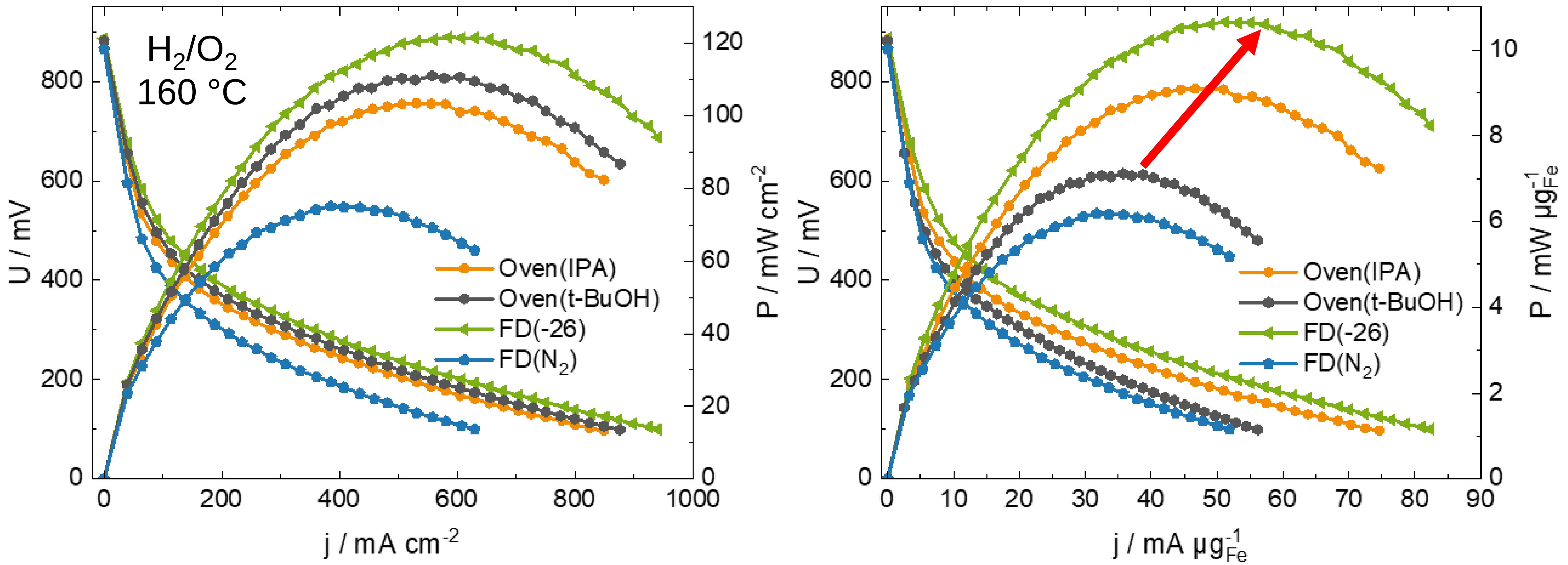
- Beneficial effect of freeze-drying on Pt-based CLs in CCMs for LT-PEMFC[1]
- Transferable to Fe-N-C-based CLs using commercial PMF-011904?[2]





[1] K. Talukdar, S.Delgado, T. Lagarteira, P. Gazdzicki, K. A. Friedrich, J. Power Sources 2019, 427, 309.
[2] T. Zierdt, et al., ChemSusChem. 2025, 18, e202500905.

Optimization of Fe-N-C-based Catalyst Layers
HT-PEMFC Performance



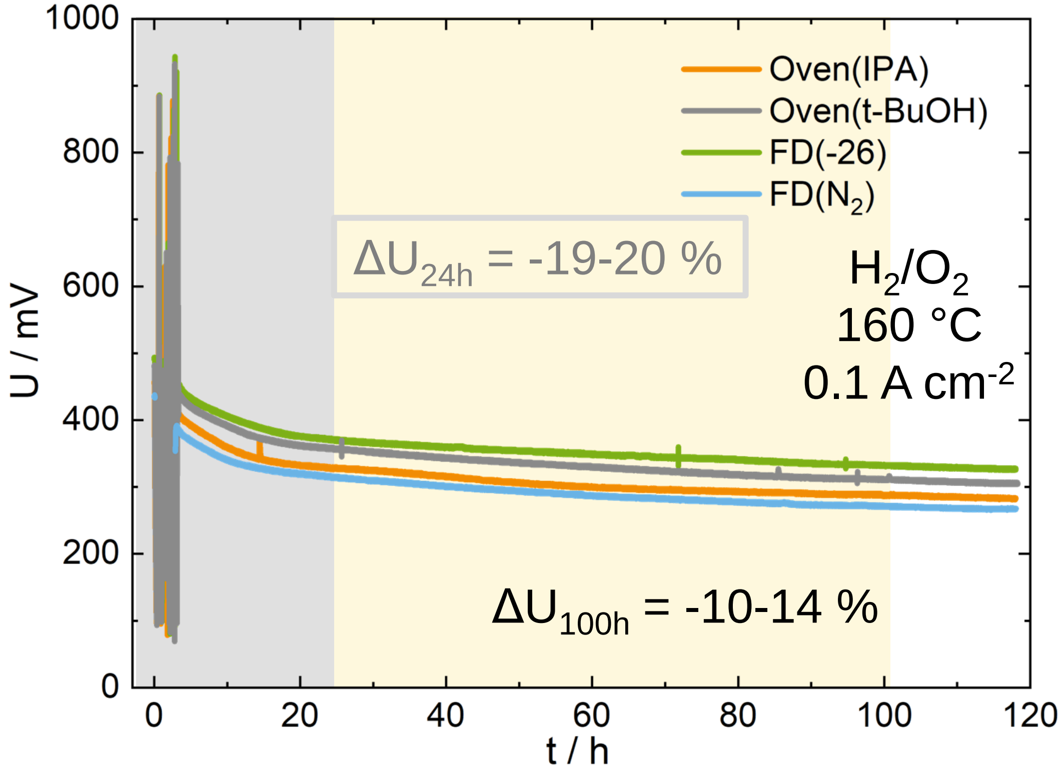
- Increase of peak power density by up to 45 % through freeze-drying of CL
  - Possibility of improved accessibility of active sites

T. Zierdt et al., ChemSusChem. **2025**, *Influence of Ink Composition and Drying Technique on the Performance and Stability of Fe–N–C-Based High-Temperature Proton Exchange Membrane Fuel Cells,* 18, e202500905.

Optimization of Fe-N-C-based Catalyst Layers

Stability over 100 h

- Stability for 100 h at 0.1 A cm^{-2}
  - Similar voltage losses independent of CL fabrication method
  - Voltage losses assumed due to formation of reactive oxygen species typically for Fe-N-Cs



→ Catalyst layer fabrication negligible effect on stability
→ Stabilisation of active Fe-N_x sites required

T. Zierdt et al., ChemSusChem. **2025**, *Influence of Ink Composition and Drying Technique on the Performance and Stability of Fe–N–C-Based High-Temperature Proton Exchange Membrane Fuel Cells,* 18, e202500905.

Outlook

Next to material development…

- **Quality control of gas diffusion electrodes using realistic operation conditions**
  - Broad range (HOR, HER, OER, ORR, CO2RR, NRR)
  - RT-160 °C and 1-6 bara
  - Testing of advanced measuring setup at DLR

TheMes

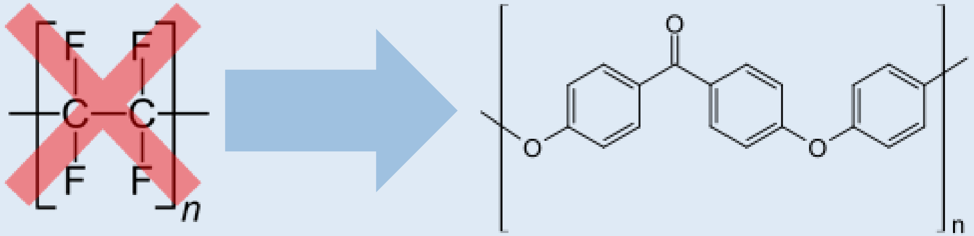
Federal Ministry
for Economic Affairs
and Energy

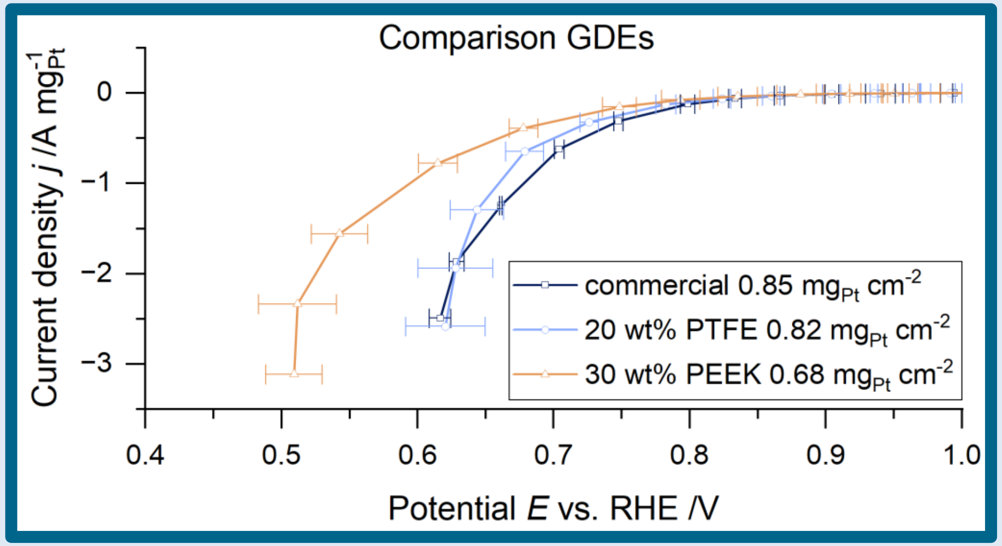
ZIM
Zentrales
Innovationsprogramm
Mittelstand

Outlook

Next to material development…

- Quality control of gas diffusion electrodes using realistic operation conditions
- **Replacement of PFAS in HT-PEMFC**
  - Implementation of PFAS-free polymers
  - Potential for fast up-scaling



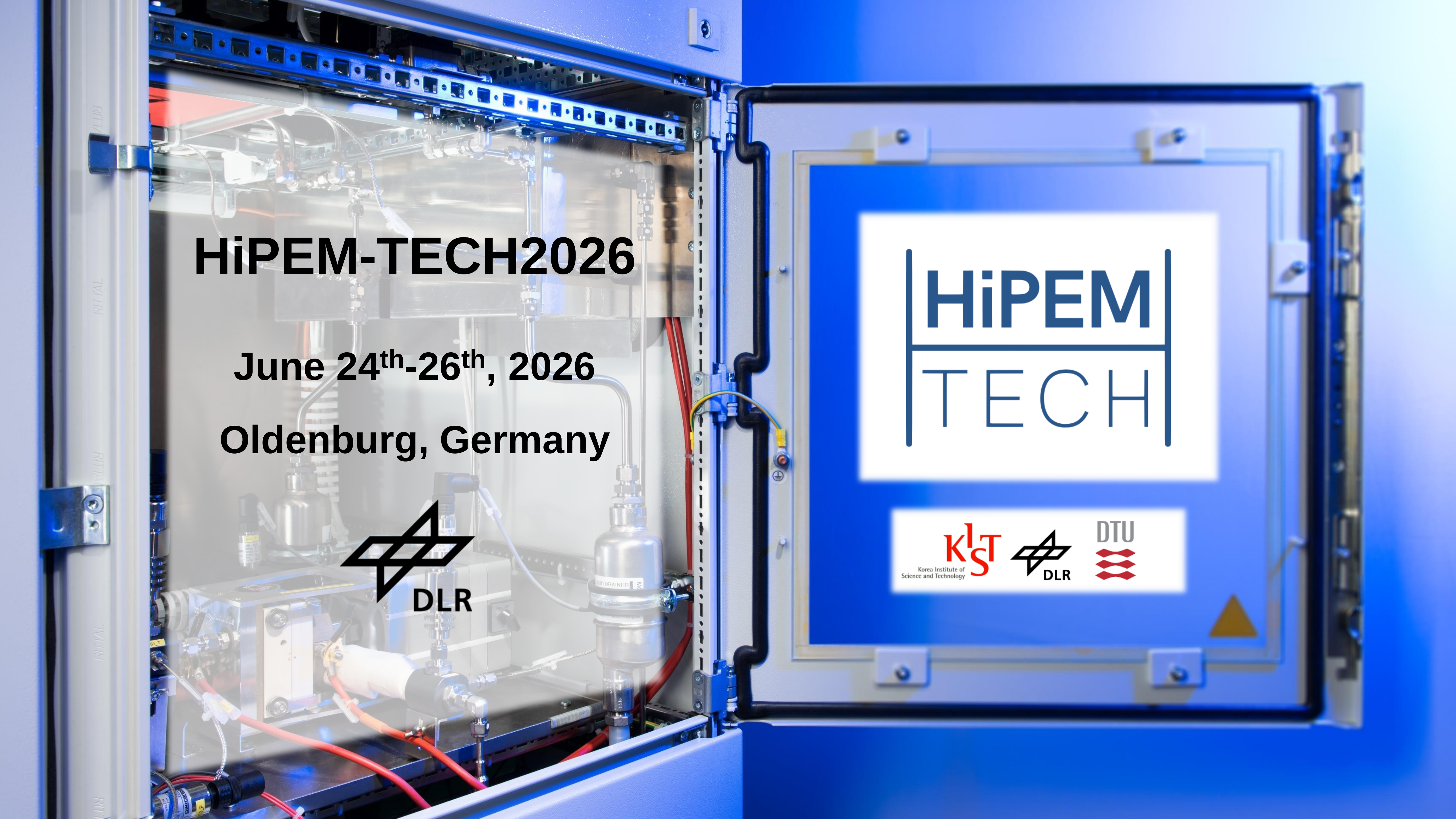


GDE half-cell test at 160 °C in conc. H_3PO_4 O_2 flow of 150 mL min^{-1}

Outlook

Next to material development…

- Quality control of gas diffusion electrodes using realistic operation conditions
- Replacement of PFAS in HT-PEMFC
- **HT-PEM fuel flexibility for case of disasters and civil protection**
  - Containerized fuel cell system – capable of running on both methanol and hydrogen
  - Coordination and multi-fuel switching and testing


HiPEM-TECH2026
June 24th-26th, 2026
Oldenburg, Germany
DLR
HiPEM
TECH
KIST
Korea Institute of Science and Technology
DLR
DTU

Thank you for your attention!

DLR

Acknowledgments

Federal Ministry of Research, Technology and Space

BASF We create chemistry

TRIGONA FUEL CELL COMPONENTS

CARL VON OSSIETZKY UNIVERSITÄT OLDENBURG

Federal Ministry for Economic Affairs and Energy

JÜLICH FORSCHUNGSZENTRUM

UNSW THE UNIVERSITY OF NEW SOUTH WALES

Leibniz Universität Hannover

CERTH CENTRE FOR RESEARCH & TECHNOLOGY HELLAS

Universität Bremen

gaskatel

ADVENT

DTU

PROACT

Universität Stuttgart Institut für Chemische Verfahrenstechnik

Contact: Dana.Schonvogel@dlr.de