09242: Systematic Optimization of Pt-free Gas Diffusion Electrodes for HT-PEMFC Application

Tanja Zierdt, Julia Müller-Hülstede, Henrike Schmies, Dana Schonvogel, Peter Wagner, K. Andreas Friedrich

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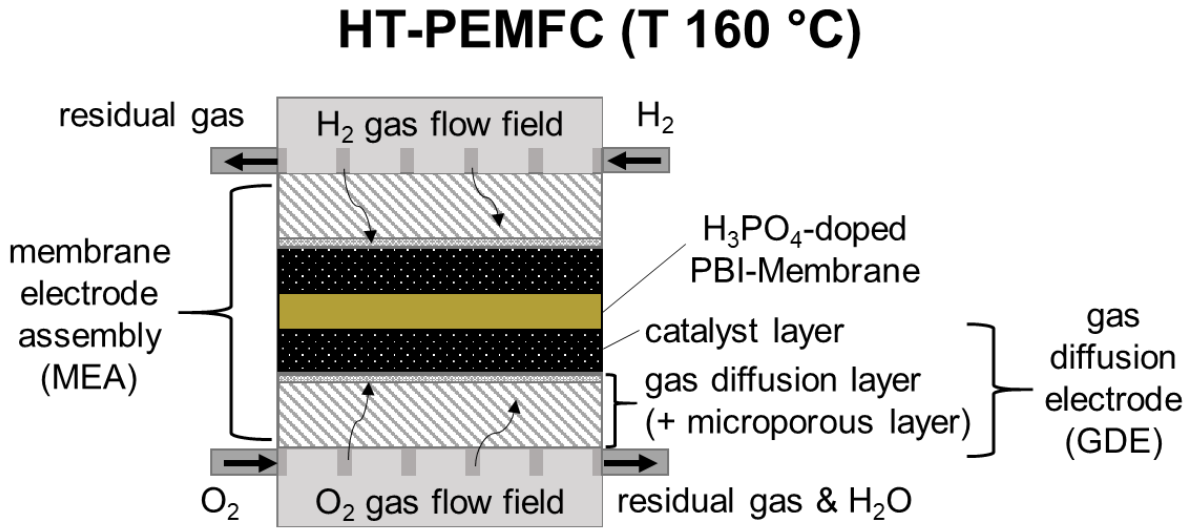
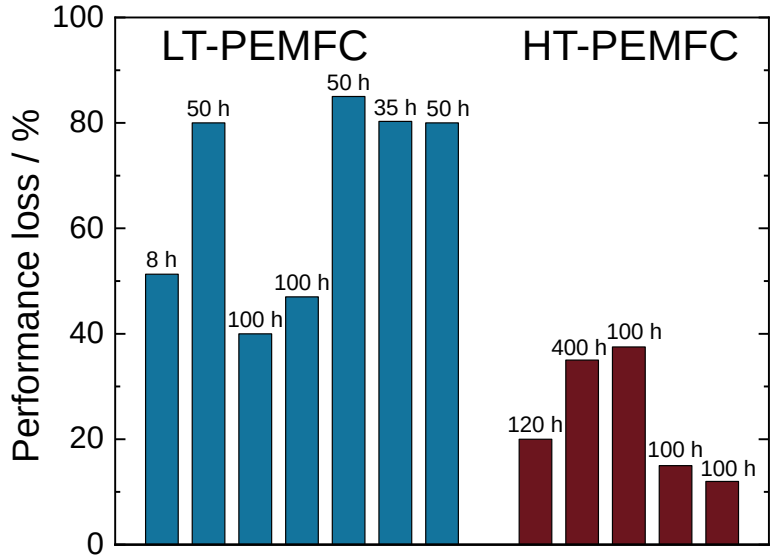
DLR

High temperature proton exchange membrane fuel cell (HT-PEMFC)

- No complex water management and increased reaction kinetics
- Pt catalyst suffers from phosphate ion poisoning, enforcing loading up to 1 $mg_{Pt}\,cm^{-2}$ → **high material costs**[1]
- **Fe-N-C** as promising alternative
  - ➢ Sufficient performance
  - ➢ Phosphate tolerance
  - ➢ Cost-efficient
  - ➢ Fast performance loss
  - ➢ Lower volumetric activity[2]





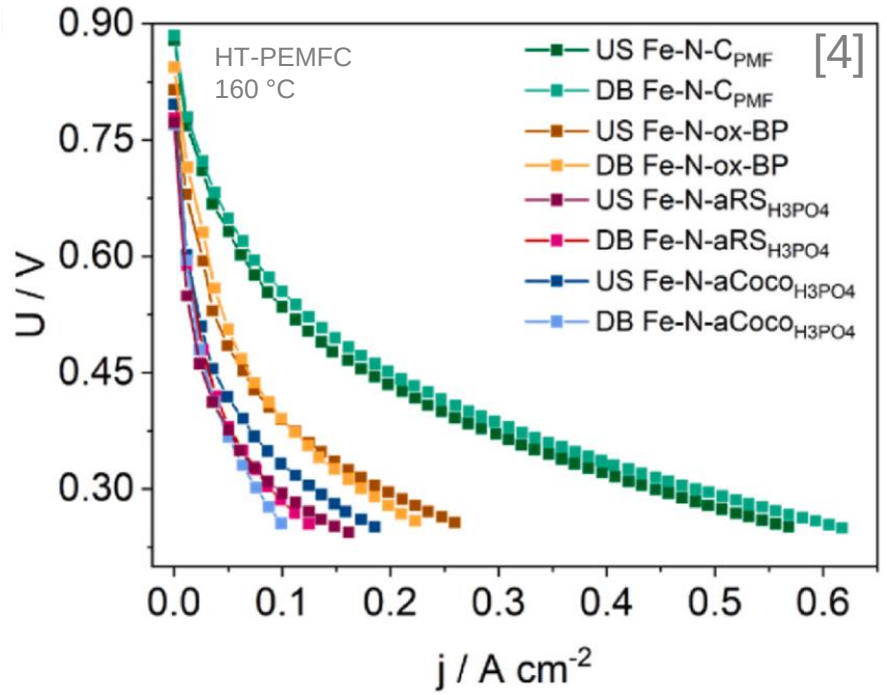
Performance of Fe-N-C-based catalyst. Inspired by [1]

[1] Meyer, Q., et al., *Electrochemical Energy Reviews*, 2023. 6(1):16, p. 2.
[2] Müller-Hülstede, J., Zierdt, T., et al., *Journal of Power Sources*, 2022. 537: p. 231529.

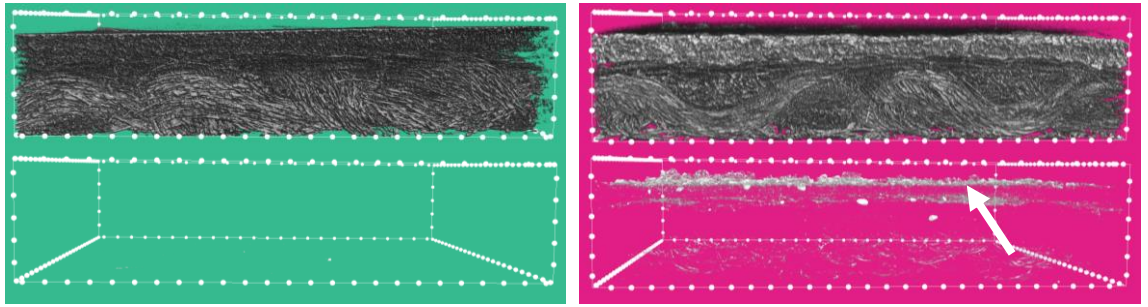
Our previous research and approach

- **Reduction of Pt loading** and varied **PTFE content** (catalyst layer binder) for HT-PEMFC cathodes[1]
- Optimization of **carbon support** for optimal Fe-N_x incorporation[2, 3]
- HT-PEMFC **stability** of different Fe-N-C catalysts (Poster B0209)
- HT-PEMFC performance of **different Fe-N-C based MEAs**[4]
  - ➢ Low impact of coating method
  - ➢ Strong impact of GDE fabrication & ink stability

→ Approach: Catalyst layer optimization to unfold full potential of Fe-N-C catalyst: investigation of **different PTFE** contents



Fe-N-C$_{PMF}$ **Fe-N-aRS$_{H3PO4}$**



Doctor blade (DB) coating [4]

[1] Schmies, H., Zierdt, T. et al., *Journal of Power Sources*, 2022. 529: p. 231276.
[2] Hülstede, J., et al., *Materials*, 2021. 14(1): p. 45 (1-19).
[3] Müller-Hülstede, J., *ACS Appl. Energy Mater*, 2021, 4(7): p. 6912-6922.
[4] Müller-Hülstede, J., Zierdt, T., et al., *Journal of Power Sources*, 2022. 537: p. 231529.

GDE fabrication and characterization

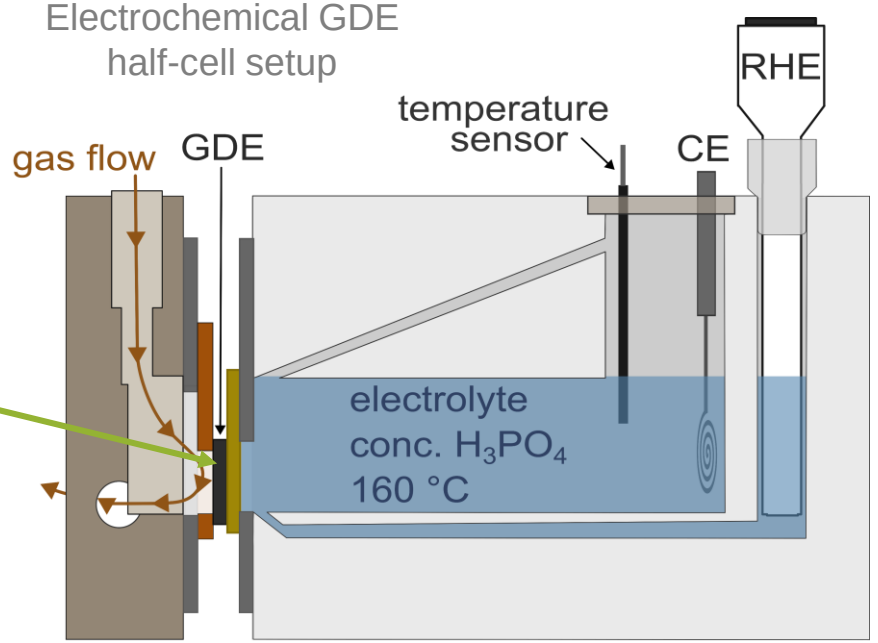
Catalyst suspension:
- Commercial Fe-N-C (PMF-0011904, Pajarito Powder)
- H_2O
- 2-Propanol
- PTFE (different amounts)

Dispersed by ultrasonication

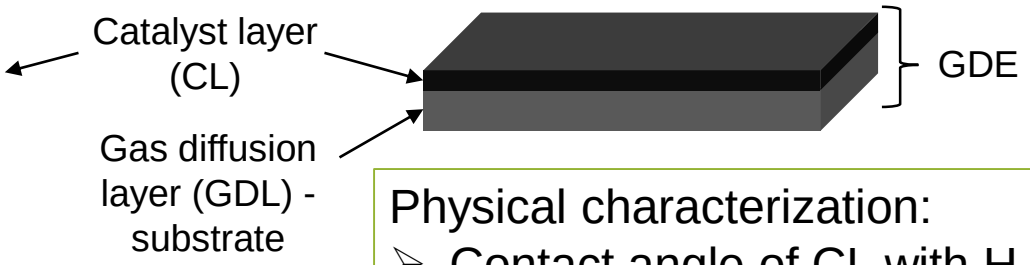
Ultrasonic spray coating

Hot pressing
polybenzimidazole (PBI) membrane
1 kN, 140 °C, 30 s

Electrochemical GDE half-cell setup

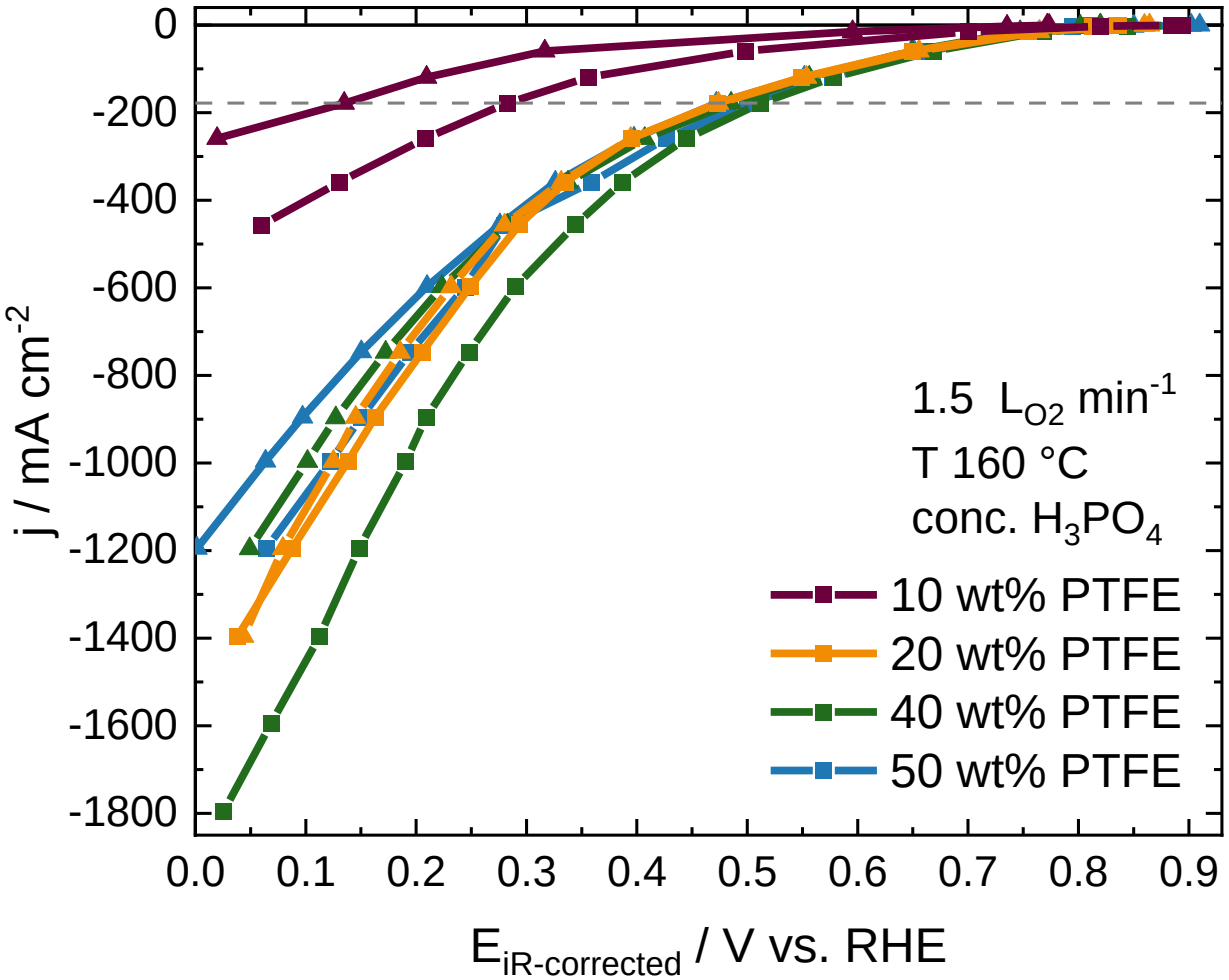


Coated layers	PTFE content	PTFE/catalyst ratio
	(in dry catalyst layer)	
168	10 wt%	0.11
188	20 wt%	0.25
240	40 wt%	0.68
298	50 wt%	0.11

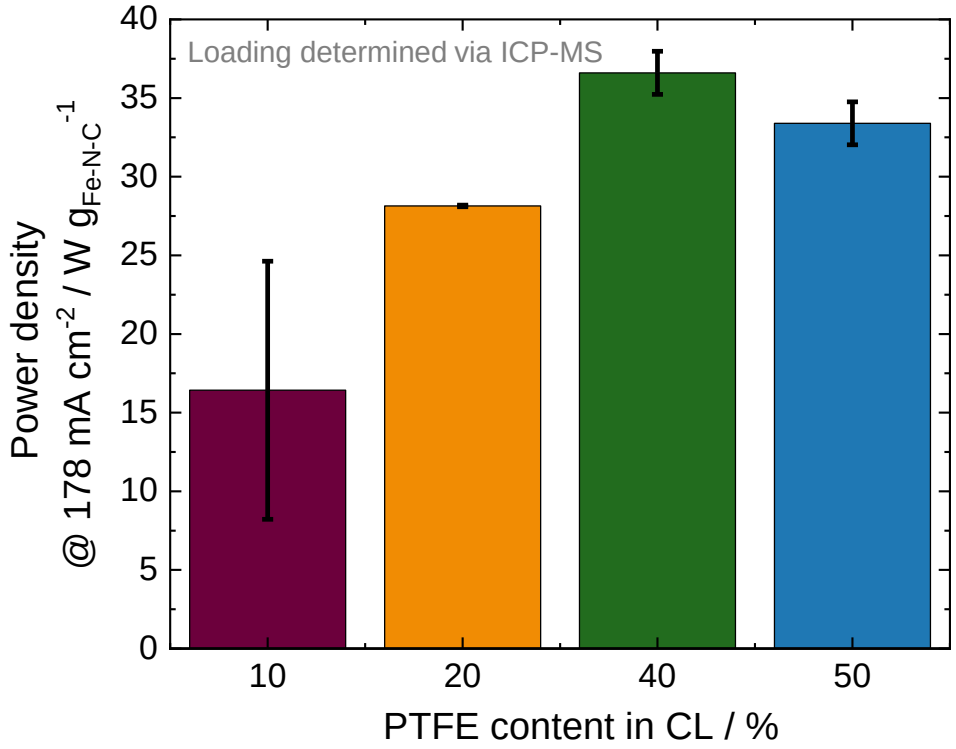


Physical characterization:
- Contact angle of CL with H_2O and H_3PO_4
- Micro-computed tomography (µ-CT)
- Inductively coupled plasma mass spectrometry (ICP-MS)

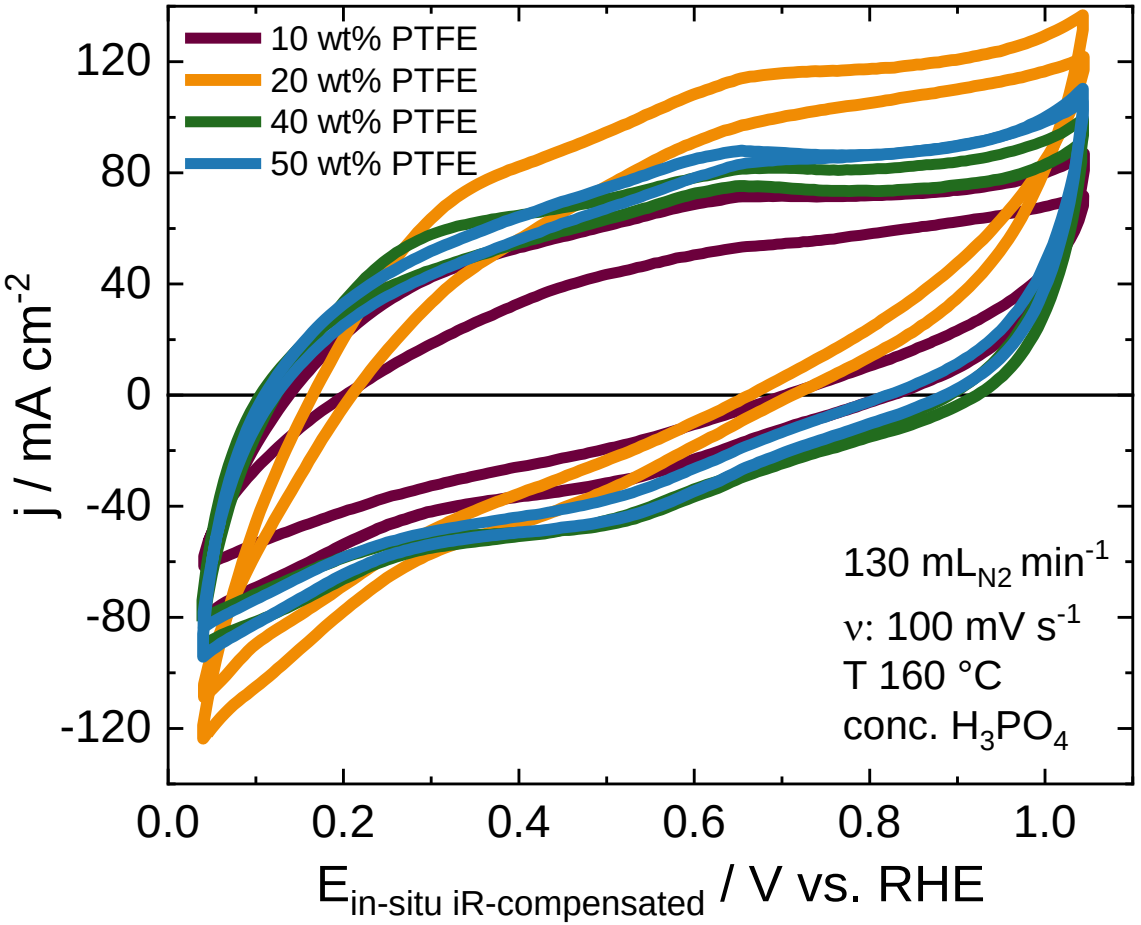
Effect of PTFE content on ORR performance

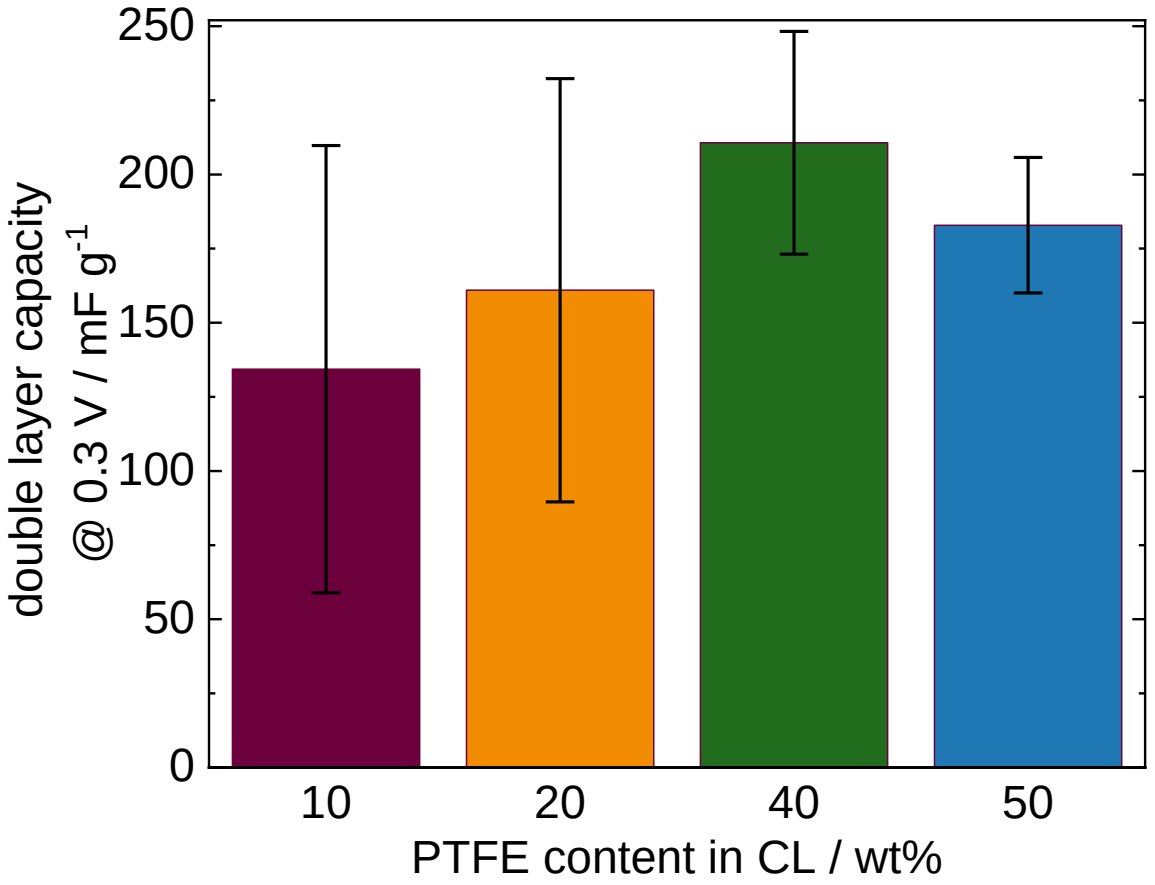


- 20, 40 and 50 wt% PTFE GDE: similar performance
- 10 wt% PTFE GDE: lowest performance and highest deviations



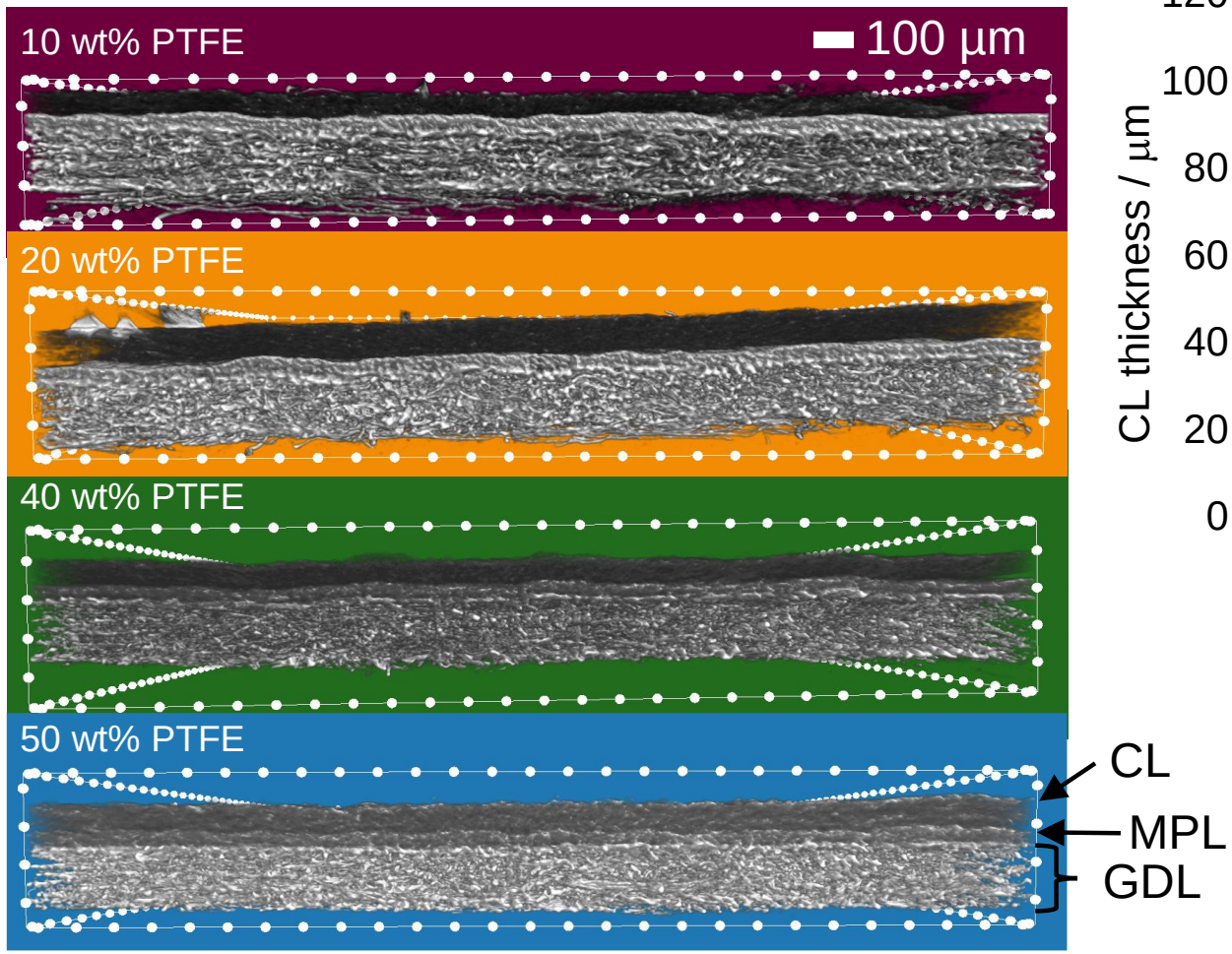
Double layer capacity



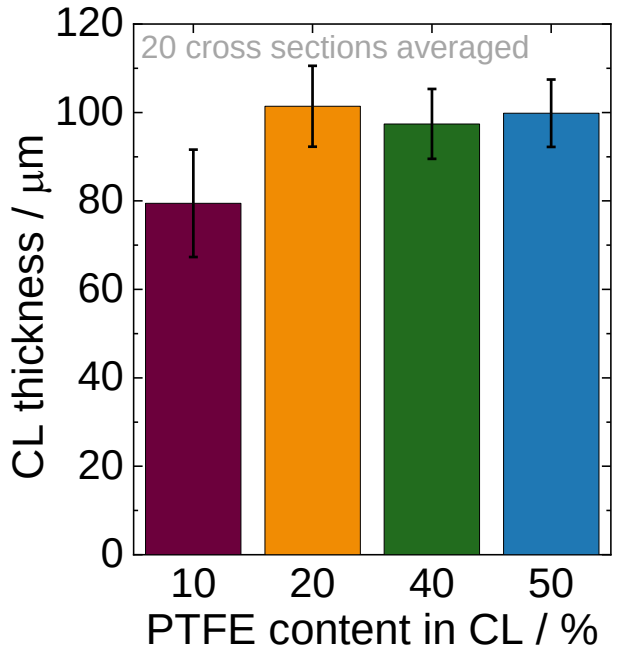


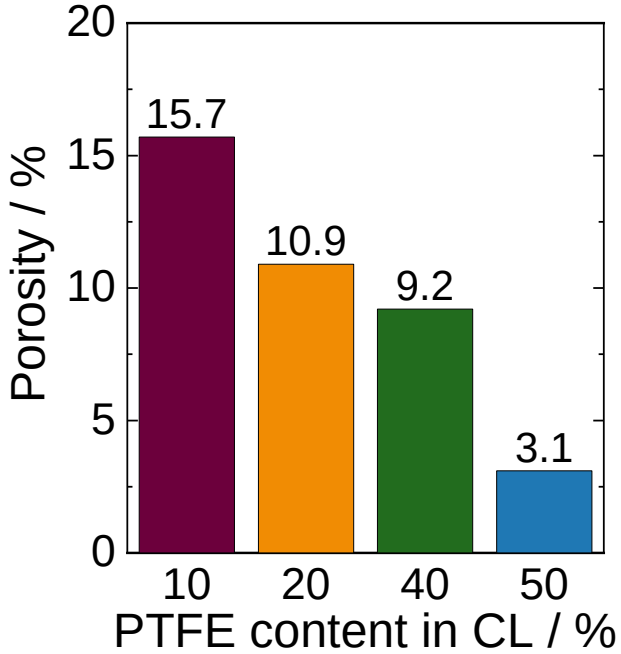
- Tilted voltammograms indicate limited electronic conductivity at begin of test
- **10, 20, 40 & 50 wt% PTFE GDE**: No significant differences in double layer capacity

Catalyst layer morphology and thickness



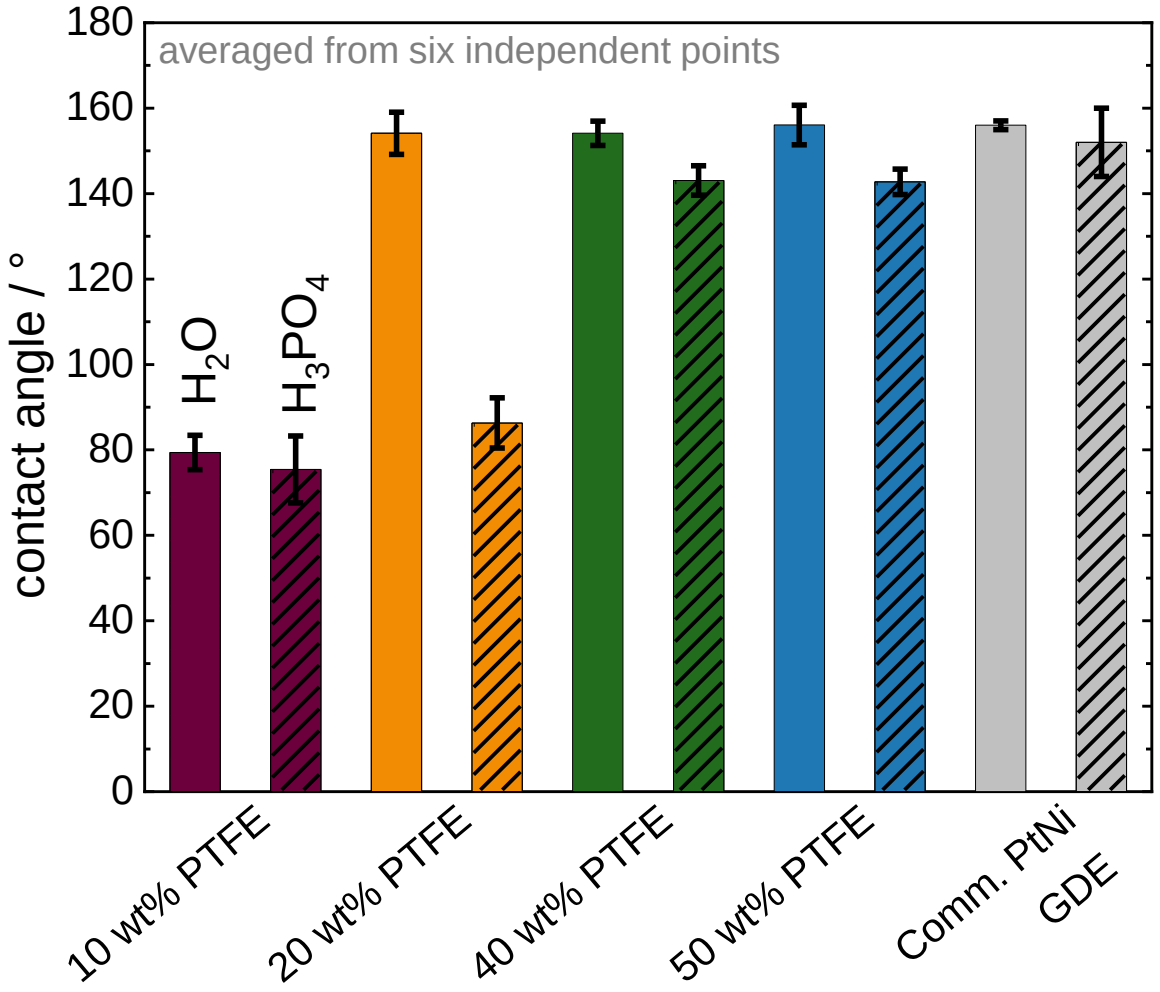
CL morphology (µ-CT images - grey values adapted for each PTFE content for optimal visibility of CL)





- 20, 40 and 50 wt% PTFE: CL thickness of ca. 100 µm
- 10 wt% PTFE GDE: Slightly thinner CL (79 ± 12 µm)
- 50 wt% PTFE: Less porous structure

Wettability of CL surface



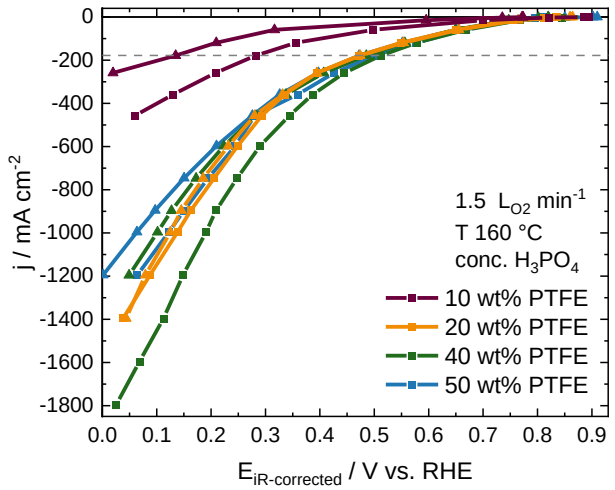
- **20, 40 & 50 wt% PTFE GDE**: Counterbalance acid drag (acidophilic oxygen and nitrogen functionalities) into catalyst layer

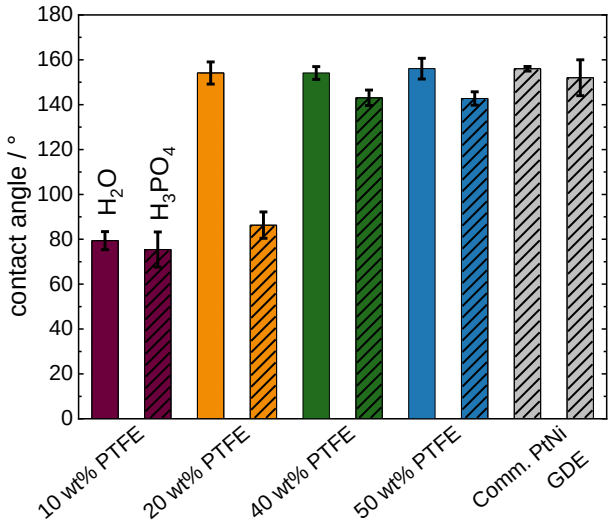
- **10 wt% PTFE GDE**: Less hydrophobic character
  - Might suffer from flooding with electrolyte
  - Not suited for Fe-N-C GDE for HT-PEMFC application

Conclusion and outlook

Conclusion

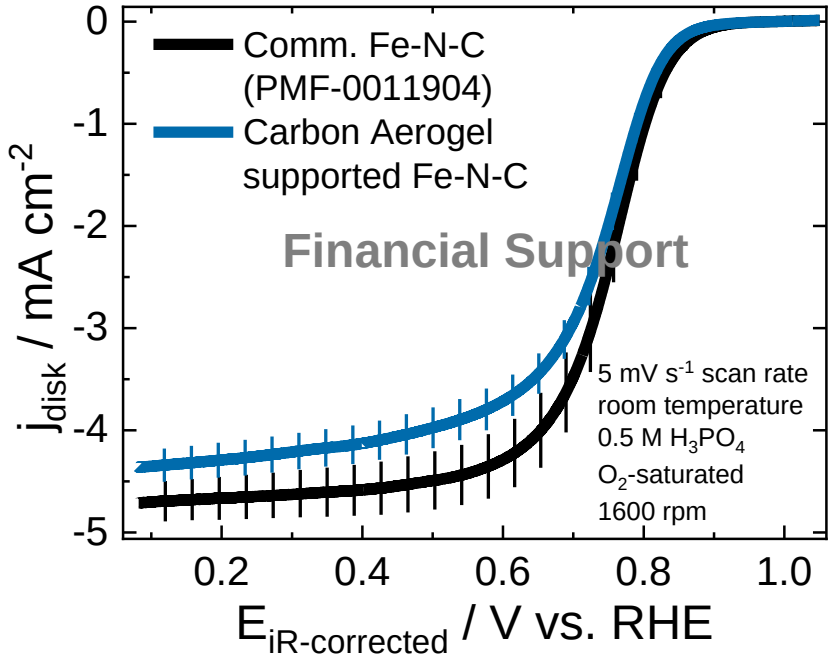
- Min. 20 wt% PTFE in Fe-N-C CL for sufficient performance
  - ➢ H_2O contact angle > 140 ° → beneficial hydrophobic properties
  - ➢ 20 wt% PTFE GDE as optimized PTFE content





Outlook

- HT-PEMFC single cell tests
- Effect of additives on Pt-free catalyst inks
- Optimized fabrication for novel carbon aerogel based Fe-N-C



Financial Support

Rotating-ring-disk electrode investigation of alternative carbon support material.

Contact: tanja.zierdt@dlr.de

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Thank you
for your attention!

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Contact: tanja.zierdt@dlr.de