A Pathway towards Pt-free Cathodes in High-Temperature Proton Exchange Membrane Fuel Cells

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Motivation

- High temperature proton exchange membrane fuel cell (HT-PEMFC)
  - Operation temperature of 160 °C enables easier heat management and higher tolerances towards SO_2 and CO enabling operation with reformate[1]
  - High Pt catalyst loading needed due to partial deactivation by phosphates from the acid doped membrane
    - ➢ Reduction or complete replacement of Pt catalyst needed for cost reduction
    - ➢ M-N-C catalyst (with M = Fe) promising as cathode catalyst

Pt-free Cathode

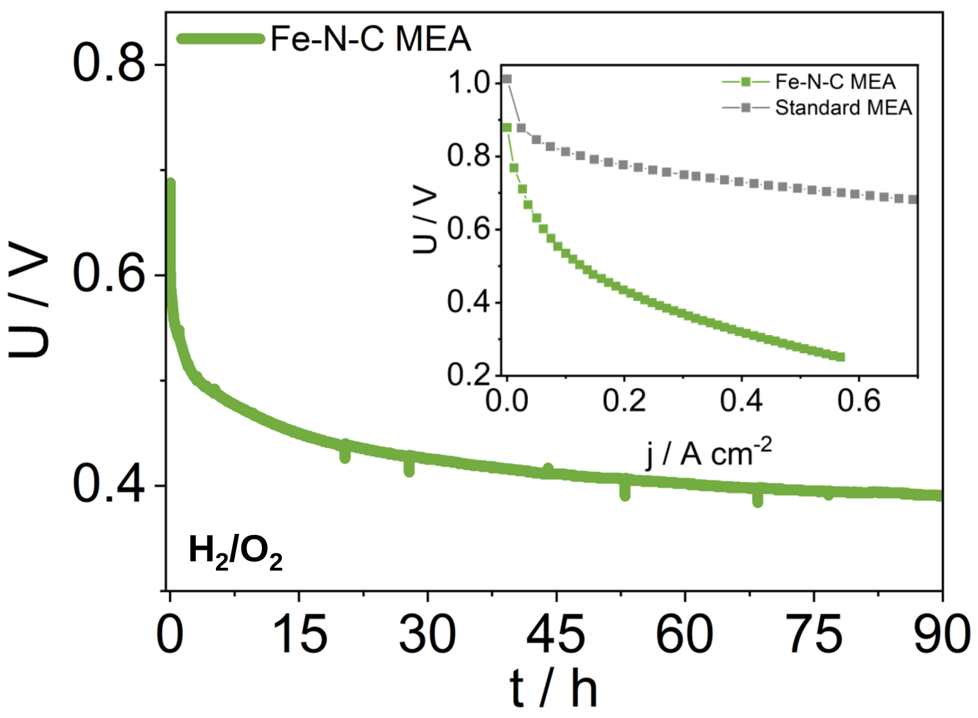


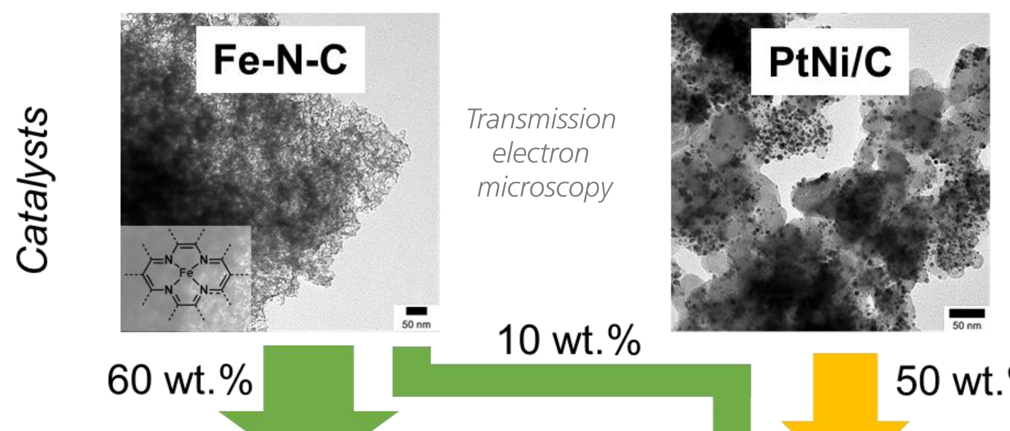
Fig. 1: Fe-N-C based MEA), cell size of 5 cm² at 0.1 A cm^{-2} and polarisation curve as inset.

HT-PEMFC performance and stability

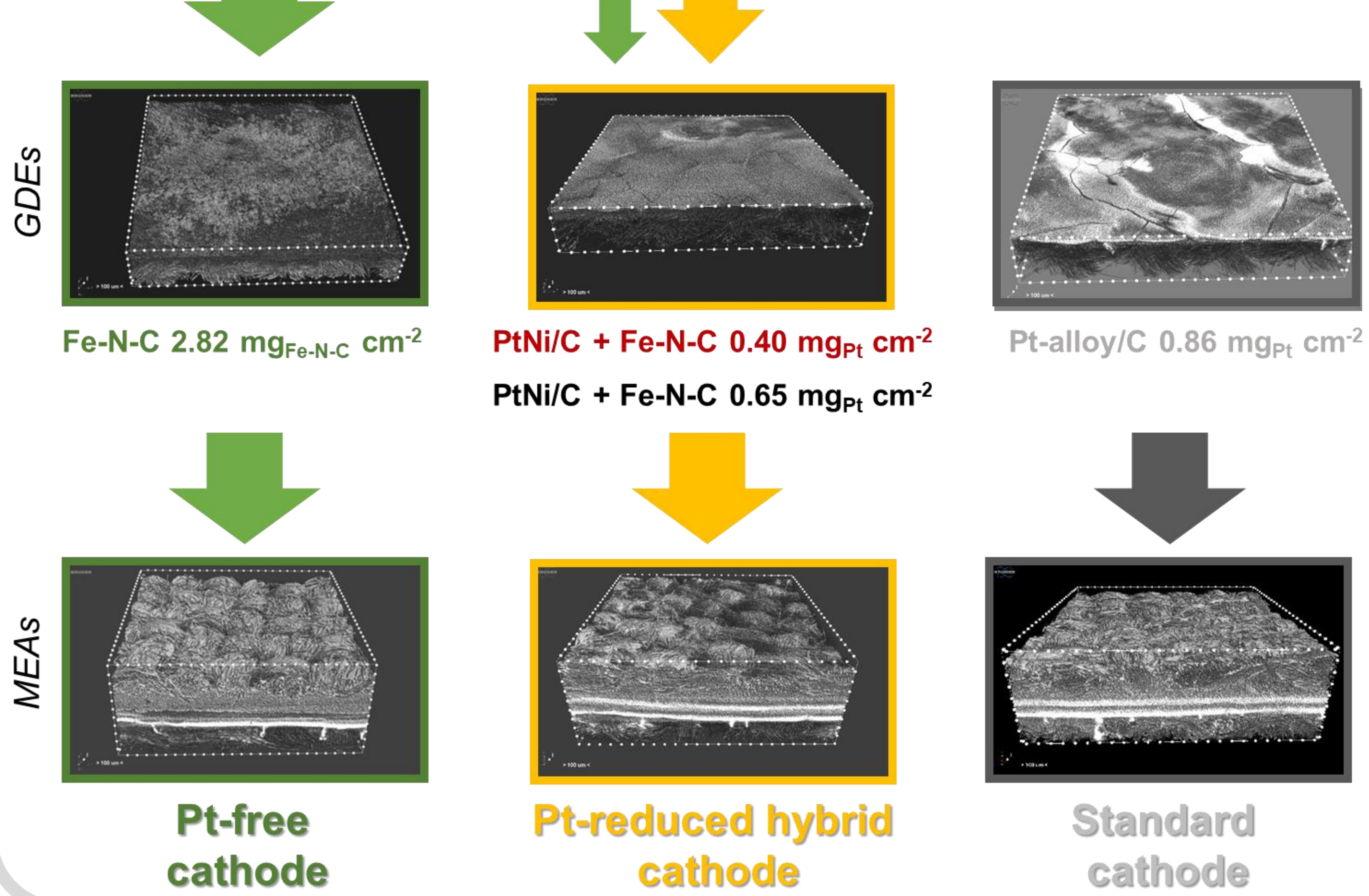
- Significantly lower performance for Fe-N-C MEA compared to standard MEA[4]
- Strong voltage decay within the first 24 h of operation
  - Pore flooding and/or deactivation of active $Fe\text{-}N_x$ sites

Experimental Approach

- Commercial Fe-N-C catalyst (PMF-011904, Pajarito Powder) and PtNi/C catalyst (24 wt.% Pt, De Nora) for GDEs; Celtec®-P1200 as standard MEA Celtec®-P1200



1. Gas diffusion electrode (GDE) fabrication using ultrasonic spray coating (40 wt.% PTFE binder)
2. Sinter step for Pt-containing GDE (350 °C,10 min, N_2)
3. Membrane electrode assembly (MEA) fabrication by hot-pressing using Celtec® membrane, Pt/C anode (0.99 mg_{Pt} cm^{-2})



Micro computed tomography images of GDEs and MEAs with cathode up in drawing. The distance between two white circles in the sample's frames displays 100 µm.[2,3]

Pt-reduced Hybrid Cathode

HT-PEMFC performance and activation (160 °C)[3]

- Increased voltage (U) for Fe-N-C + PtNi/C hybrid MEAs over time (Fig. 2)
  - ➢ Typical break-in duration (~60 h) not sufficient
- Abrupt U increase after electrochemical measurements during BoT for Pt-based MEA (Fig. 2 b)
  - ➢ Catalyst activation e.g. dealloying
- U increase during activation 2 for hybrid MEAs (Fig. 2 b)
  - ➢ Electrolyte redistribution in presence of Fe-N-C
- Comparable performances for standard and Pt-reduced MEA_0.65 (Fig. 3)
- Lower performance/voltages in air, attributed to different coating method of hybrid compared to standard MEA

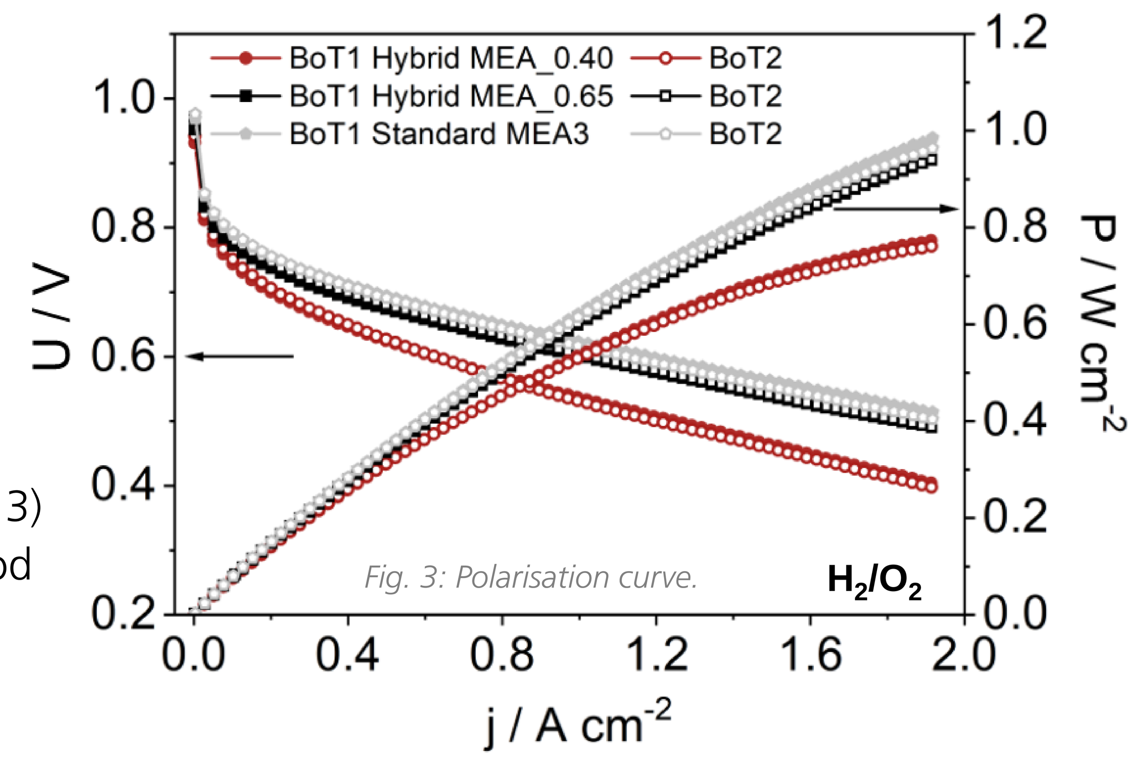


Fig. 3: Polarisation curve.

Rates and resistances[3] (Fig. 4)

- Distribution of relaxation times (DRT) analysis from electrochemical impedance spectra (EIS)
- Verification that longer activation for hybrid MEAs is caused by mass transport (MT) limitations.
- MT rate increased and MT resistance decreased during activation 2.

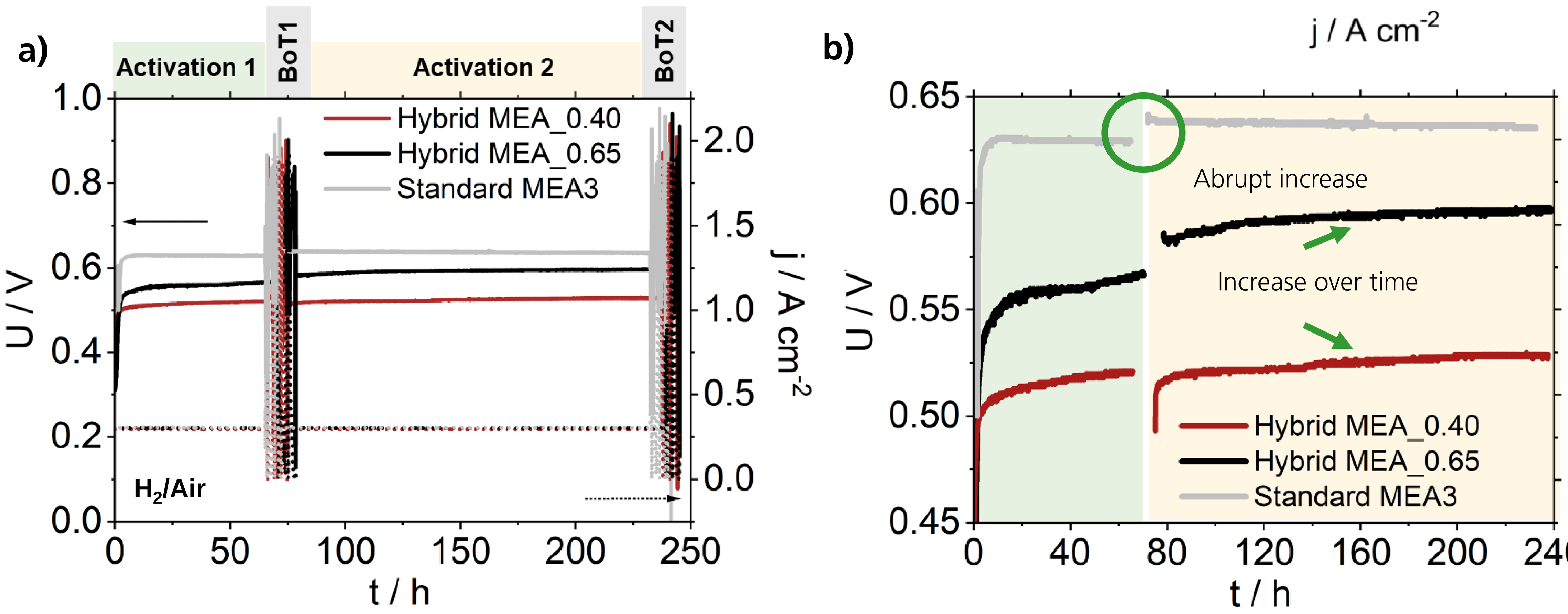


Fig. 2: a) Cell size of 25 cm² with two activation periods and begin of test (BoT) analysis with indicated Pt loadings of hybrid MEAs and b) voltage-time plot excluding BoT1 and BoT2.

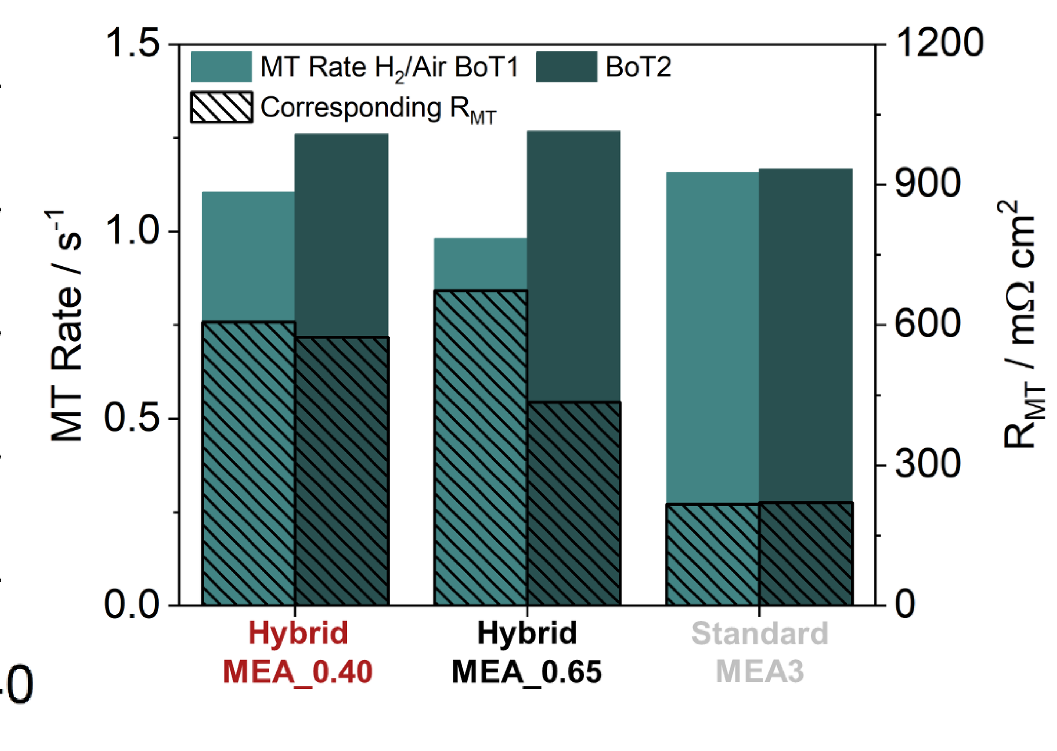


Fig. 4: Mass transport (MT) rate and MT resistance R_{MT} determined from DRT analysis from EIS at 0.3 A cm^{-2} under H_2/air (1.5/2.0) operation.

Conclusion & Outlook

- Cathodes with Fe-N-C only need optimization of performance and stability
  - Implementation of carbon aerogels in Fe-N-C for increased stability, activity and up-scaling possibility
- Hybrid electrodes show promising results for Pt-loading reduction in HT-PEMFC MEAs
  - Optimization of Pt-free and hybrid electrode fabrication e.g. ink and coating process
- Adjustment of activation procedures for hybrid electrodes needed

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