Impedance characterization of supported oxide ion conducting electrolytes

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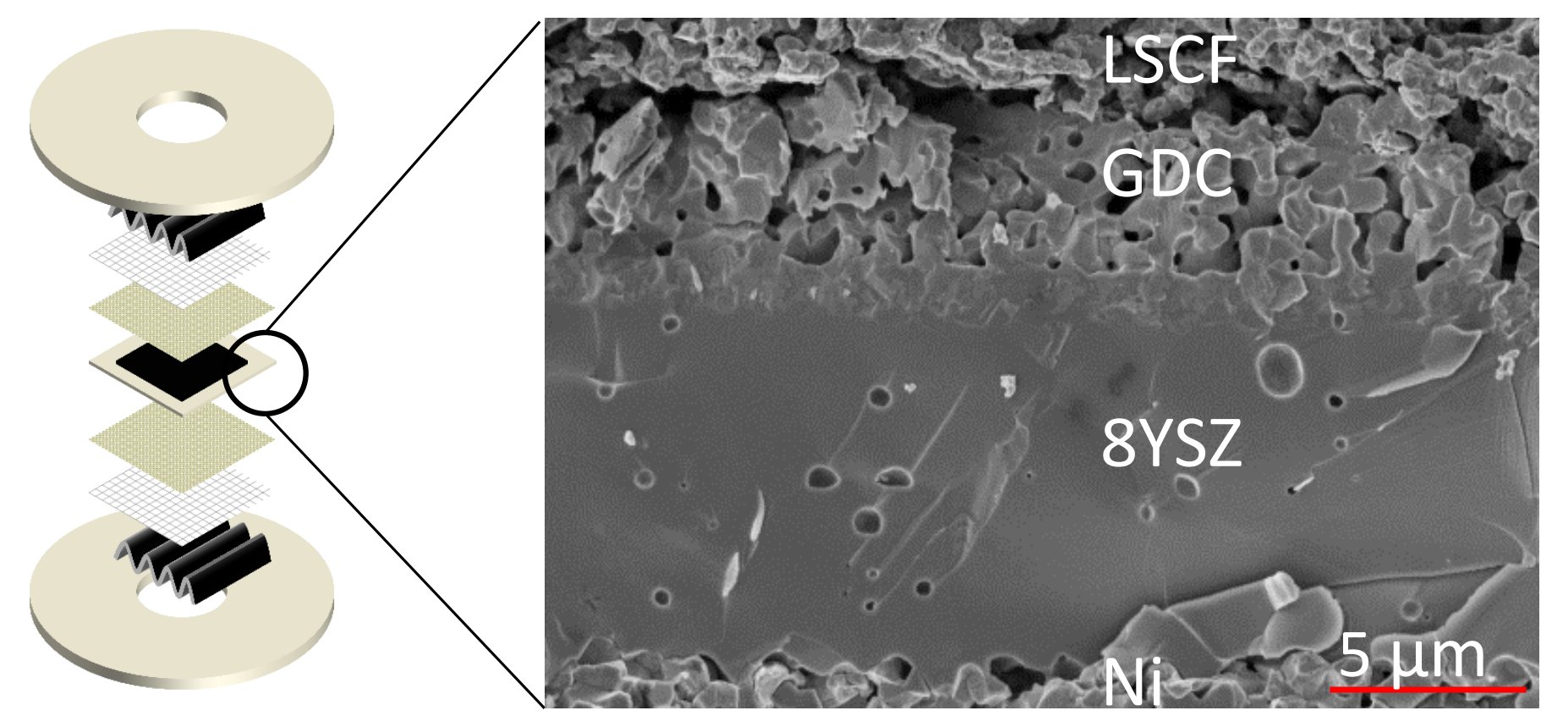
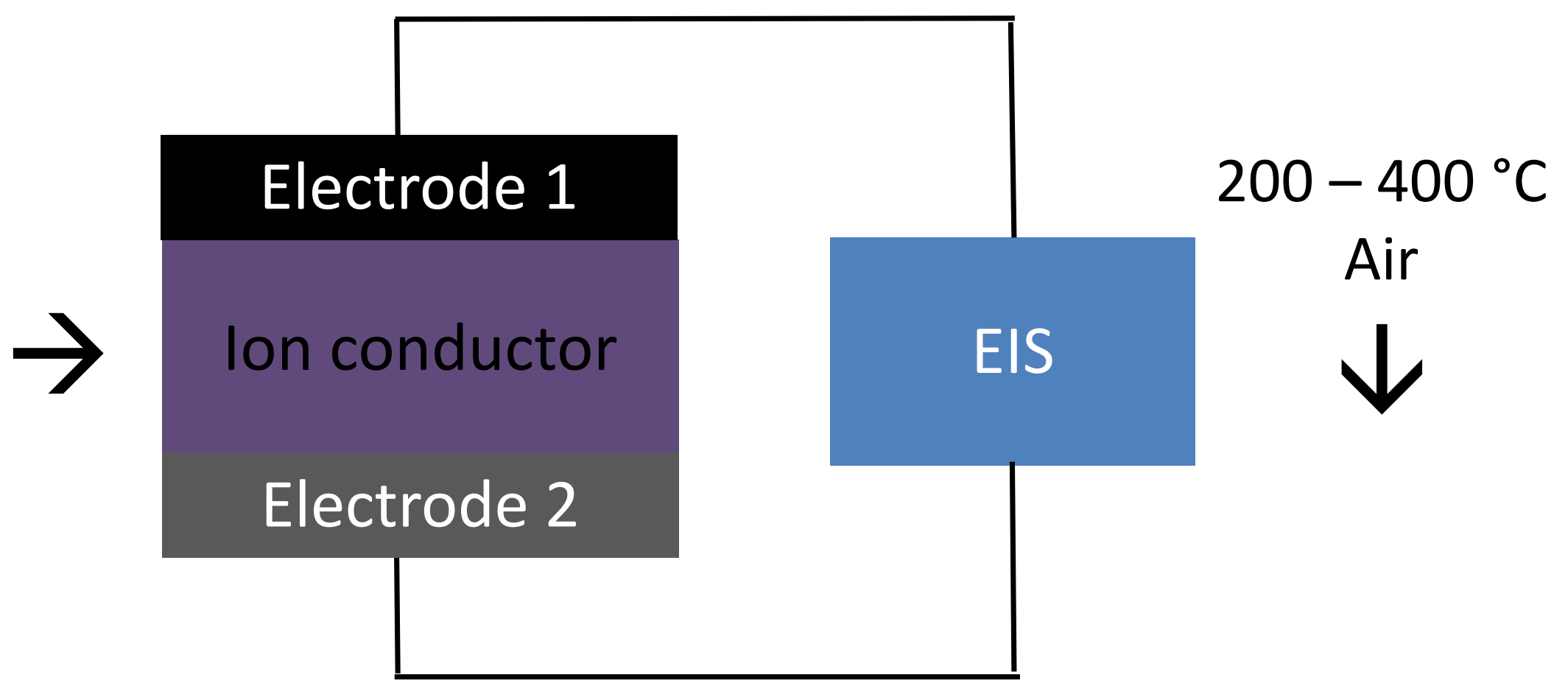
Background

Electrochemical impedance spectroscopy is a widely used tool to study electrochemical reactions in batteries, fuel cells and other electrochemical energy conversion devices. However, conduction processes in the electrolyte of high temperature fuel or electrolysis cells (SOFC / SOEC) are inaccessible during operation, severely restricting the information that can be obtained about performance and degradation of the electrolyte. Using the distribution function of relaxation times (DRT), we study the ionic conduction properties and degradation phenomena in multi-layered solid electrolytes, ex situ and at low temperatures.

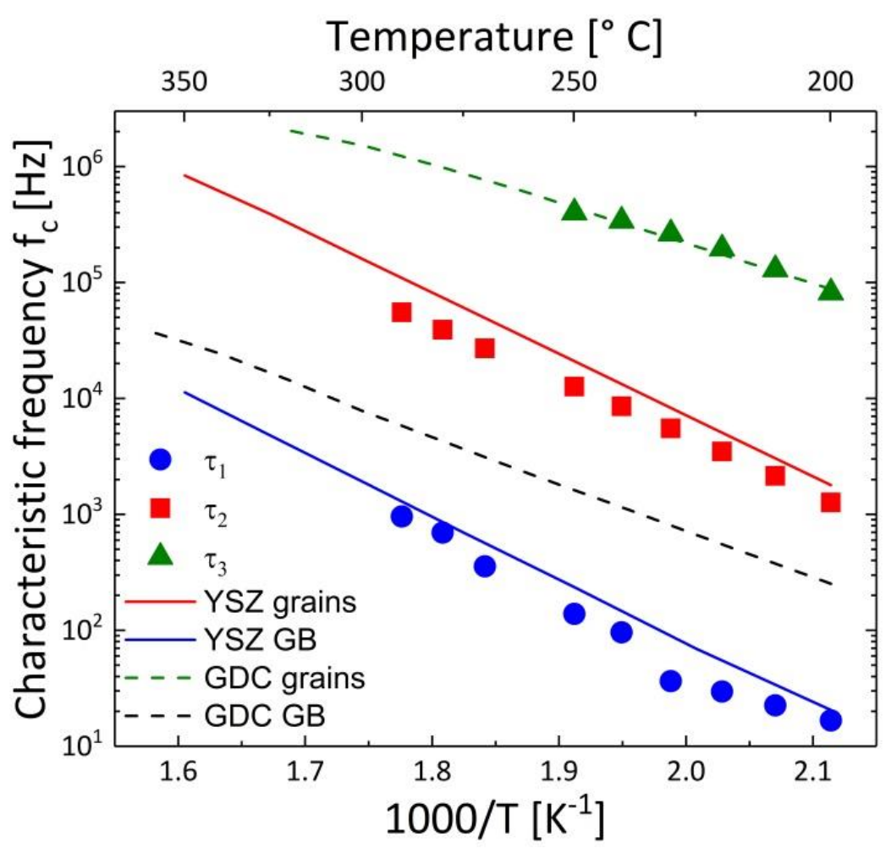
Methodology

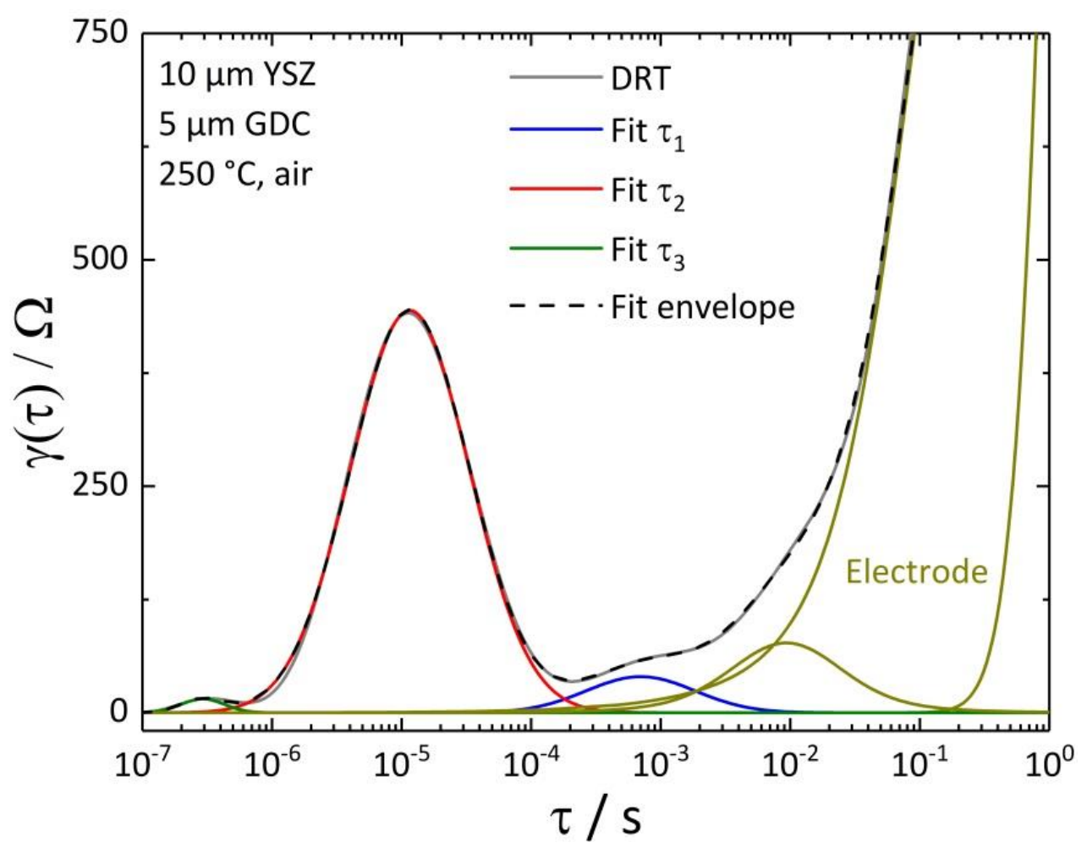
➢ Can we measure the actual ionic conductivity of a supported electrolyte?

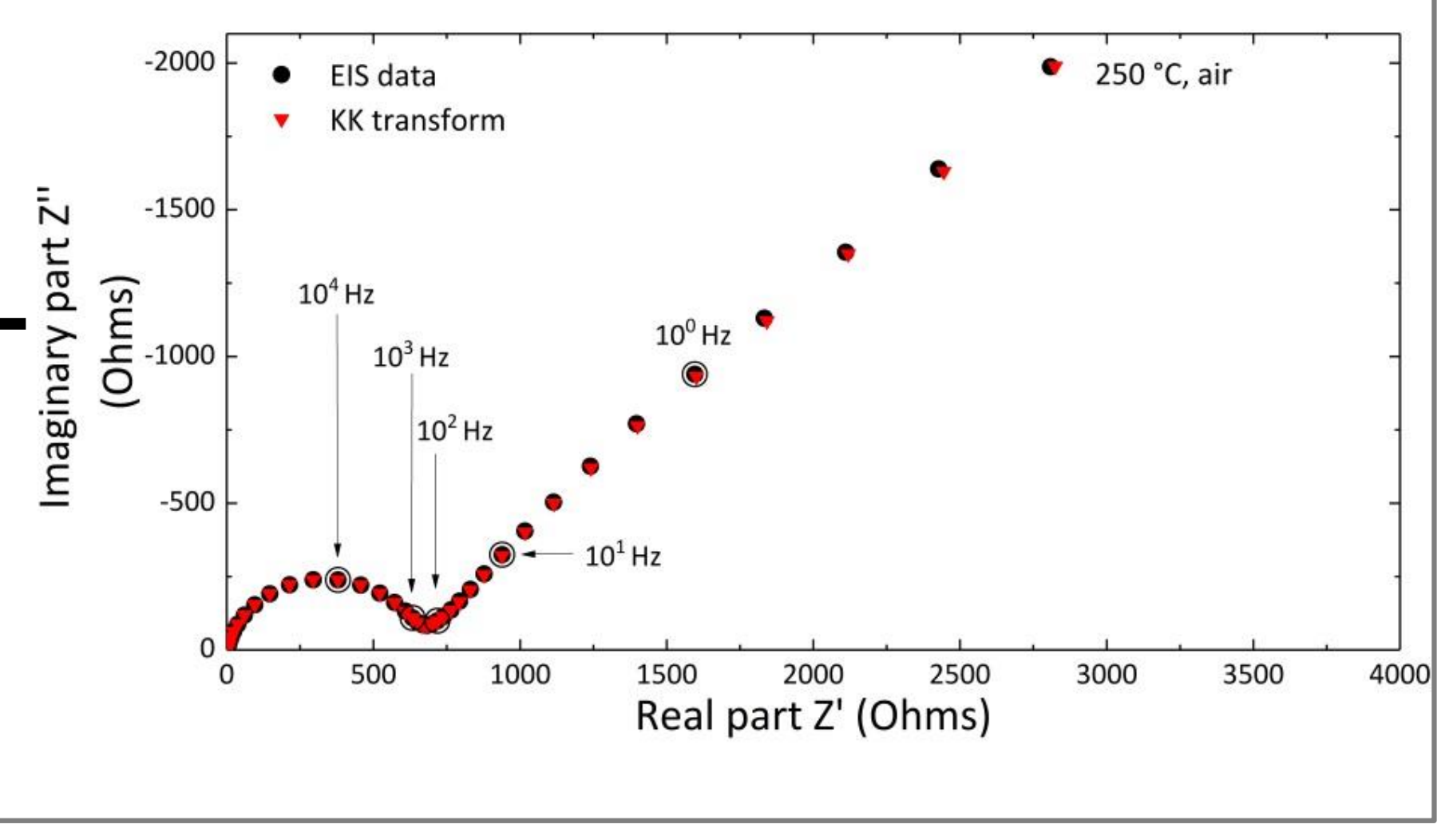




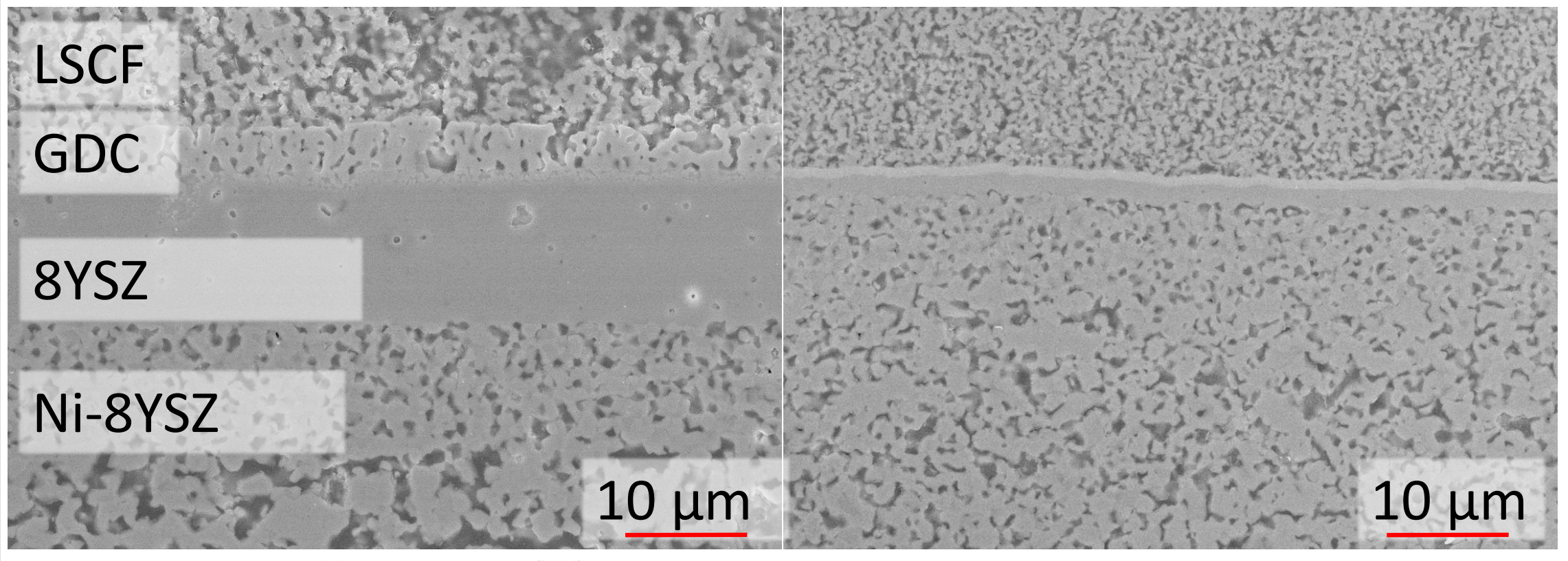
$\frac{1}{2\pi f_c} = \tau_{RC} = RC$ DRT fitting Impedance data

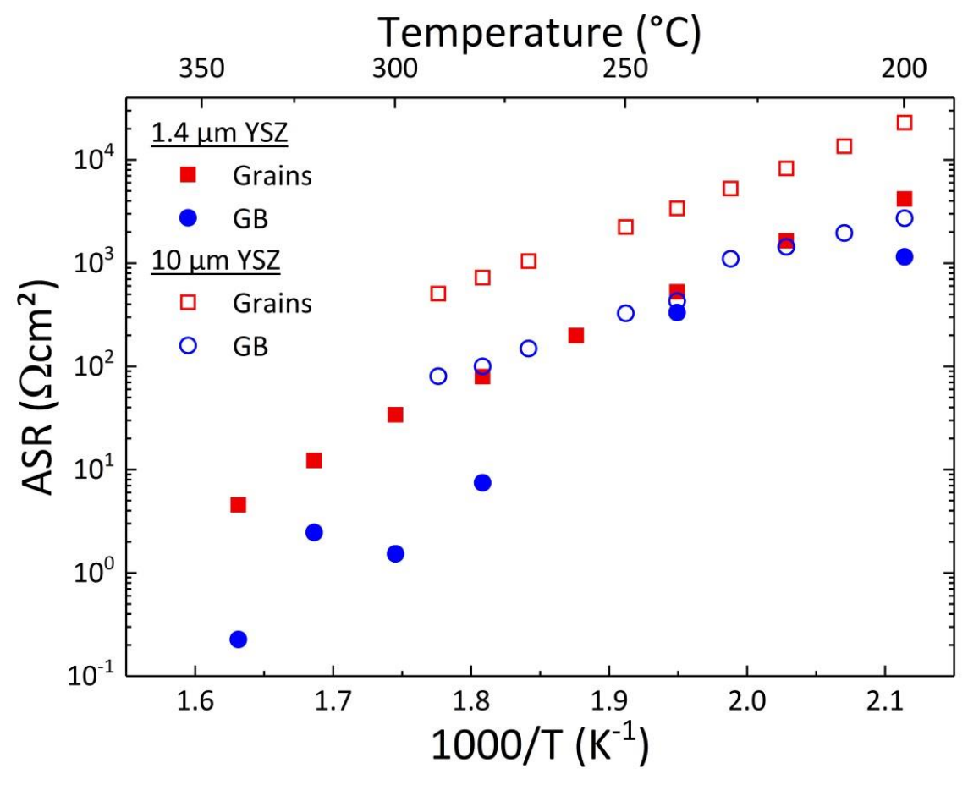






Validation

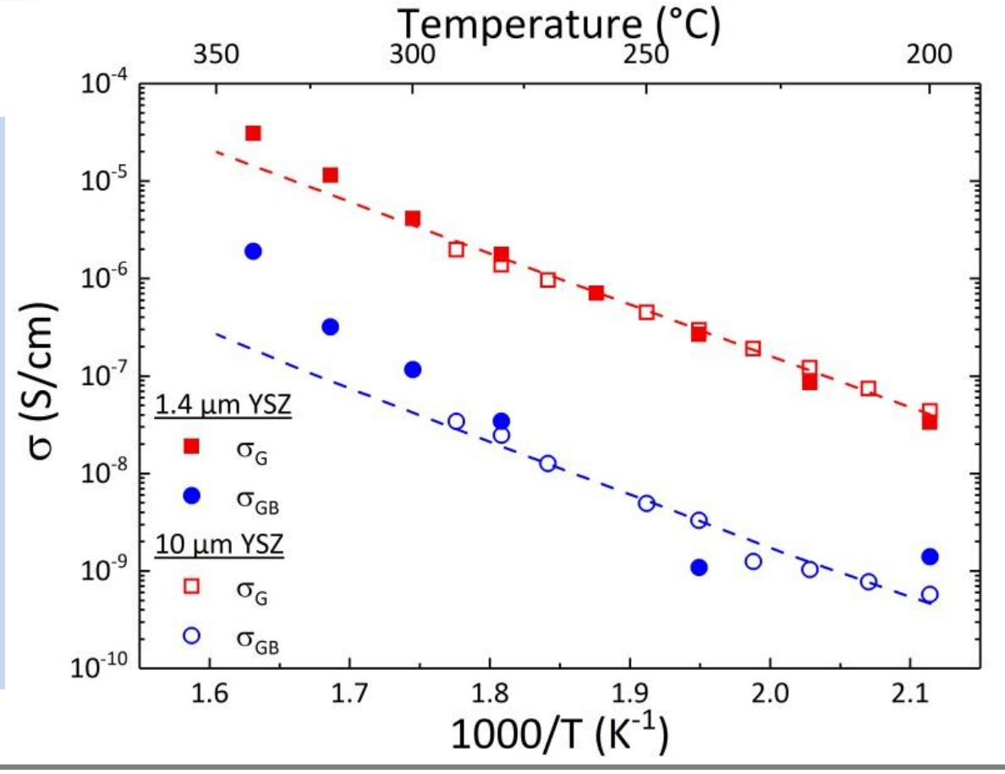




- Supported YSZ with different thickness
- Full cells measured in air show different ASR values

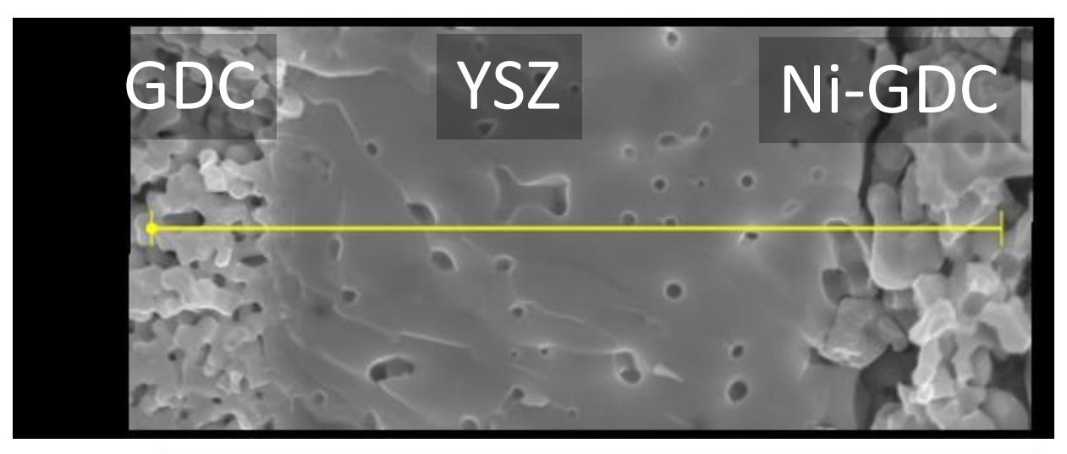
- Electrolyte conductivity is identical to bulk YSZ

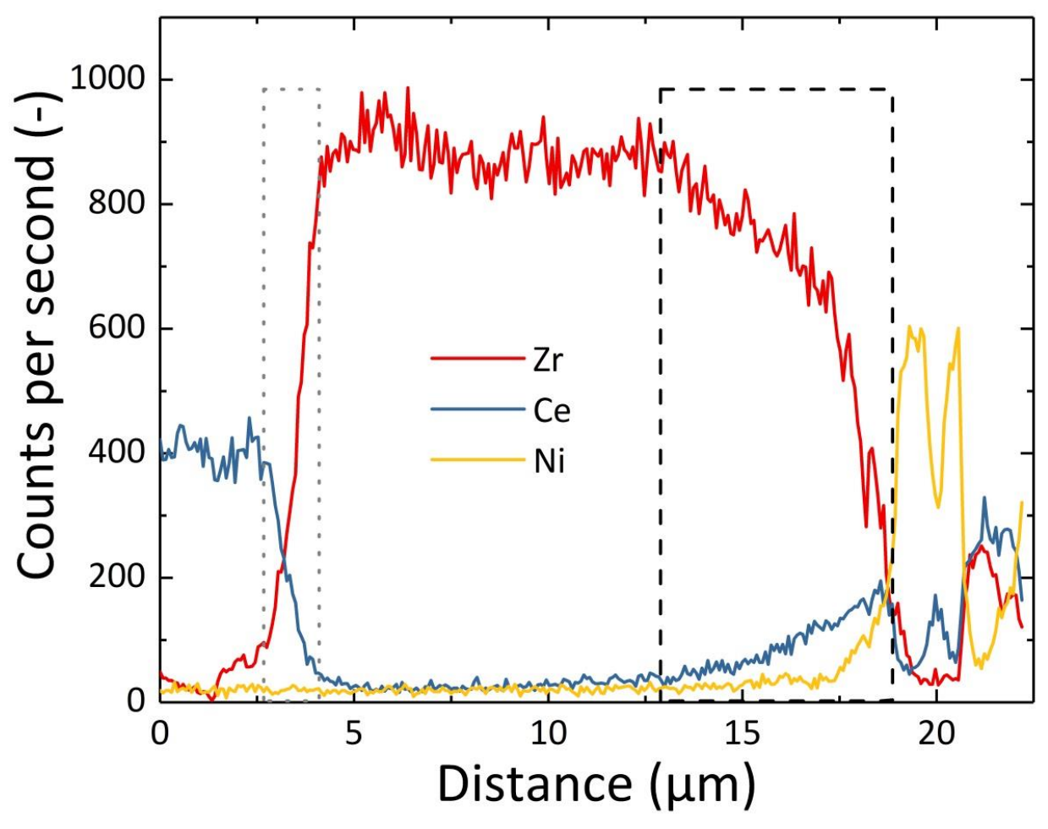
➢ DRT fitting is suitable to quantify differences in cell design

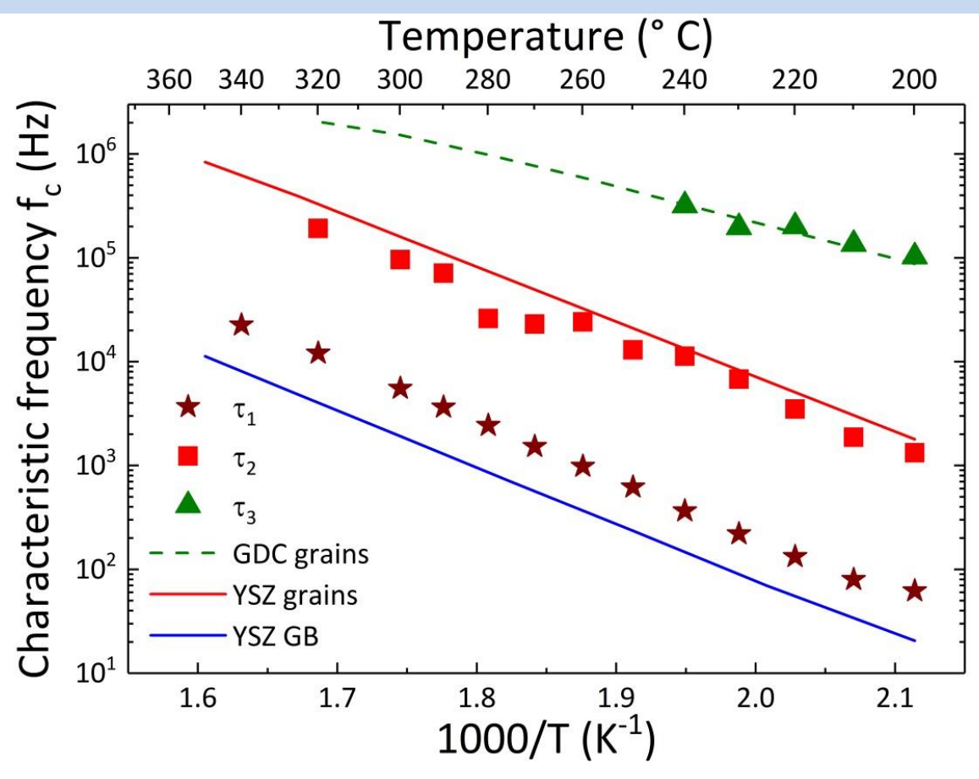


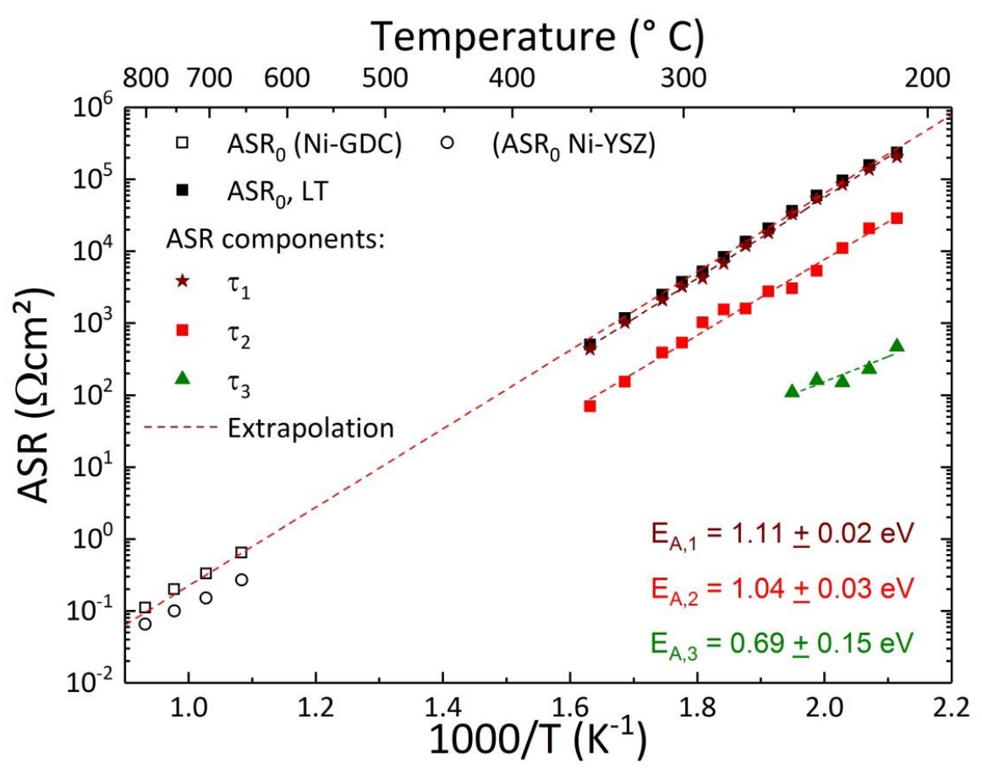
Interdiffusion

- Co-sintering of NiO-GDC anode and YSZ electrolyte leads to lower cell performance compared to NiO-YSZ anode[1]
- SEM-EDX shows interdiffusion between GDC and YSZ after co-sintering



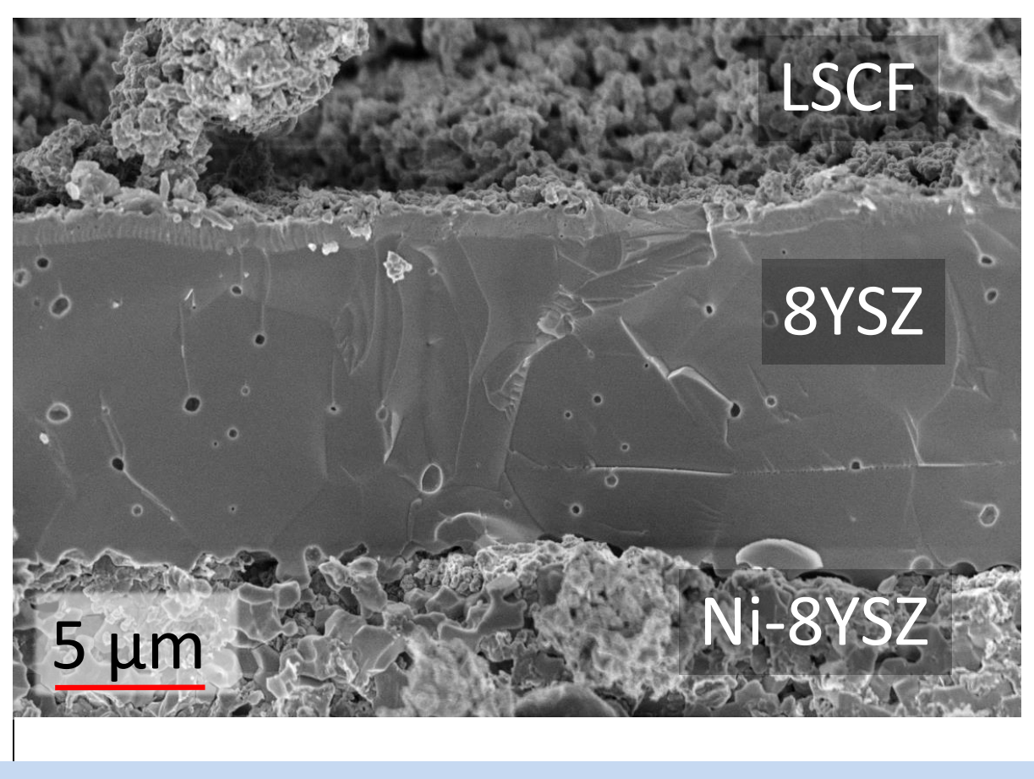






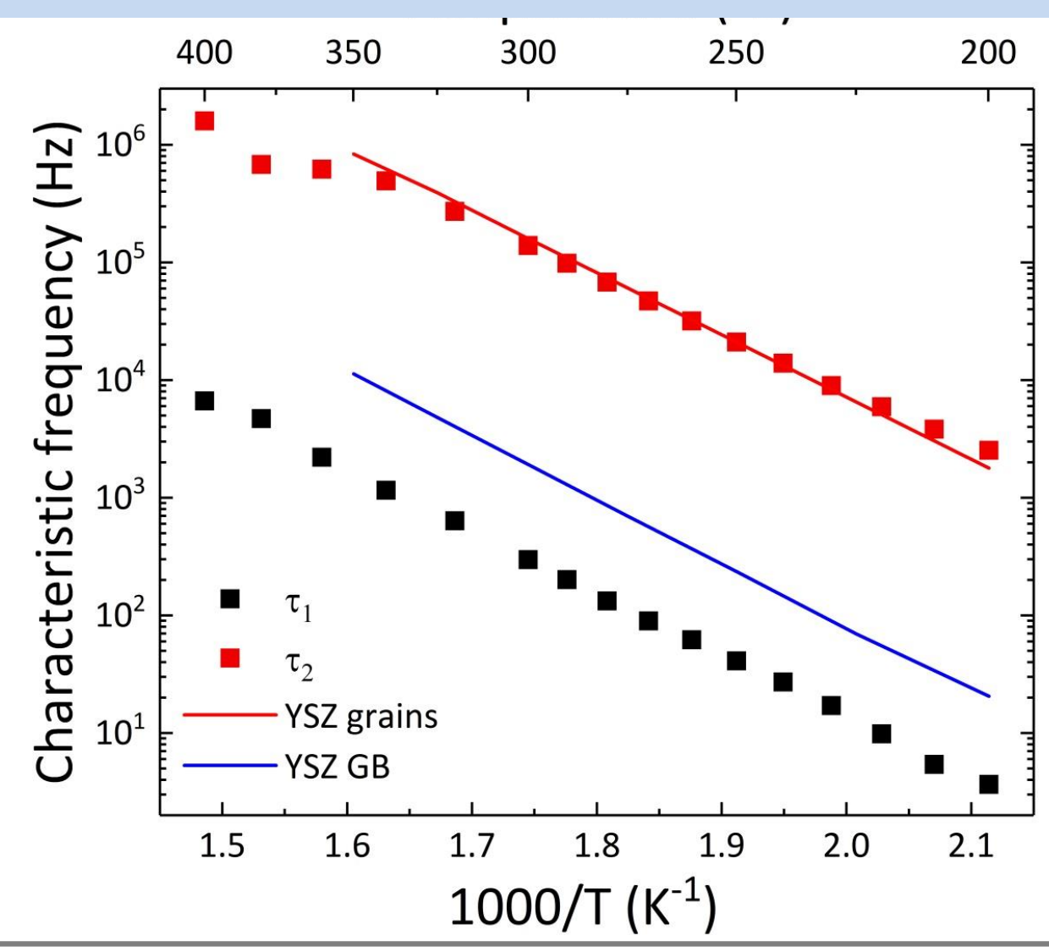
- Contributions from YSZ and GDC can be identified by DRT
- Additional contribution due to interdiffusion of YSZ and GDC at the anode / electrolyte interface (ε ~ 10^{-12} F/cm)
- Ohmic ASR mainly due to interdiffusion phase (τ_1)
- Resistive phase forms between anode and electrolyte

SOFC – Mn diffusion



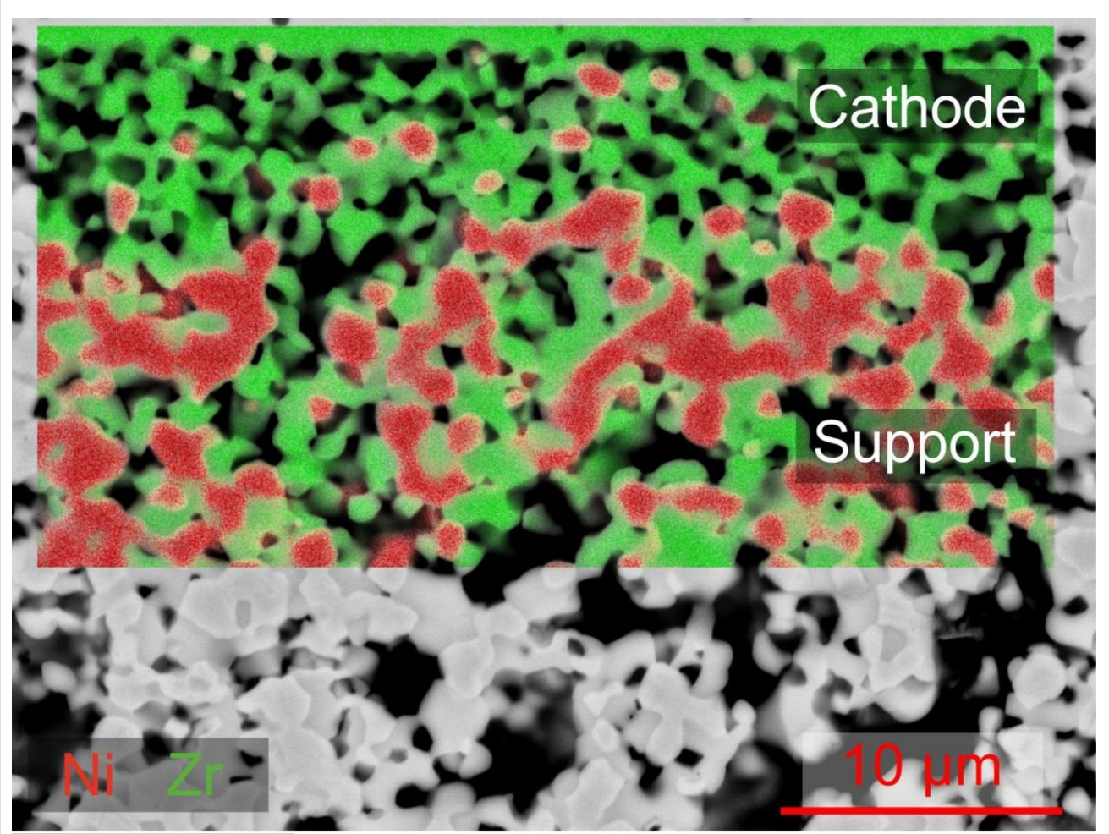
- SOFC long-term operation (~35 kh) shows Mn diffusion into the electrolyte [2]
- YSZ grain boundary contribution shows lower f_c and lower conductivity (ε ~ 10^{-9} F/cm)

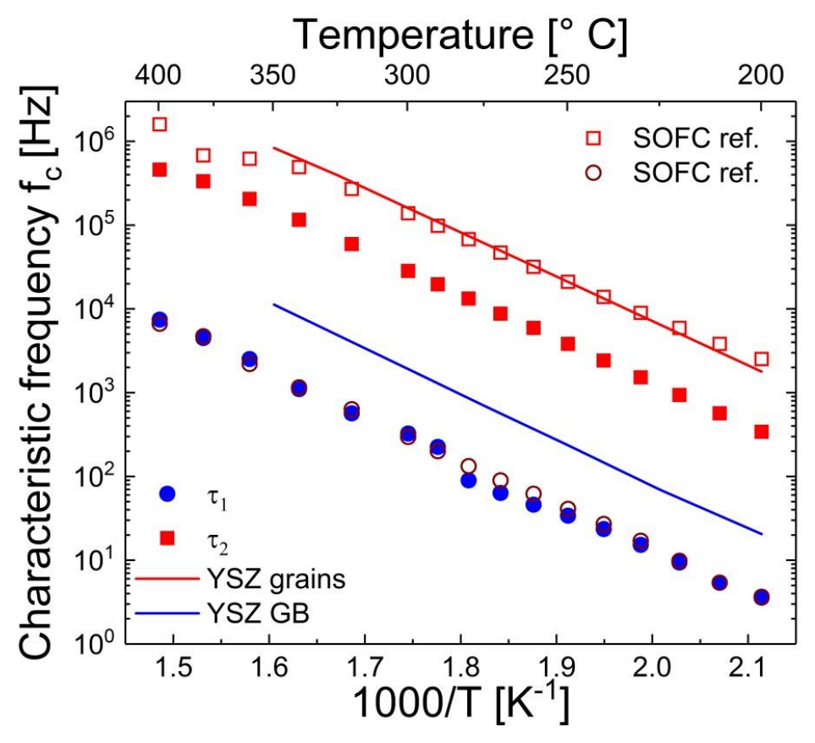
➢ Mn diffusion into the electrolyte can be traced via EIS!



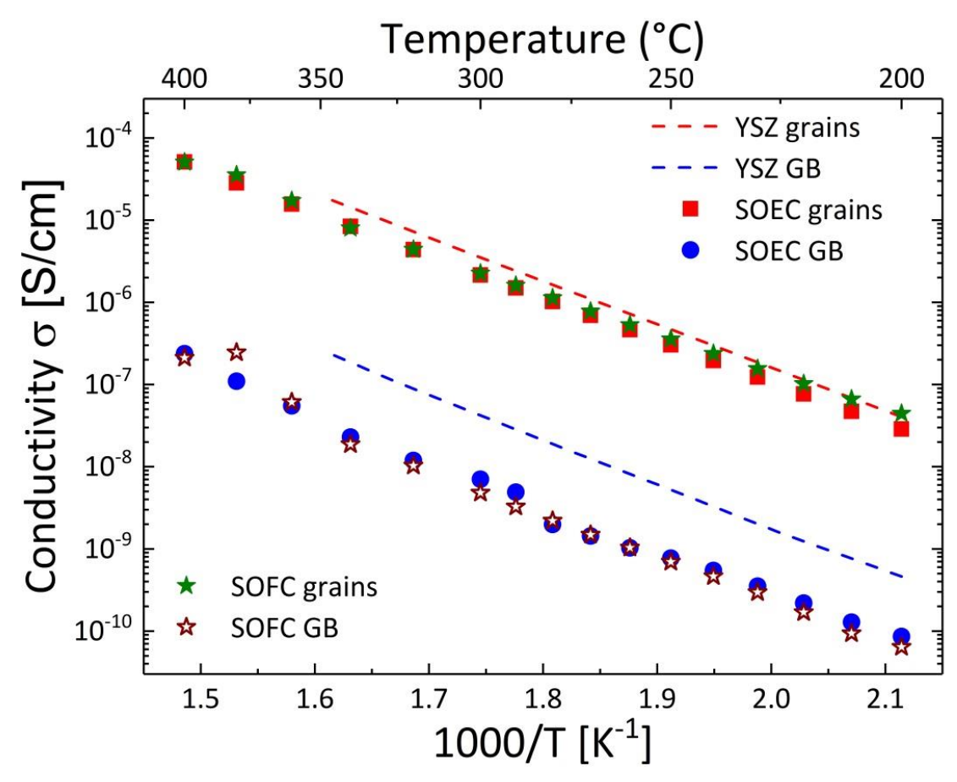
SOEC – Ni migration

- After ~ 20 kh of SOEC operation, the ohmic resistance increased significantly [3]
- Post-test analysis revealed Ni depletion at the fuel electrode / electrolyte interface





- Porosity in fuel electrode P ~ 55%
- $ASR_{ohm} = \sigma_{YSZ}^{-1}(l_{Electrolyte} + l_{Anode} * (1-P)^{2.74})$ [4]
- Corrected values agree well with bulk YSZ -> porosity in the fuel electrode causes increased ohmic resistance



Conclusions

The investigation of full cells in air enables a detailed analysis of the conductivity of supported electrolytes as thin as 1 µm, as well as the relative contributions of multilayered electrolytes. Furthermore, three case studies are presented concerning the degradation mechanism in SOFC and SOEC operation, showcasing the ability of this technique to distinguish the effects of grain boundary contamination, formation of solid solutions and the formation of porosity on the ionic conductivity of thin, supported electrolytes. [5]

References

[1] C. Lenser et al., *Journal of the American Ceramic Society* **101** (2017) (2) 739.
[2] N.H. Menzler, et al., *Journal of Power Sources* **374** (2018) 69.
[3] Q. Fang, et al., *Journal of The Electrochemical Society* **165** (2018) (2) F38.
[4] M.C. Steil, et al., *Journal of The Electrochemical Society* **144** (1997) (1) 390.
[5] C. Lenser, N.H. Menzler, *Solid State Ionics* **334** (2019) 70.