High temperature electrolysis for sustainable energy carriers

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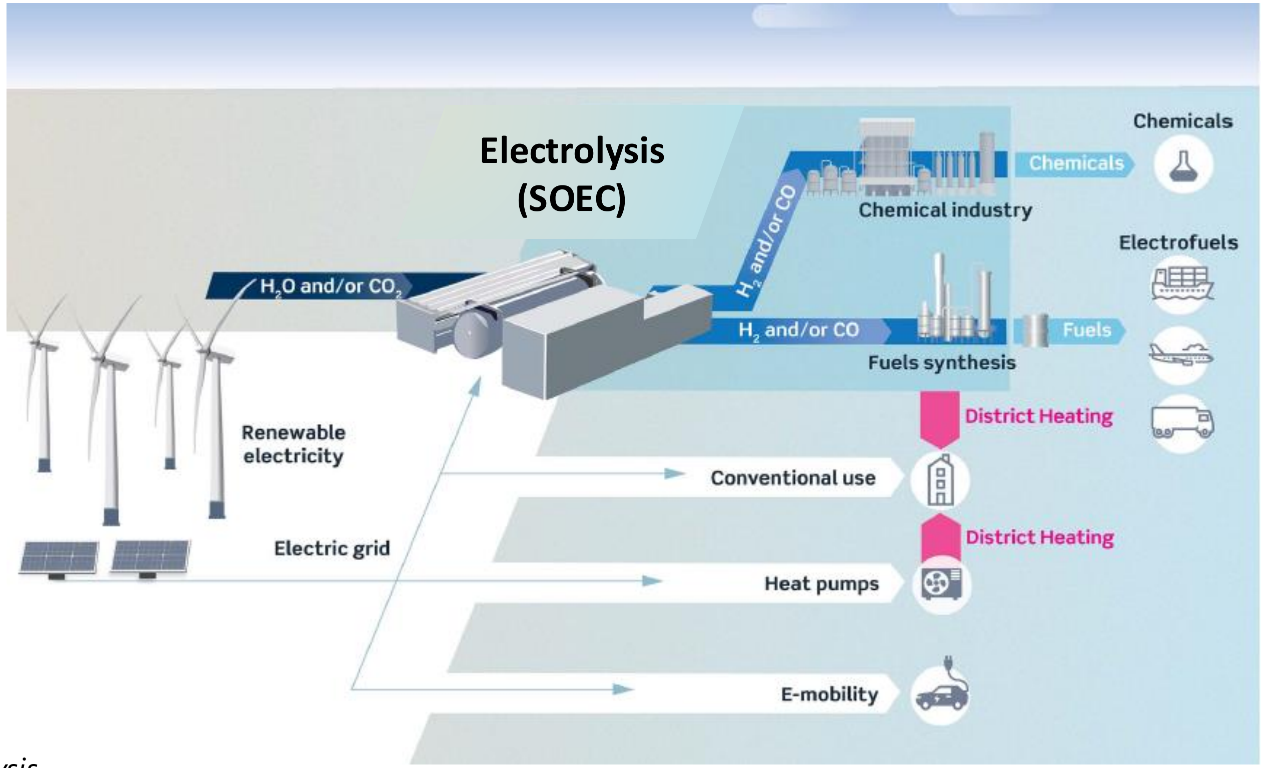
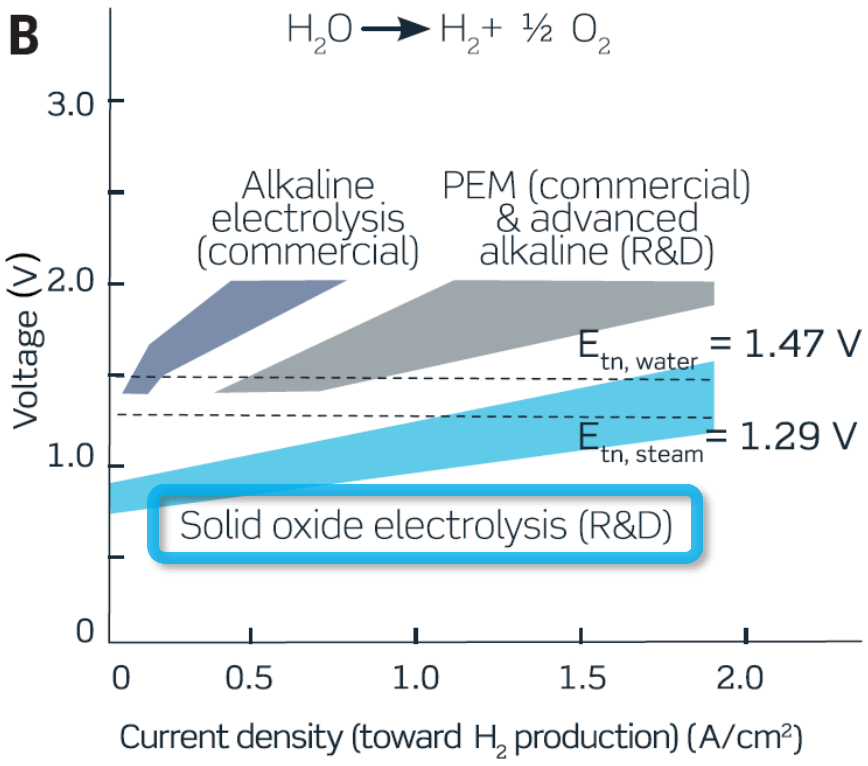
Solid oxide electrolysis

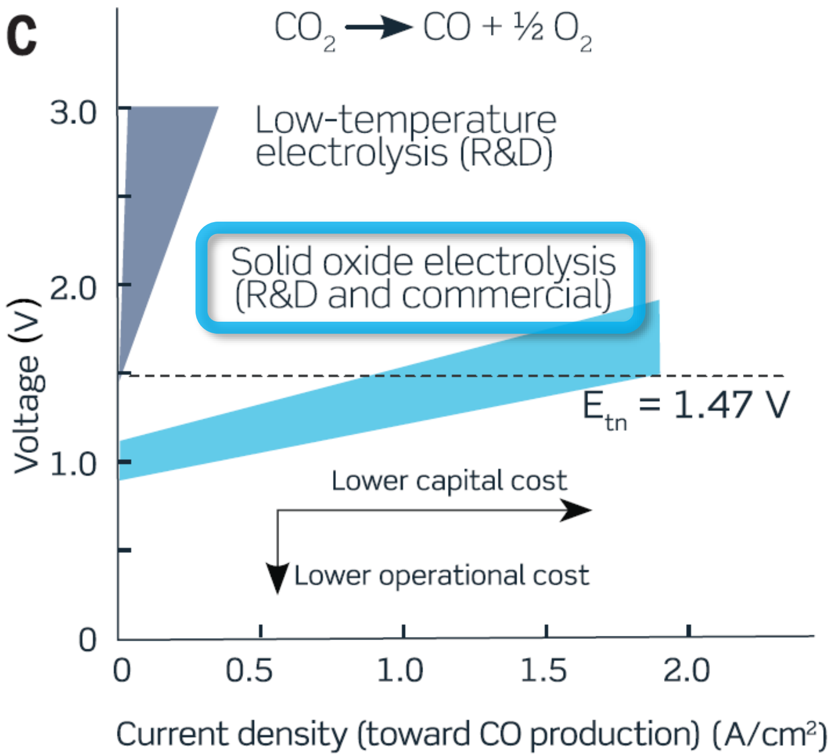
- Central role in future energy systems – link between grids (electric, gas, thermal)
- Intermittent renewable energy
  → convert electrical energy into chemical energy
- Decoupling production of transport fuels and chemicals from fossil resources
- Challenges: performance, degradation, scale-up



Hauch et al., *Recent advances in solid oxide cell technology for electrolysis*, Science 370 (2020) 6513, eaba6118, https://doi.org/10.1126/science.aba6118

Performance of electrolysis technologies





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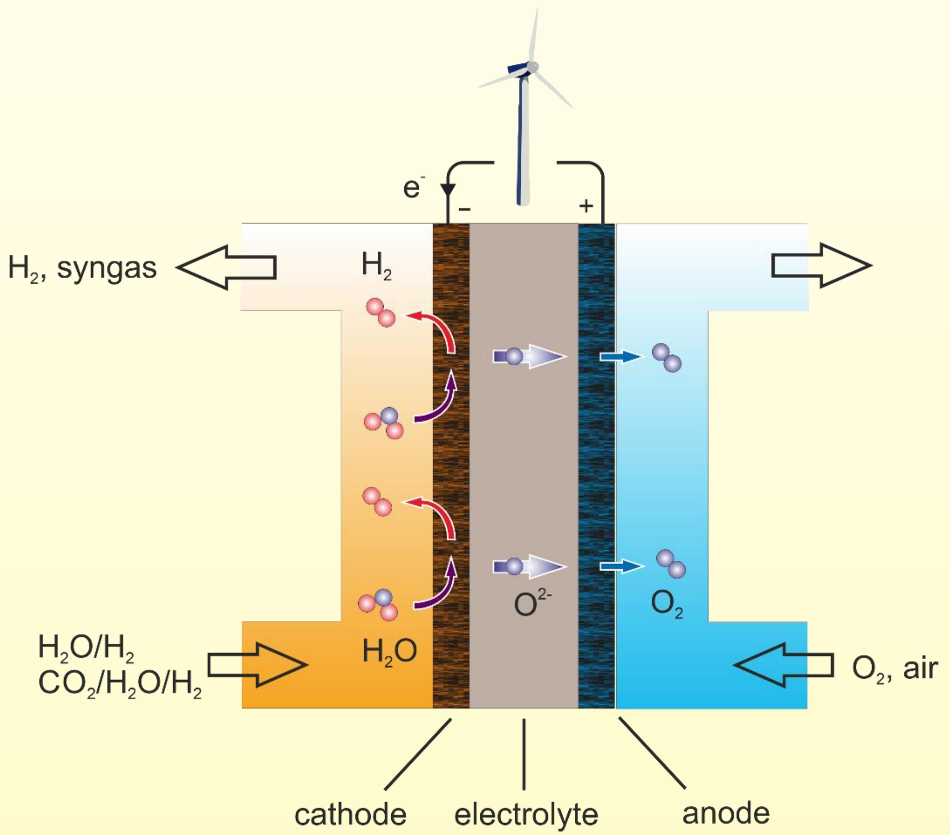
Solid oxide electrolysis

- High operating temperatures ~800°C, favourable thermodynamics and kinetics, $CO_2 + H_2O \rightarrow CO + H_2 + O_2$ (endothermal)
- Highest conversion efficiencies η_{el} of 80 % (SOEC) vs. 60 % (low-temp. electrolysis)
- Can be integrated with exothermal syntheses, e.g. of e-fuels, methanol, methane, ammonia (PtL, PtG)
  - $CO + 3\ H_2 \rightarrow CH_4 + H_2O$
  - $CO + 2\ H_2 \rightarrow CH_3OH$
  - $N_2 + 3\ H_2 \rightarrow 2\ NH_3$



Solid oxide electrolysis cell (SOEC)

Montanuniversität Leoben – Key expertise

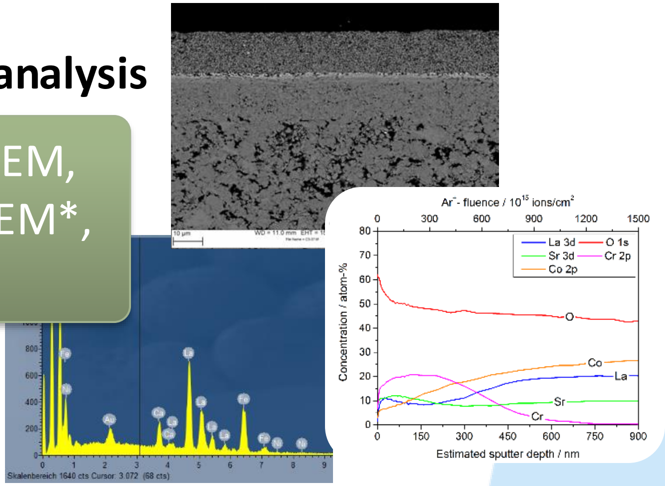
Synthesis

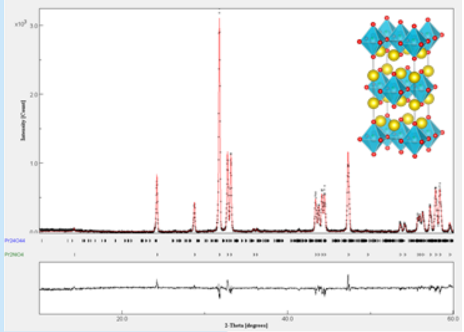
- Sol-gel processes, milling, calcination, sintering, cutting, polishing

Post-test analysis

- XRD, SEM, XPS, TEM*, AFM*

* In coope-ration with partners

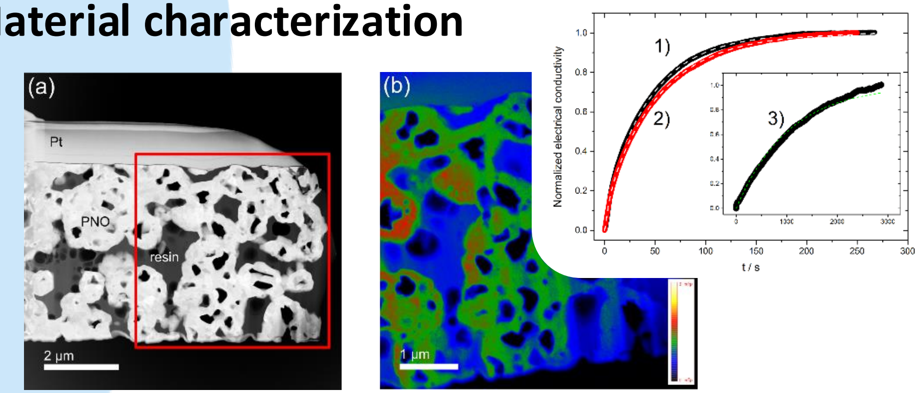




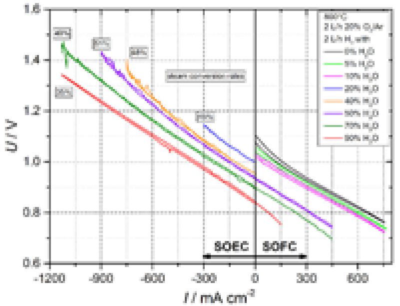
- XRD, TG, DSC, DIL, PSD, SEM, TEM*
- dc-conductivity (relaxation)

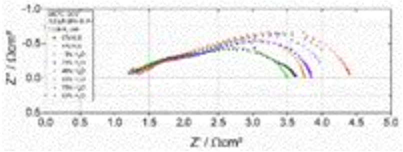
* In cooperation with partners

Material characterization



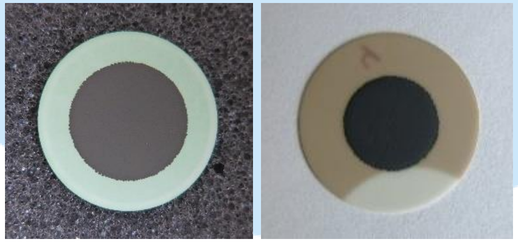
Electrochemical tests





- EIS, i-U, DRT

Cell preparation



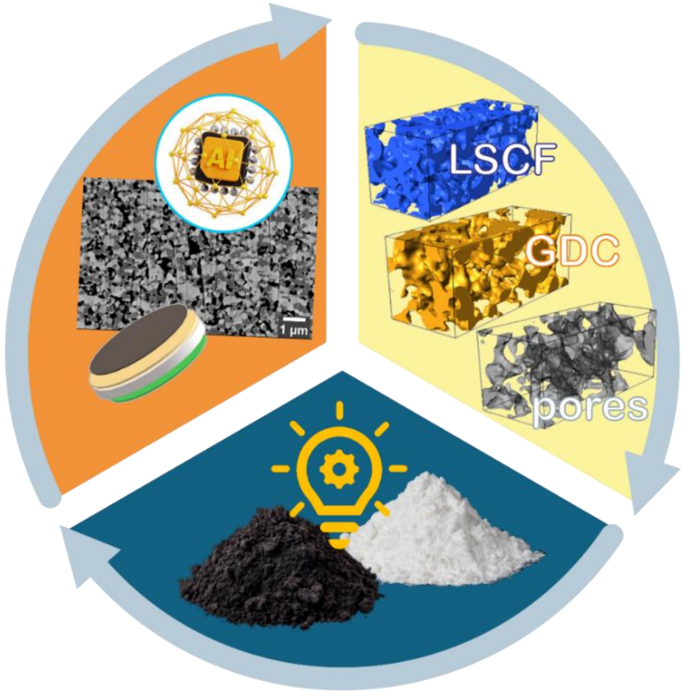
- Screen-printing, tape-casting*, additive manufacturing*, sintering

* In cooperation with partners

Material and cell design

Fundamental and technical aims

- Study complex relations between morphology, chemical composition and cell performance
- Improve functional and structural properties of electrodes and solid electrolytes
- Design materials with excellent performance and improved long-term stability
- Improve sustainability and decrease critical raw materials
- Develop guidelines for tailored structures and compositions



Partners

- e.g.

AVL LITHOZ MCL ISFK

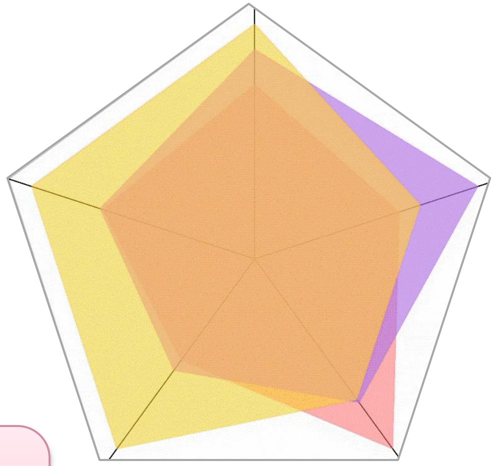
1

Relations between 3D-morphology of porous air electrodes and their electrochemical performance

Cooperation with Materials Center Leoben Forschung GmbH & AVL List GmbH

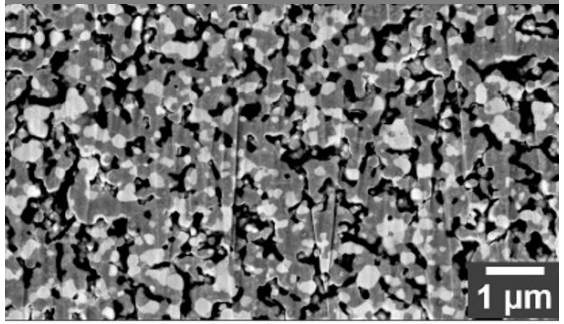
Solid oxide cell air electrodes

- Mixed conducting perovskites, e.g. $(La,Sr)(Co,Fe)O_{3-\delta}$, also applied as composites with $(Gd,Ce)O_{2-\delta}$
  - High catalytic activity for oxygen exchange
  - High electronic and ionic conductivity
  - High stability, tolerance to impurities
  - Good adherence on electrolyte
  - Optimum solid phase morphology, high porosity

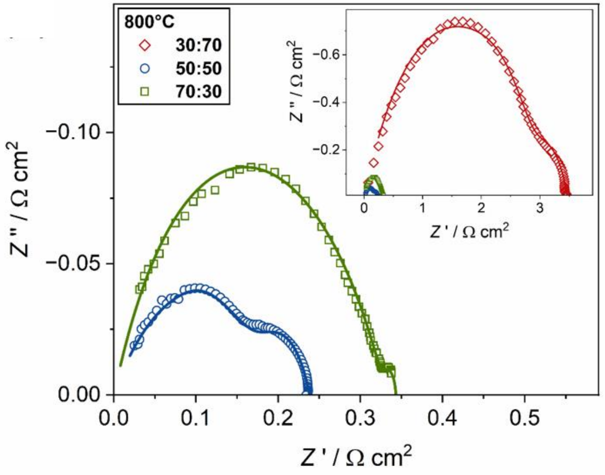


An optimization problem in many parameters!

Microstructure (SEM-EDX)

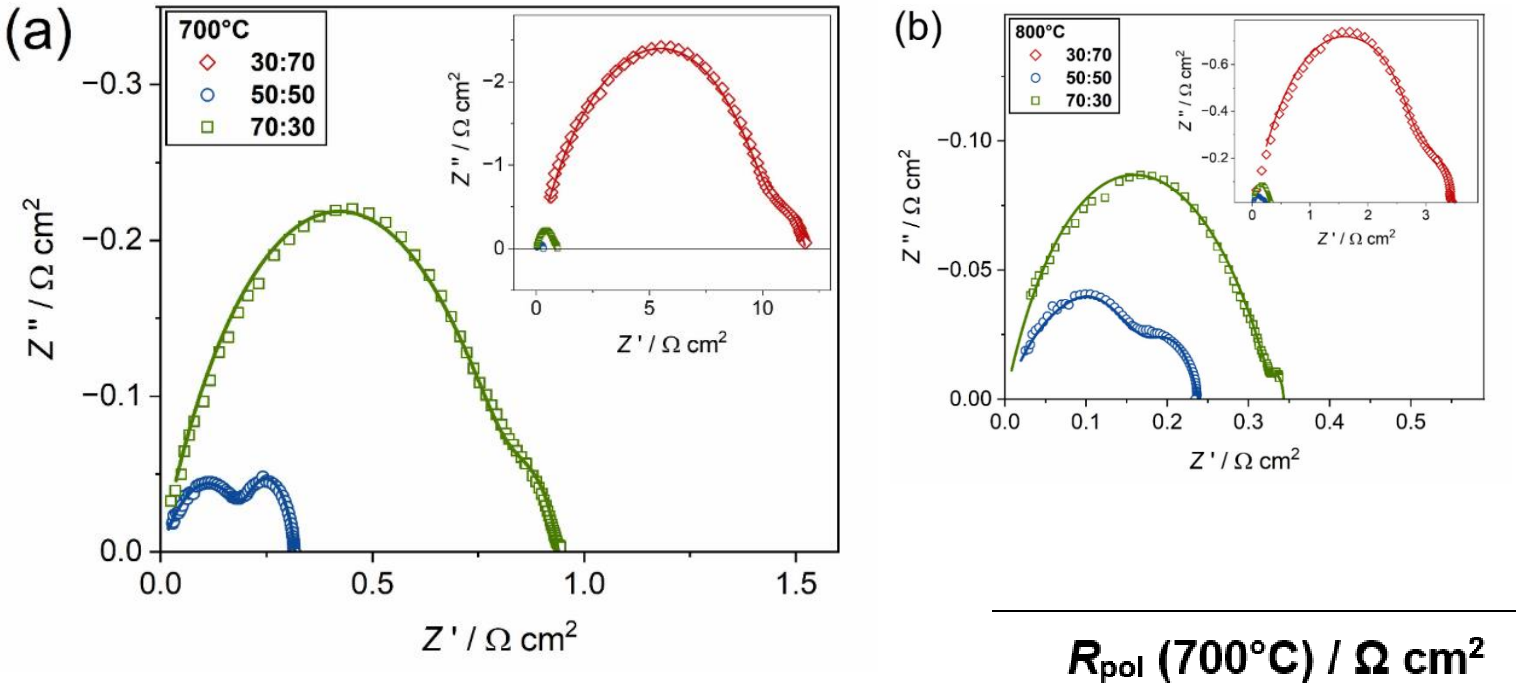


Electrochemical performance



Electrochemical performance

- EIS of symmetric cells with different LSCF-GDC ratio electrodes

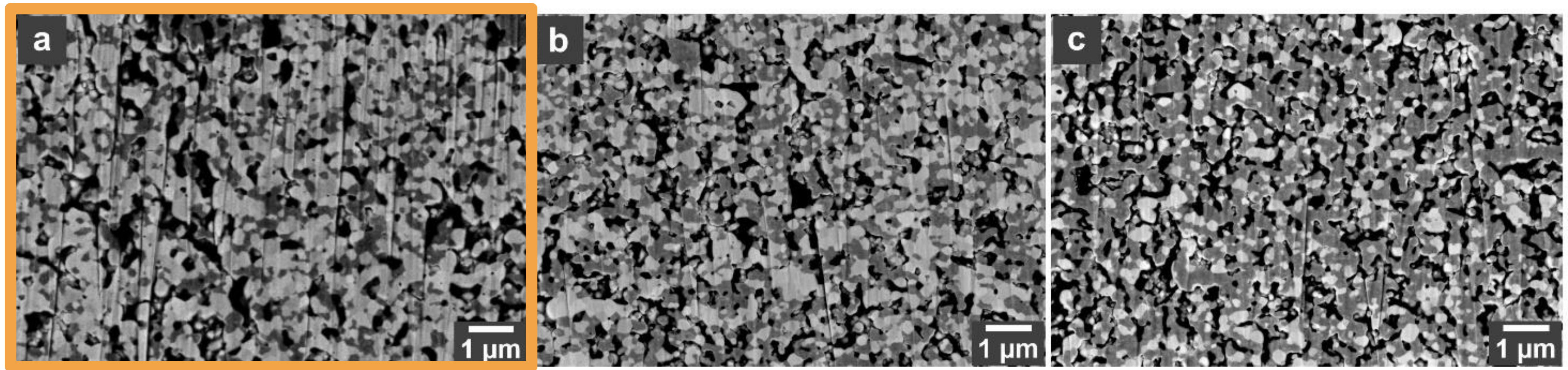


	LSCF content / wt.%		
	30	50	70
R_{pol} (700°C) / Ω cm²	11.9	0.32	0.94
R_{pol} (800°C) / Ω cm²	3.46	0.24	0.34

EIS … Electrochemical impedance spectroscopy
LSCF … $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$
GDC … $Gd_{0.1}Ce_{0.9}O_{2-\delta}$

2D electrode morphology

- FESEM-BSE of electrodes with different LSCF-GDC ratios



(a) 30 wt% LSCF

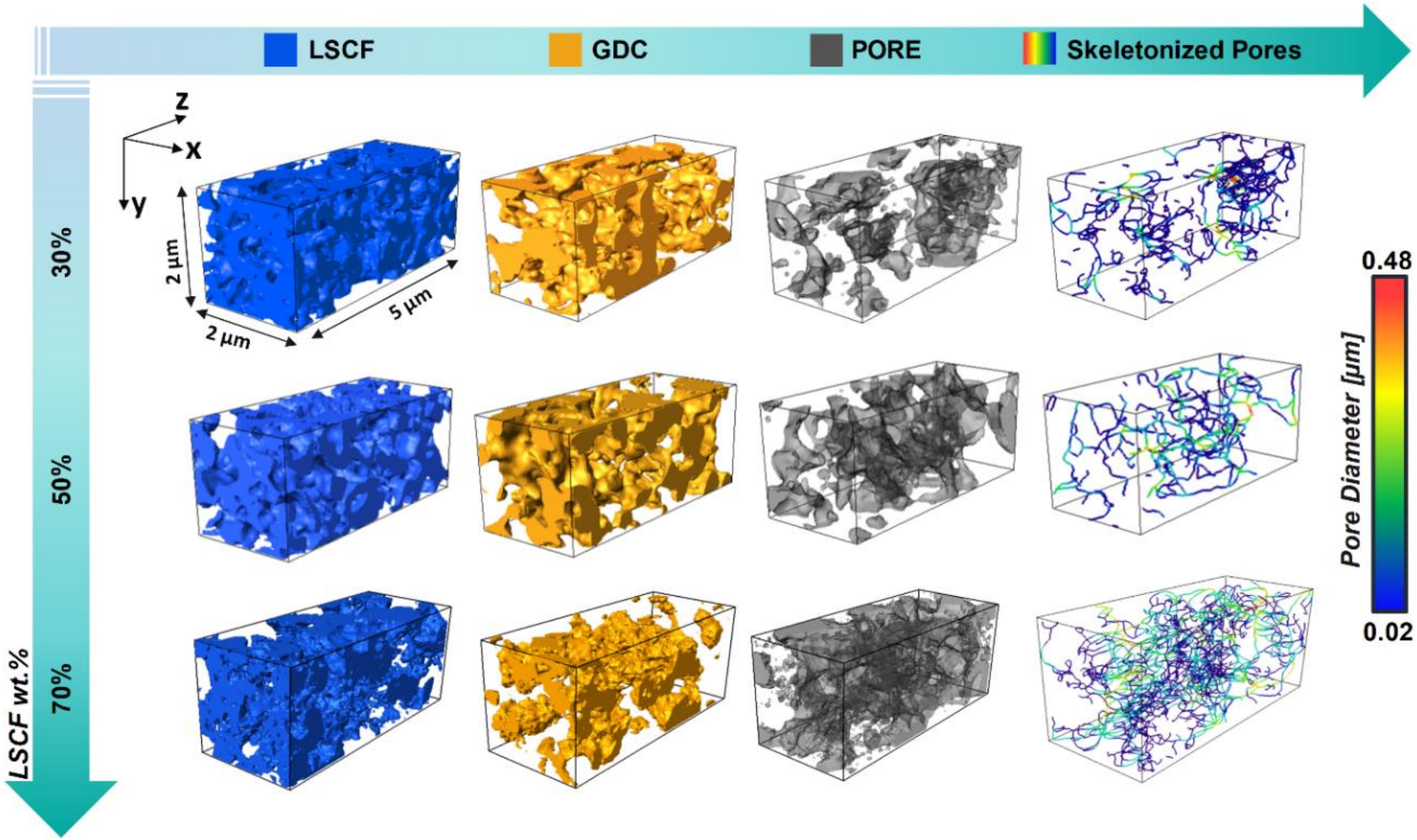
(b) 50 wt% LSCF

(c) 70 wt% LSCF

EDX

LSCF … $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$
GDC … $Gd_{0.1}Ce_{0.9}O_{2-\delta}$

3D electrode morphology



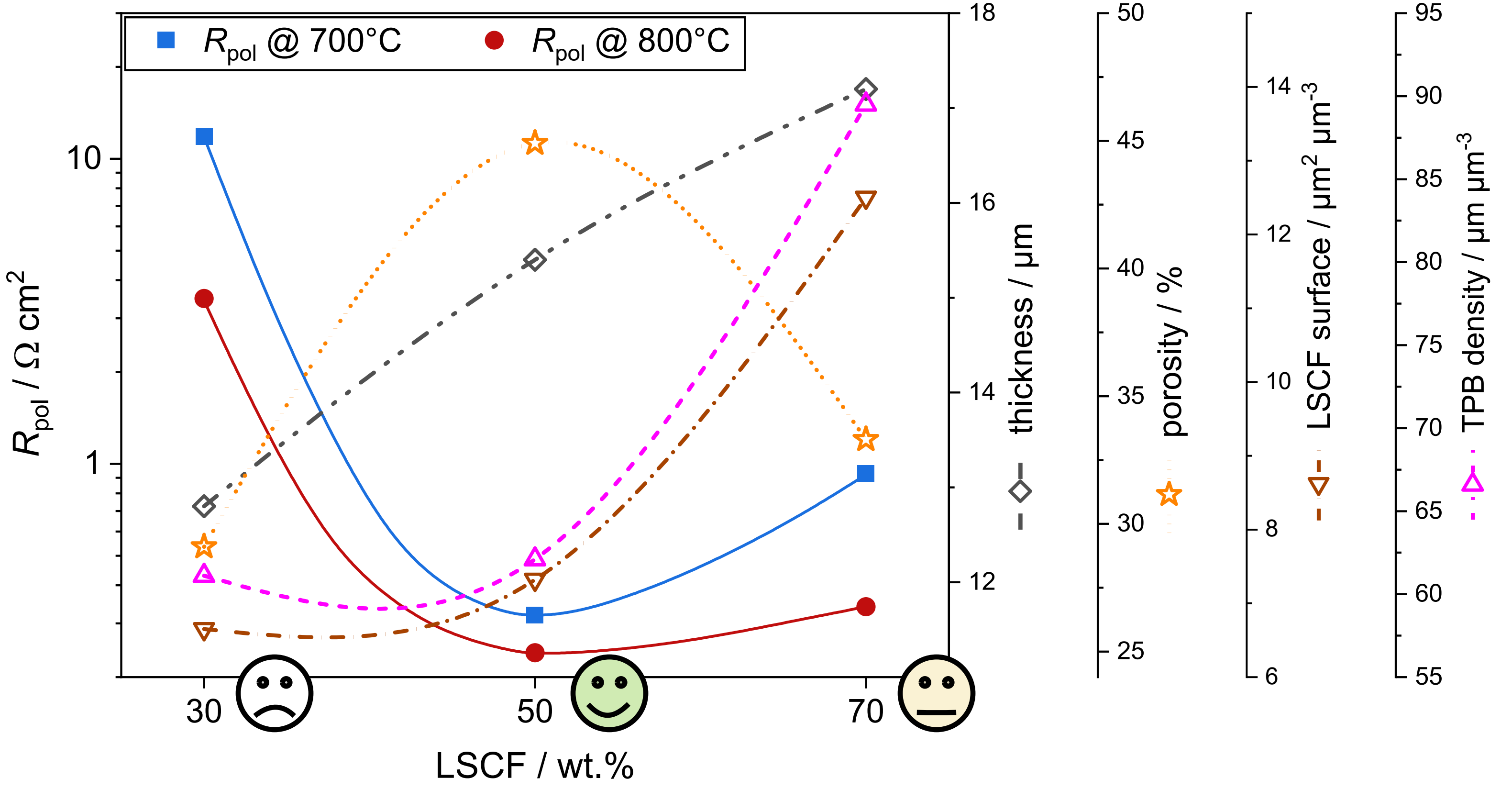
3D-reconstructed microstructures of LSCF-GDC electrodes with different phase ratios

(FIB-SEM, VOI 2×2×5 µm^3)

LSCF … $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$
GDC … $Gd_{0.1}Ce_{0.9}O_{2-\delta}$

3D electrode morphology

- Features of composite electrodes with different LSCF-GDC phase ratios

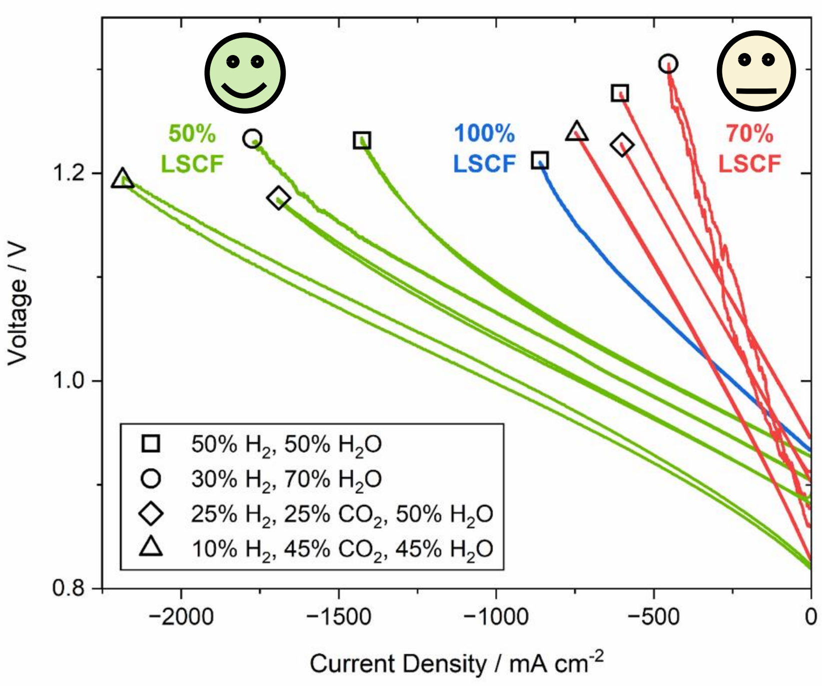


TPB … triple phase boundary
LSCF … $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$
GDC … $Gd_{0.1}Ce_{0.9}O_{2-\delta}$

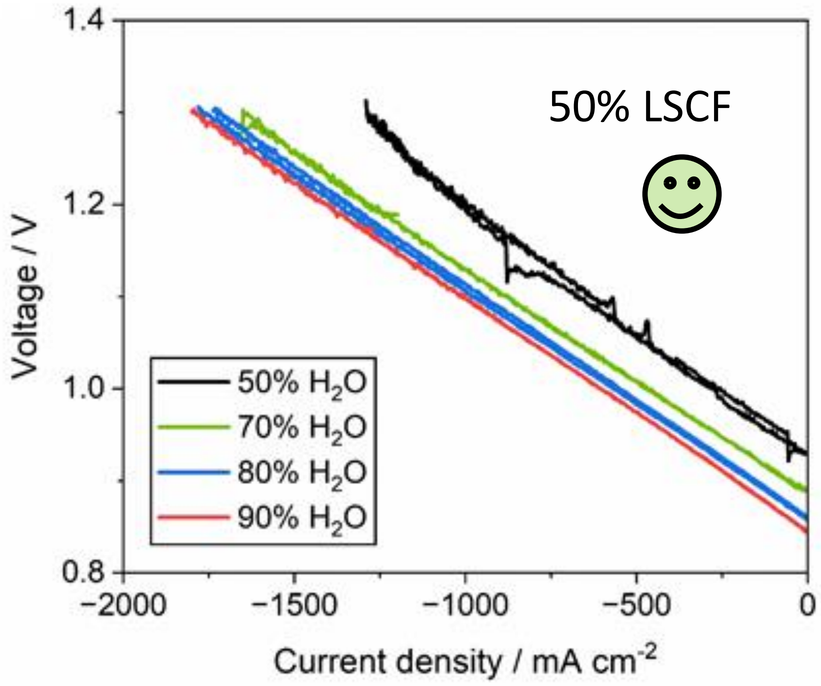
Electrochemical performance

- Current-voltage curves of anode-supported full cells at 800°C

2 cm button cell



5×5 cm² cell



2

Development of new air electrodes based on uncritical raw materials

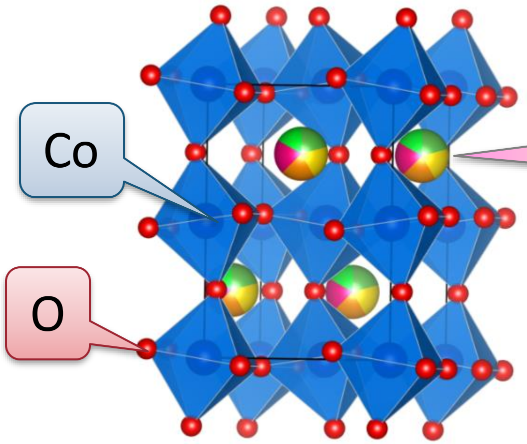
DESIGN-SOEC

Cooperation with Materials Center Leoben Forschung GmbH & AVL List GmbH

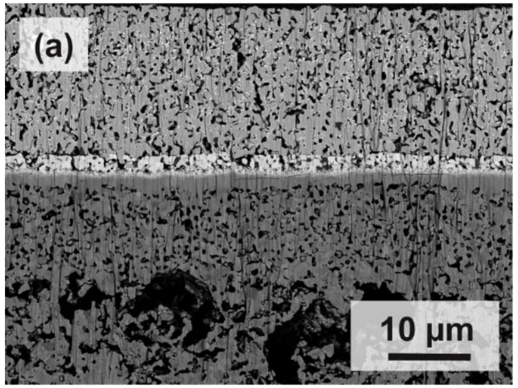
FFG Forschung wirkt.

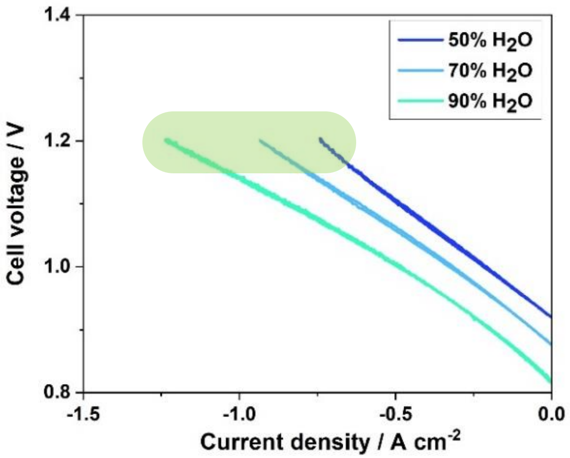
High-entropy perovskites ($\Delta S_{config} \geq 1.5R$)

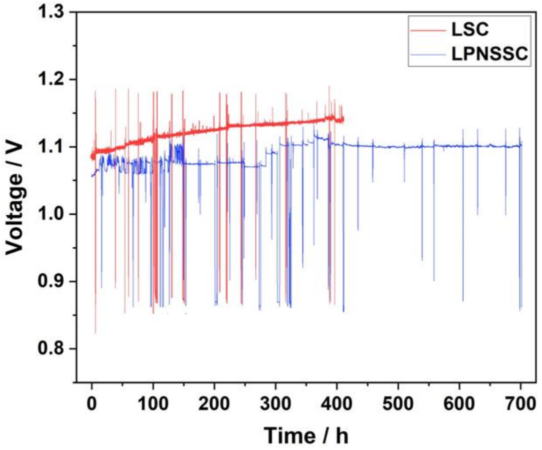
- $La_{0.2}Pr_{0.2}Nd_{0.2}Sm_{0.2}Sr_{0.2}CoO_{3-\delta}$
  - \+ Single-phase perovskite
  - \+ Excellent electrolysis performance
  - \+ High long-term stability
  - – Contains Co (critical element)



Crystal structure of LPNSSC, SG: *Pnma*, $S_{conf} = 1.61\,R$







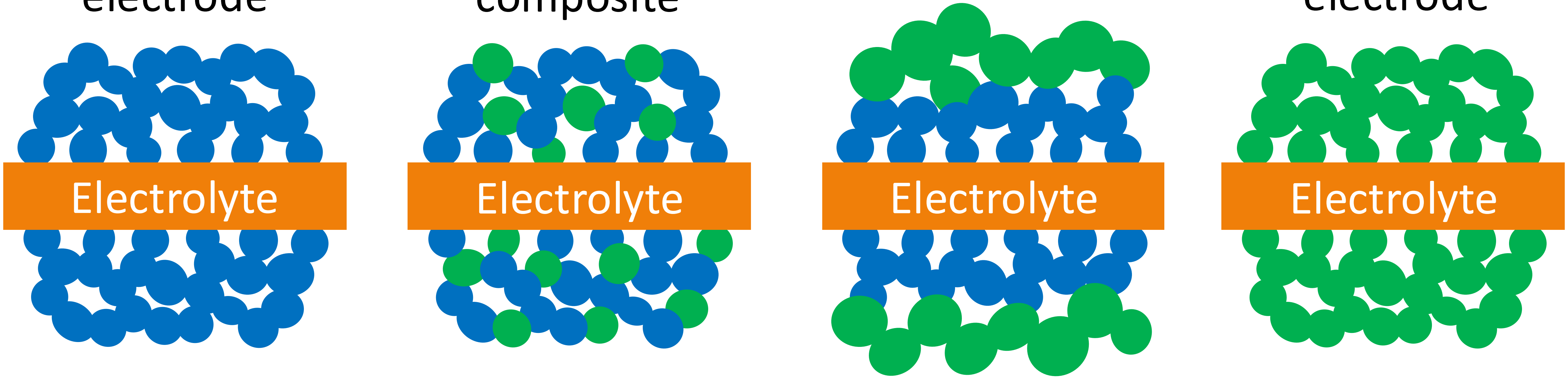
P. Pretschuh, A. Egger, R. Brunner, E. Bucher: Fuel Cells 23 (2023) 6, 377-386, https://doi.org/10.1002/fuce.202300036

$La_{0.2}Pr_{0.2}Nd_{0.2}Sm_{0.2}Sr_{0.2}\mathbf{Co}O_{3-\delta} \rightarrow La_{0.2}Pr_{0.2}Nd_{0.2}Sm_{0.2}Sr_{0.2}\mathbf{Fe}O_{3-\delta}$?

Property	LPNSS**Co**$O_{3-\delta}$	LPNSS**Fe**$O_{3-\delta}$
Single-phase perovskite	YES	YES
High electronic conductivity (800°C)	1500 S cm^{-1}	90 S cm^{-1} ☹
High ionic conductivity	YES	YES
High catalytic activity	YES	YES
High electrochemical performance	YES	?
High long-term stability	YES	?
Critical elements, expensive, issues with disposal (hazardous)	Cobalt ☹	No cobalt

Porous electrodes on symmetric cells

Pure LPNSSF electrode | LPNSSF-LNF composite | Pure LPNSSF LNF on top | Pure LNF electrode

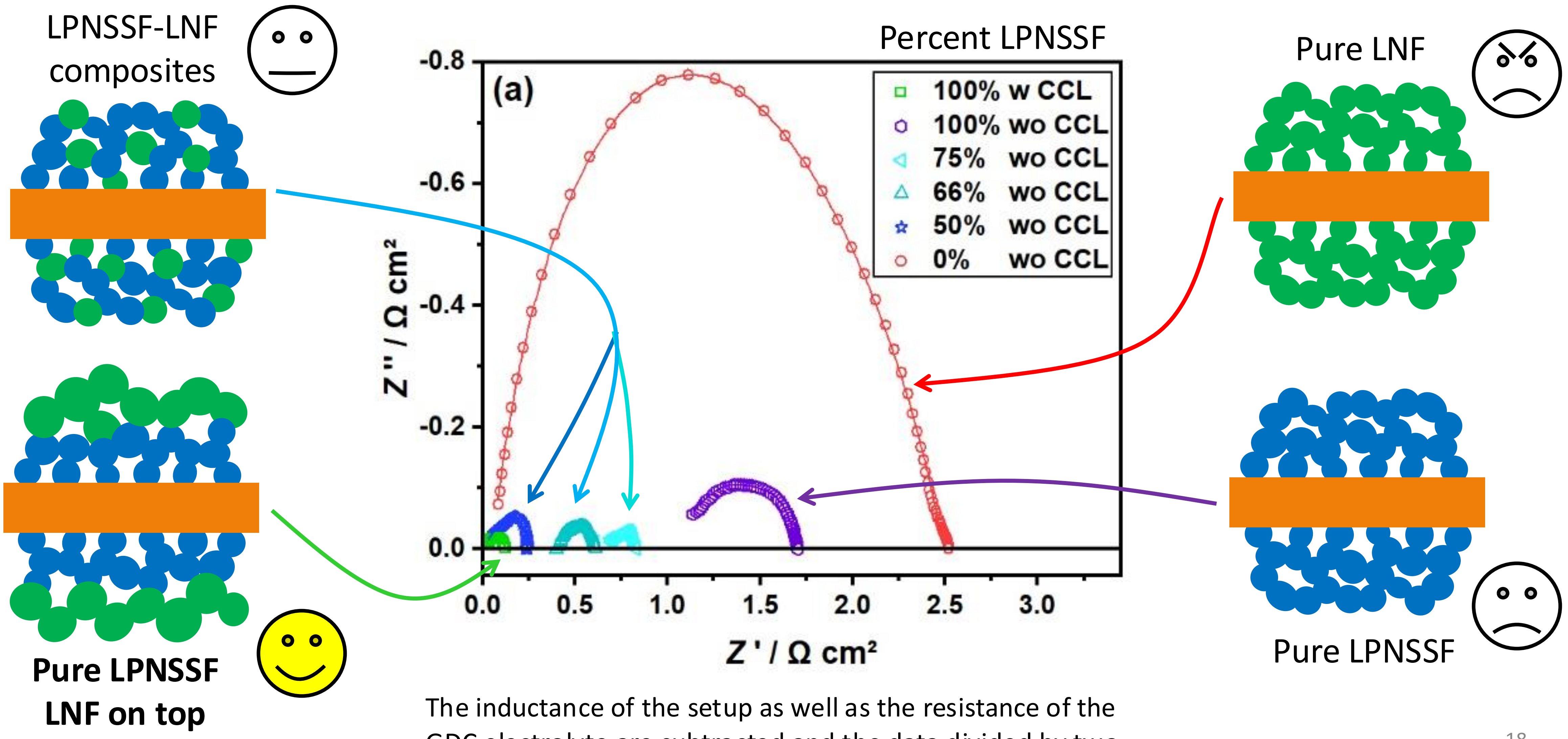


$La_{0.2}Pr_{0.2}Nd_{0.2}Sm_{0.2}Sr_{0.2}FeO_{3-\delta}$ (90 S cm^{-1})

$LaNi_{0.6}Fe_{0.4}O_{3-\delta}$ (580 S cm^{-1})

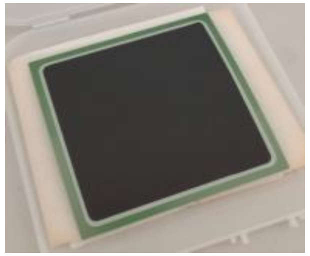
$Gd_{0.1}Ce_{0.9}O_{2-\delta}$

Impedance spectra at 800°C

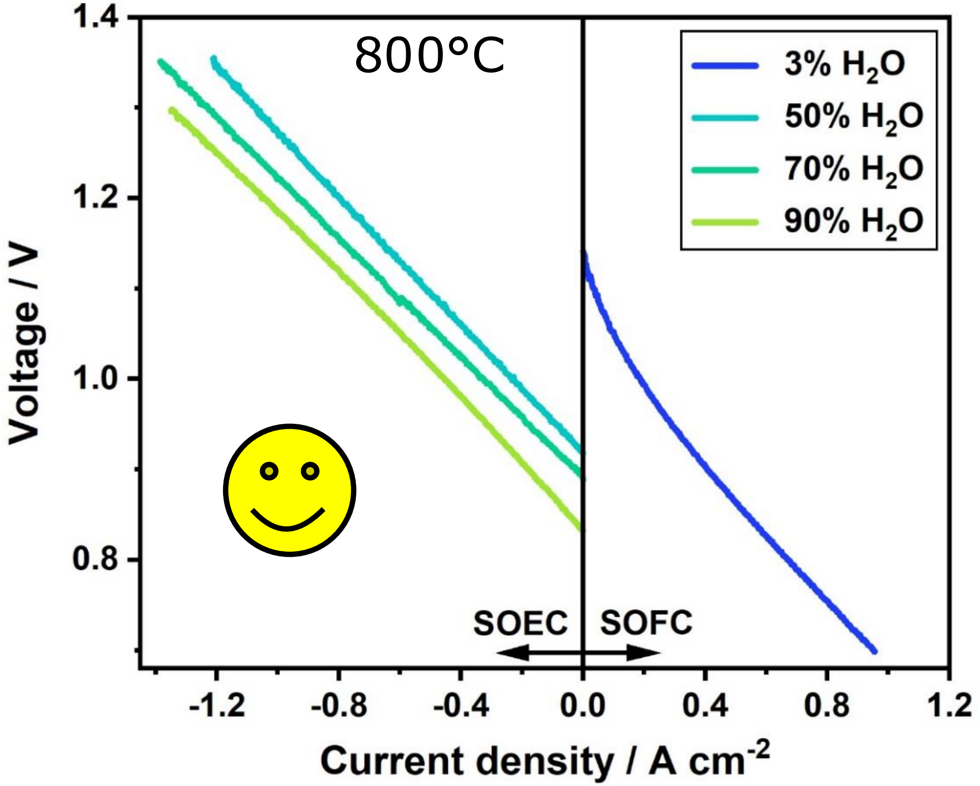


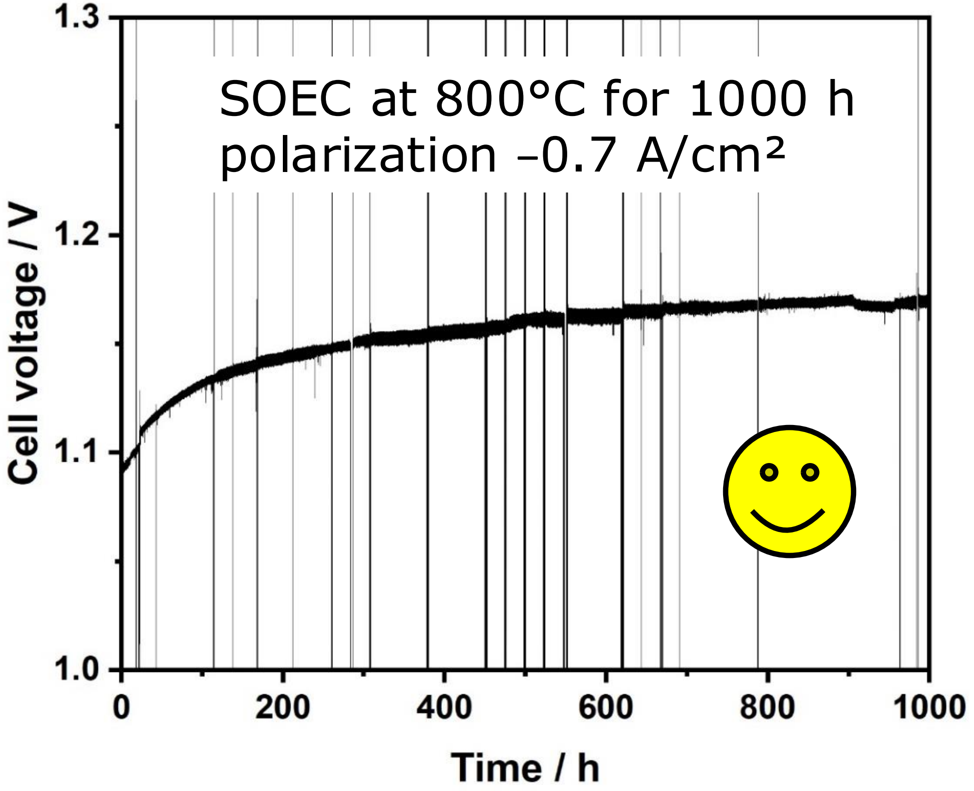
The inductance of the setup as well as the resistance of the GDC electrolyte are subtracted and the data divided by two.

$La_{0.2}Pr_{0.2}Nd_{0.2}Sm_{0.2}Sr_{0.2}FeO_{3-\delta}$ on 5×5 cm² cell



Air electrode: pure LPNSSF, LNF on top



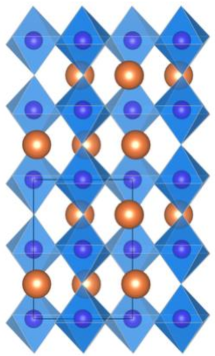


High current density (electrolysis)

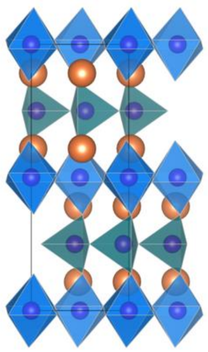
High long-term stability (electrolysis)

Electrodes without any critical elements?

- Avoid not only Co – but also rare earth elements and strontium
- *Uncritical* raw materials
  - $B^{3+/4+/2+}$: Fe, Mn, Ni, Cu
  - A^{2+}: Ca
  - A^{3+}: …?
  - X^{2-}: O



Perovskite
$ABO_{3-\delta}$



Brownmillerite
$A_2B_2O_{5-\delta}$

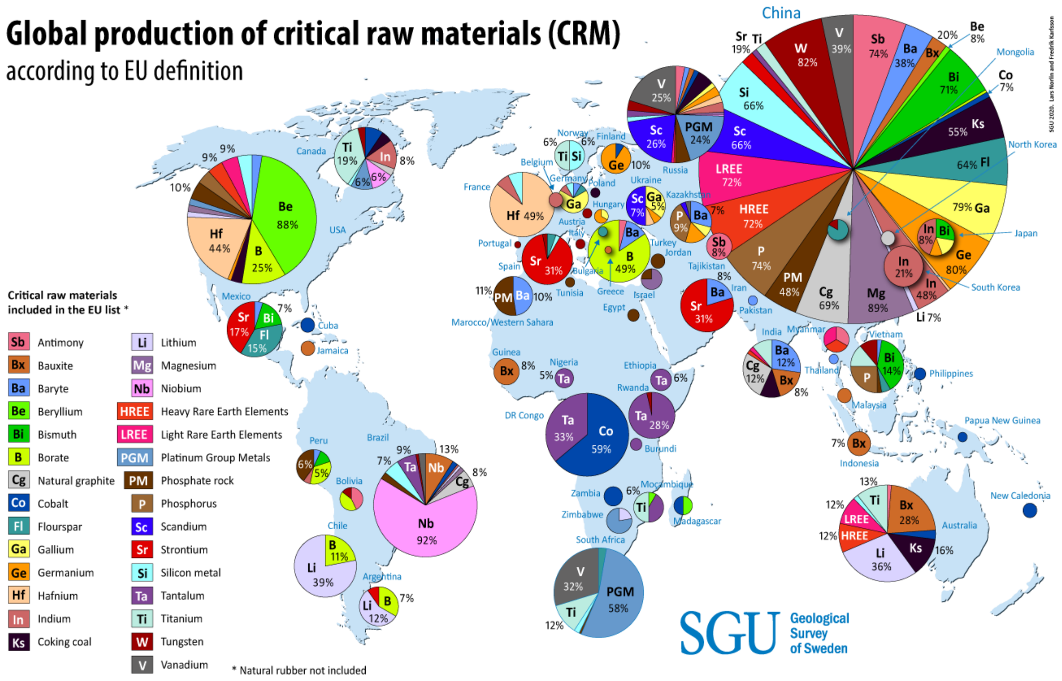
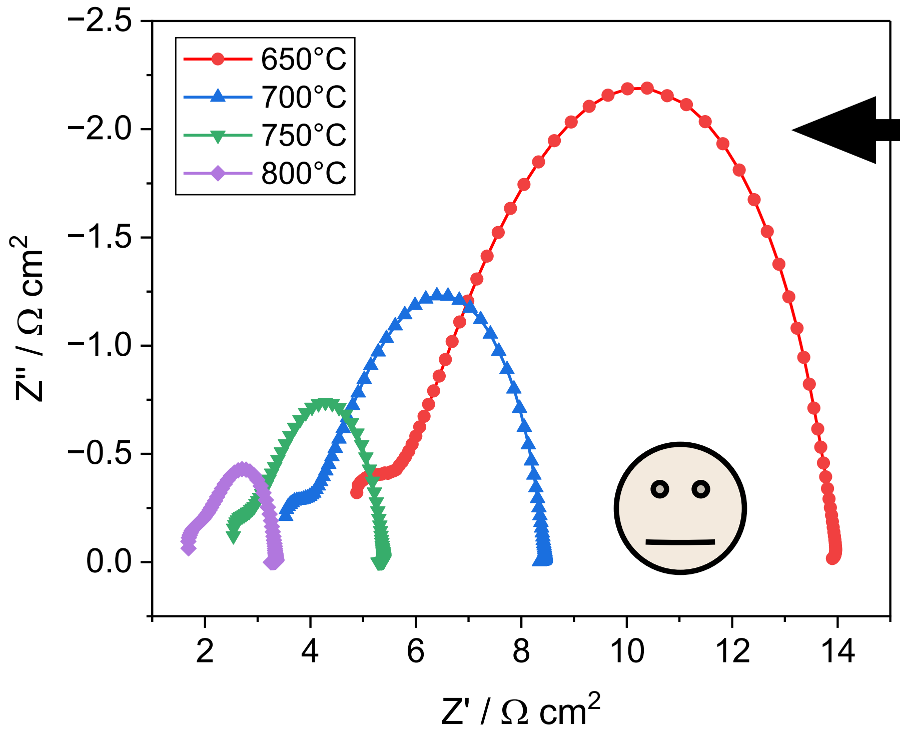
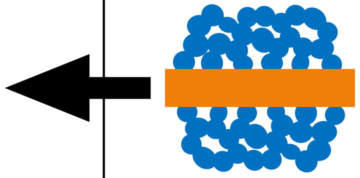


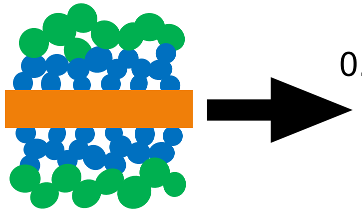
Image: https://www.sgu.se/en/mineral-resources/critical-raw-materials/

Brownmillerite-based materials

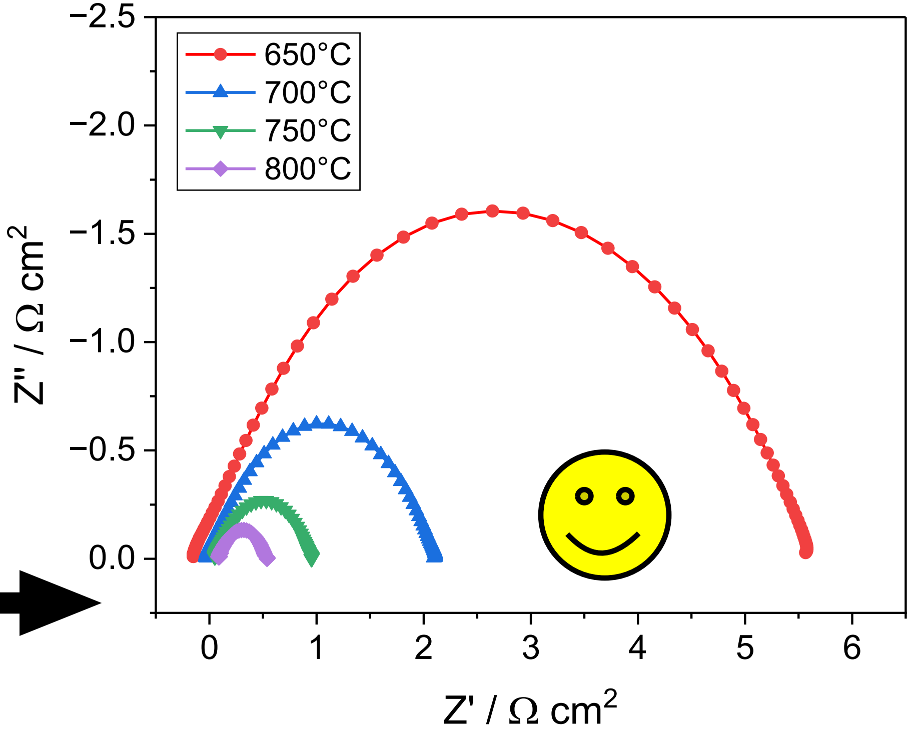
Symmetric cell CFO I GDC I CFO







Symmetric cell LNF I CFO I GDC I CFO I LNF



$Ca_2Fe_2O_5$ single-phase <u>wo</u> CCL

$Ca_2Fe_2O_5$ single-phase <u>w</u> CCL

The inductance of the setup as well as the resistance of the GDC electrolyte are subtracted and the data divided by two.

3

Advanced fabrication methods for unconventional cell designs

ADDEUS
SOLID OXIDE CELL

Cooperation with Chair of Structural and Functional Ceramics at MUL & Lithoz GmbH

FFG
Forschung wirkt.

AddEus – consortium and tasks

LITHOZ — Method development & additive manufacturing

- Development of printing processes
- Development of slurries
- Development of sintering processes
- Printing of fine structures
- Printing of new materials (esp. GDC)

ISFK — Processing & mechanical characterization

- Development of multi-material tape casting
- Development of graded electrolytes
- Mechanical properties of thin electrolyte substrates
- Microstructure tailoring, incl. non-conventional sintering

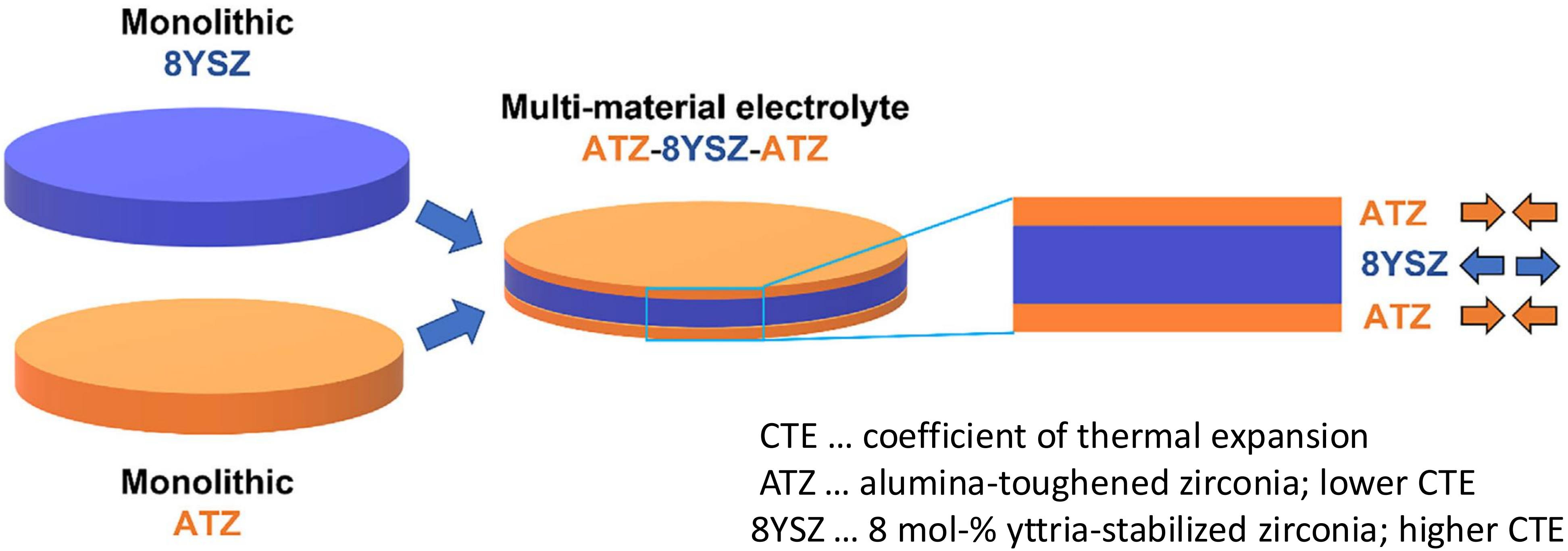
LPC physical chemistry — Synthesis & electrochemical characterization

- Synthesis of new electrode materials, esp. cobalt-free electrodes
- Preparation of porous electrodes, diffusion barriers and current collectors (screen printing)
- Ionic conductivity measurements
- Electrochemical cell tests

Maximizing cell performance, reliability and stability, as well as independence from critical raw materials

Multimaterial electrolytes

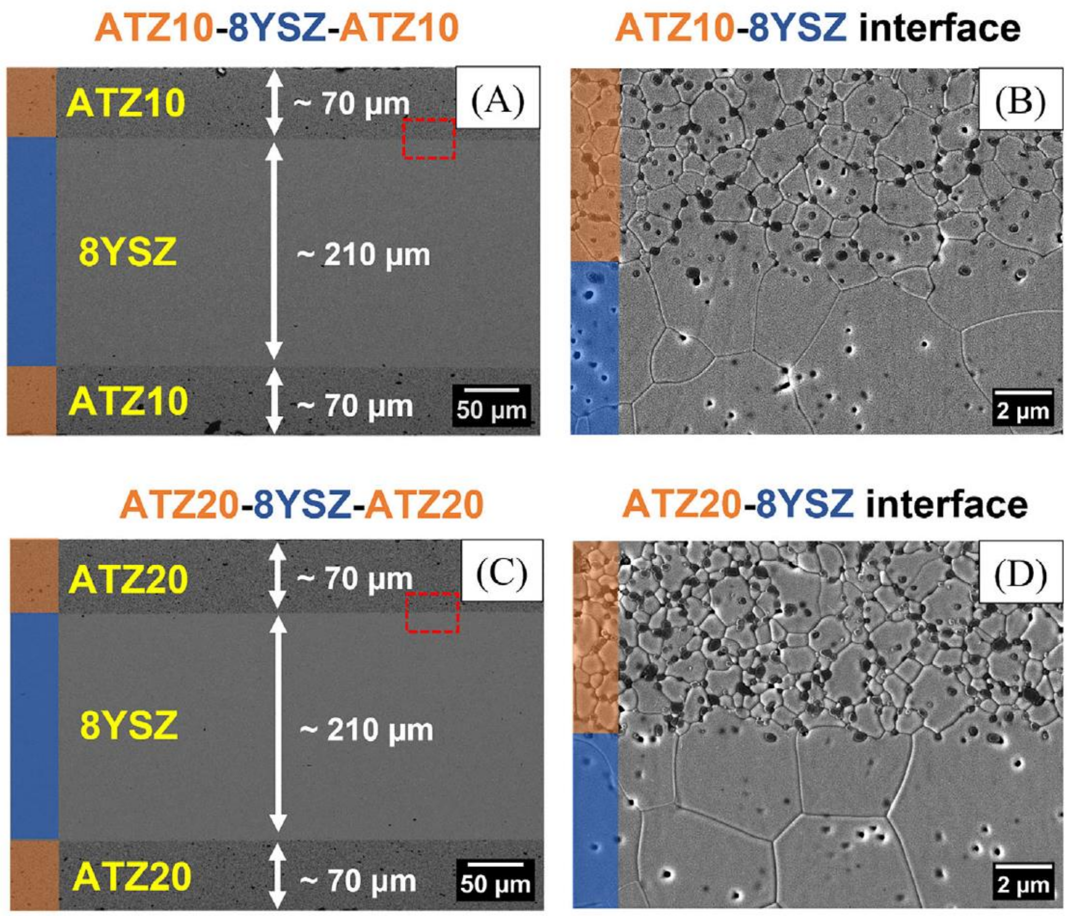
- In-plane compressive (ATZ) and tensile (8YSZ) residual stresses develop during cooling from sintering temperature



CTE … coefficient of thermal expansion
ATZ … alumina-toughened zirconia; lower CTE
8YSZ … 8 mol-% yttria-stabilized zirconia; higher CTE

Multimaterial electrolytes

- SEM microstructure images of multimaterial architectures
  (a) ATZ10–8YSZ–ATZ10
  (c) ATZ20–8YSZ–ATZ20
- Magnified views of interfaces confirm excellent bonding
  (b) ATZ10–8YSZ
  (d) ATZ20–8YSZ

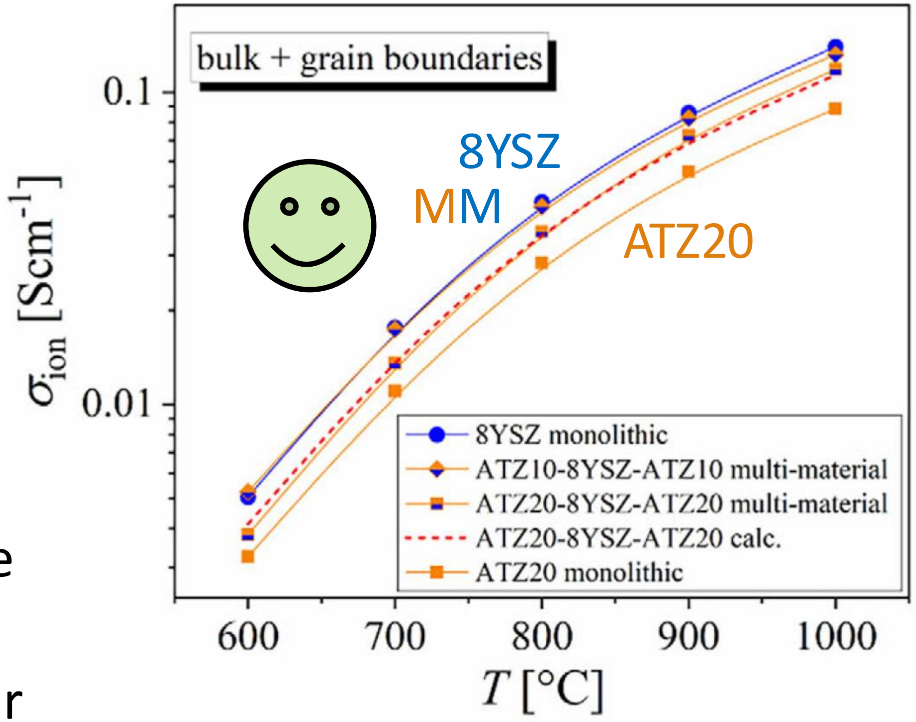


Multimaterial electrolytes

- Characteristic strength σ_0 and ionic conductivity σ_{ion}

Specimen set	**σ_0 (MPa)**	**m (–)**
8YSZ	650 [587–721]	5 [3–6]
ATZ10	767 [716–823]	7 [5–10]
ATZ20	733 [695–775]	9 [6–12]
ATZ10–8YSZ–ATZ10	771 [713–835]	6 [4–8]
ATZ20–8YSZ–ATZ20	853 [794–917]	7 [5–9]

Values in square brackets correspond to 90% confidence intervals.



- ATZ improves mechanical strength of 8YSZ-electrolyte by up to ~30%
- Decrease in ionic conductivity is small, even for higher Al_2O_3-content

Publications

[1] B. Buxbaum, A. Egger, J. Kramer-Schöggl, F.F. Chamasemani, R. Brunner, E. Bucher, *Electrochemical performance and advanced 3D reconstruction of LSCF-GDC air electrodes for solid oxide electrolysis cells*, Journal of Power Sources (2025), https://doi.org/10.1016/j.jpowsour.2025.238174

[2] P. Pretschuh, A. Egger, P. Paulachan, J. Schöggl, R. Brunner, E. Bucher, *Cobalt-free high-entropy perovskite* $La_{0.2}Pr_{0.2}Nd_{0.2}Sm_{0.2}Sr_{0.2}FeO_{3-\delta}$ *solid oxide cell air electrode with enhanced performance*, Fuel Cells 24 (3) (2024) e202400068, https://doi.org/10.1002/fuce.202400068

[3] A. Jana, J. Schlacher, I. Kraleva, A. Egger, E. Bucher, R. Bermejo, *Tailoring strength and ionic conductivity in zirconia-based solid oxide electrolytes using a multimaterial approach*, Journal of the American Ceramic Society (2025) e20632, https://doi.org/10.1111/jace.20632

Find out more …

Research group Bucher at Montanuniversität Leoben

- Webpage: https://physchem.unileoben.ac.at/forschung/research-group-bucher

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