Optimizing the performance of solid oxide electrolysis cells (SOECs) and stacks for Power-to-Gas applications

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**Motivation and Background:**
Thermal integration of HT-electrolysis with catalytic methanation

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**Low-temperature** electrolysis (PEM, alkaline):  
\[ \text{H}_2\text{O}(l) \rightarrow \text{H}_2 + 0.5 \text{O}_2, \Delta H^\circ = 286 \text{ kJ mol}^{-1} \ (1.48 \text{ V}) \]

**High-temperature** electrolysis (SOEC):  
\[ \text{H}_2\text{O}(g) \rightarrow \text{H}_2 + 0.5 \text{O}_2, \Delta H^\circ = 242 \text{ kJ mol}^{-1} \ (1.29 \text{ V}) \]

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**Figure:** Adapted from [1].

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**Table:** Advantages and R&D demands of high-temperature (HT) solid oxide electrolysis cells (SOECs) for Power-to-Methane (PtM) applications.

<table>
<thead>
<tr>
<th>Advantages</th>
<th>R&amp;D requirements</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Thermodynamic:</strong></td>
<td>Scale-Up</td>
</tr>
<tr>
<td>High efficiency</td>
<td></td>
</tr>
<tr>
<td><strong>Kinetic:</strong></td>
<td>Optimization of process parameters</td>
</tr>
<tr>
<td>Thermal barriers</td>
<td></td>
</tr>
<tr>
<td><strong>Thermal integration possible</strong></td>
<td>Selection of optimal cell design</td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Promising perspectives regarding investment costs</strong></td>
<td>Degradation</td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Dynamics</td>
</tr>
</tbody>
</table>

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Methodology:
Multi-scale modeling framework

Planar RU (Repeating Unit)
2D modelling approach (axial + radial)
without interconnects, adapted cell area → single cell simulations

Electrolyte: dense, O²⁻-conductor; Y-doped ZrO₂ (YSZ)
Anode: porous, single phase O²⁻ and e⁻-mixed conductor; doped perovskite (LSCF, LSC)
Barrier layer: O²⁻-conductor; Gd-doped CeO₂ (CGO)
Cathode: porous, Ni cermet; doped CeO₂ or ZrO₂ (Ni-YSZ, Ni-CGO)
Gas channel

Ferritic interconnector: Crofer® 22 APU steel

MEA

Temperature profile
Heat source terms

Methodology:
From single cell to system level

- Testing of commercial SOEC cells
  - electrode + electrolyte supported configurations

- SOEC cell model adaption
  - calibration & validation procedure
  - Performance analysis
  - Optimize operation conditions

- Scale-up to stack level
  - Optimize operation conditions
  - Correlate performance with single cell level

- Scale-up to integrated PtM system model
  - integrate developed SOEC stack model
  - use Matlab® Simulink
  - assess and compare different process chains for CNG/LNG production

this presentation
Electrochemical characterization of single cells: Test facility at the ENERMAT laboratory

- Test bench from Fiaxell
- Sealed cell housing
  - Avoid thermal reaction between H₂ and air
  - Removal of product gas
- Coupling of test bench to analysis systems
  - Product gas flow via MFCs
  - Polarization curves (iV) and Electrochemical Impedance Spectroscopy (EIS)
## Model parametrization: Microstructural analysis

### Table: Model input parameters for isothermal single cell simulations. Units of partial pressures are given in atm. fl: functional layer, dl: diffusion layer.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Cell parameters</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Active cell area</td>
<td>10.18 cm²</td>
<td>Measurement</td>
</tr>
<tr>
<td>Cell length</td>
<td>1.8 cm</td>
<td>Measurement</td>
</tr>
<tr>
<td>Channel height</td>
<td>1.0 mm</td>
<td>Measurement</td>
</tr>
<tr>
<td>Channel width</td>
<td>5.65 mm</td>
<td>Calculated</td>
</tr>
<tr>
<td>Cathode thickness (fl/dl)</td>
<td>7.2 / 25.0 μm</td>
<td>SEM</td>
</tr>
<tr>
<td>Anode thickness</td>
<td>47.5 μm</td>
<td>SEM</td>
</tr>
<tr>
<td>Electrolyte/Barrier layer thickness</td>
<td>81.0 / 7.0 μm</td>
<td>SEM</td>
</tr>
<tr>
<td><strong>Microstructural parameters</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cathode porosity (fl/dl)</td>
<td>0.2 / 0.3</td>
<td>SEM</td>
</tr>
<tr>
<td>Anode porosity</td>
<td>0.2</td>
<td>SEM</td>
</tr>
<tr>
<td>Cathode particle diameter (fl/dl)</td>
<td>0.44 μm</td>
<td>SEM</td>
</tr>
<tr>
<td>Anode particle diameter</td>
<td>0.4 μm</td>
<td>SEM</td>
</tr>
<tr>
<td>N\text{\textdegree} volume fraction in cathode</td>
<td>0.5</td>
<td>EDX</td>
</tr>
<tr>
<td>Ni electronic conductivity</td>
<td>3.27×10^{-1}×10653+T S cm\textsuperscript{-1}</td>
<td>[4]</td>
</tr>
<tr>
<td>YSZ ionic conductivity</td>
<td>334×e\textsuperscript{-2336T} S cm\textsuperscript{-1}</td>
<td>[5]</td>
</tr>
<tr>
<td>YSZ ionic conductivity</td>
<td>37.8×e\textsuperscript{444T} S cm\textsuperscript{-1}</td>
<td>[6]</td>
</tr>
<tr>
<td>CGO ionic conductivity</td>
<td>(1.09×10\textsuperscript{-7})×e\textsuperscript{-3.499T}(1+150.05) S cm\textsuperscript{-1}</td>
<td>[7, 8]</td>
</tr>
<tr>
<td>Electronic conductivity</td>
<td>(3.46×10\textsuperscript{-7})×e\textsuperscript{-8777T}×e\textsuperscript{-0.235T} S cm\textsuperscript{-1}</td>
<td>[7]</td>
</tr>
<tr>
<td>Extra oxygen vacancies</td>
<td>1.3×10\textsuperscript{-8}×e\textsuperscript{0.385T}×e\textsuperscript{-0.265}</td>
<td>[7]</td>
</tr>
<tr>
<td>LSC Electronic conductivity</td>
<td>4860.1-2.996×10\textsuperscript{T} S cm\textsuperscript{-1}</td>
<td>[9]</td>
</tr>
<tr>
<td>LSC Ionic conductivity</td>
<td>1.31×10\textsuperscript{-6}×e\textsuperscript{-0.079T} S cm\textsuperscript{-1}</td>
<td>[10]</td>
</tr>
<tr>
<td>LSCF Electronic conductivity</td>
<td>983.25-0.627×10\textsuperscript{T} S cm\textsuperscript{-1}, T &gt; 923.15 K</td>
<td>[11]</td>
</tr>
<tr>
<td>LSCF Ionic conductivity</td>
<td>7.35×10\textsuperscript{-6}×e\textsuperscript{-1489T} S cm\textsuperscript{-1}</td>
<td>[12]</td>
</tr>
</tbody>
</table>

**References:**

[1] ESC: Electrolyte-supported cell (ESC)
[2] CSC: Cathode-supported cell (CSC)
[3] 3YSZ
[4] Ni
[5] YSZ
[6] CGO
[7] Ni-CGO
[8] dl
[9] fl
[10] Ni
[12] CGO
Model calibration and validation:
Extract electrokinetics of half-cell reactions

- Model able to reach a near-quantitative accordance with experimental iV-data for both cell types
- Activation energies and pressure exponents (preferably from high-pressure data) for all materials used from literature

**Figure**: Comparison of simulated (continuous) and measured (dotted) polarization curves of a cathode-supported cell (CSC) at $p = 1$ atm, $T = 600$-$750 \, ^{\circ}C$, $90/10 \, H_2O/H_2$, air.

**CSC**: Ni-YSZ|YSZ|CGO|LSC, $A_{cell} = 10.18 \, cm^2$

**ESC**: Ni-CGO|3YSZ|CGO|LSCF, $A_{cell} = 10.18 \, cm^2$

$p = 1$ atm, $90/10 \, H_2O/H_2$, air
Model calibration and validation:
Extract electrokinetics of half-cell reactions

Figure: Comparison of simulated and measured ohmic resistances of the ESC and CSC single cells. Experimental ASR values were extracted from EIS measurements (HF intercepts) at OCV at different temperatures. Aside from the ohmic resistance of the electrolyte itself, simulated ASR values also contain the ohmic resistance contributions due to charge transport across the electrodes.

Table: Butler-Volmer fit parameters acquired from the single cell calibration procedure. Units of partial pressures are given in atm.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>ESC</th>
<th>Value</th>
<th>CSC</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cell type</td>
<td>Ni-CGO</td>
<td>Ni-YSZ</td>
<td></td>
</tr>
<tr>
<td>Exchange current density</td>
<td>$5.9 \times 10^3 \times e^{-111900/RT} \times p_{O_2}$</td>
<td>$0.048 \times e^{-105000/RT} \times p_{O_2}$</td>
<td></td>
</tr>
<tr>
<td>Anodic CT-coefficient</td>
<td>1.5</td>
<td>1.18</td>
<td></td>
</tr>
<tr>
<td>Cathodic CT-coefficient</td>
<td>0.5</td>
<td>0.82</td>
<td></td>
</tr>
<tr>
<td>Anode material</td>
<td>LSCF</td>
<td>LSC</td>
<td></td>
</tr>
<tr>
<td>Exchange current density</td>
<td>$1.8 \times 10^5 \times e^{138000/RT} \times p_{O_2}$</td>
<td>$6.0 \times 10^4 \times e^{150000/RT} \times p_{O_2}$</td>
<td></td>
</tr>
<tr>
<td>Anodic CT-coefficient</td>
<td>0.5</td>
<td>1.3</td>
<td></td>
</tr>
<tr>
<td>Cathodic CT-coefficient</td>
<td>0.5</td>
<td>0.7</td>
<td></td>
</tr>
</tbody>
</table>
Model calibration and validation:
Pressurized operation

Pressure influence on physico-chemical processes:

- **Reversible voltage:**
  \[ E_{\text{rev}} = \frac{\Delta G^0}{2F} + \frac{RT}{2F} \ln \left( \frac{p_{\text{H}_2,\text{fe}}p_{\text{O}_2,\text{ae}}^{0.5}}{p_{\text{H}_2\text{O},\text{fe}}} \right) \]

- **Porous medium gas transport:**
  \[ J_k = - \sum_{i=1}^{K_g} D_{kl}^{\text{DGM}} \frac{\partial [X_i]}{\partial y} + \sum_{i=1}^{K_g} \left( \frac{D_{kl}^{\text{DGM}} [X_i]}{D_{i,k}^{\text{eff}}} \right) B_g \frac{\partial p}{\partial y} \]

- **Electrochemical kinetics:**
  \[ i_{0,\text{H}_2/\text{H}_2\text{O}} = A_{\text{H}_2/\text{H}_2\text{O}} \exp \left( - \frac{E_{\text{H}_2/\text{H}_2\text{O}}}{RT} \right) f(p_{\text{H}_2}, p_{\text{H}_2\text{O}}) \]
  \[ i_{0,\text{O}_2} = A_{\text{O}_2} \exp \left( - \frac{E_{\text{O}_2}}{RT} \right) f(p_{\text{O}_2}) \]

- **Electrical conductivities:**
  \( \sigma_{\text{el}}/\sigma_{\text{io}} \) due to stoichiometry changes, oxidation states...

Use literature data:

Figure: Comparison of simulated (continuous) polarization curves with experimental data (dotted) of an cathode-supported Ni-YSZ/YSZ/CGO|LSCF|LSC single cell from Bernadet et al. [13].

\( A_{\text{cell}} = 3.14 \text{ cm}^2, T = 800 \, ^\circ \text{C}, 35/58.5/6.5 \text{ N}_2/\text{H}_2\text{O}/\text{H}_2, \text{ air.} \)

Optimization of operating conditions of SOEC single cells: Parametric analysis

**ESC:** Ni-CGO | 3YSZ | CGO | LSCF, $A_{cell} = 10.18 \text{ cm}^2$

$T = 750 ^\circ \text{C}$, $p = 1 \text{ atm}$, 90/10 $\text{H}_2\text{O}/\text{H}_2$, air

Table: Confrontation of single cell efficiency definitions.

<table>
<thead>
<tr>
<th>$\eta_{\text{tot}}$ (LHV, DC)</th>
<th>$\eta_{\text{elec}}$ (LHV, DC)</th>
</tr>
</thead>
<tbody>
<tr>
<td>With respect to total heat+electrical energy input</td>
<td>With respect to electrical input</td>
</tr>
</tbody>
</table>

System scale (coupling with catalytic methanation, BoP):
- Theoretical minimum; zero thermal integration or recuperation
- Theoretical maximum; zero steam generation or electrical heating required

- **Oppositional trends** between $\eta_{\text{elec}}$ and $V_{H_2}$
- Surpassing conv. of $\sim 80\%$ leads to performance losses due to reactant starvation zones
Optimization of operating conditions of SOEC single cells:
Parametric analysis

**CSC: Ni-YSZ|3YSZ|CGO|LSC**  
\[A_{cell} = 10.18 \text{ cm}^2\]

\[p = 1 \text{ atm}, 90/10 \text{ H}_2\text{O/H}_2, \text{ air}\]

- Efficiency losses at conv. >75%  
  (compared to >80%, KeraCell III)

- Electrode-supported cell design achieves significantly higher performance, e.g.:

<table>
<thead>
<tr>
<th>Operation conditions</th>
<th>Electrode-supported cell</th>
<th>Electrolyte-supported cell</th>
</tr>
</thead>
<tbody>
<tr>
<td>(T_{in} = 750 \degree C, 1 \text{ atm}, 1.3 \text{ V}, 80% \text{ conv.},^\star)</td>
<td>(\dot{V}_{H_2} = 6.5 \text{ SL h}^{-1})</td>
<td>(\dot{V}_{H_2} = 0.8 \text{ SL h}^{-1})</td>
</tr>
<tr>
<td>(\eta_{tot} = 60.4%)</td>
<td>(\eta_{tot} = 60.4%)</td>
<td></td>
</tr>
</tbody>
</table>

\(^\star\)inlet: 90/10 \text{ H}_2\text{O/H}_2, \text{ air}\)

\(\rightarrow\) Pre-Identification of suitable operation windows for stack simulations.
Scale-up from cell to planar stack level:
Simulation parameters

**Table**: Model input parameters for stack simulations.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Stack parameters</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Active cell area</td>
<td>127.8 cm²</td>
<td>[14]</td>
</tr>
<tr>
<td>Active cell length</td>
<td>9 cm</td>
<td>[14]</td>
</tr>
<tr>
<td>Active cell width</td>
<td>14.2 cm</td>
<td>[14]</td>
</tr>
<tr>
<td>Channels per layer</td>
<td>24</td>
<td>[14]</td>
</tr>
<tr>
<td>Channel height</td>
<td>1.0 mm</td>
<td>[14]</td>
</tr>
<tr>
<td>Channel width</td>
<td>4.4 mm</td>
<td>Calculated [14]</td>
</tr>
<tr>
<td>Interconnect thickness</td>
<td>0.5 mm</td>
<td>[14]</td>
</tr>
<tr>
<td>Cells per Stack</td>
<td>150</td>
<td>Simulation</td>
</tr>
<tr>
<td>Insulation layer thickness</td>
<td>5 cm</td>
<td>Simulation</td>
</tr>
<tr>
<td><strong>Cell / Microstructural / Kinetic parameters</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Operating conditions</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Temperature</td>
<td>1073.15-1173.15 K</td>
<td>Simulation</td>
</tr>
<tr>
<td>Pressure</td>
<td>1-20 bar</td>
<td>Simulation</td>
</tr>
<tr>
<td>Inlet gas composition</td>
<td>90-95% H₂O, air</td>
<td>Simulation</td>
</tr>
<tr>
<td>Conversion</td>
<td>75-85%</td>
<td>Simulation</td>
</tr>
</tbody>
</table>

**ESC**: Ni-CGO | 3YSZ | CGO | LSCF, 150-cell stack

\[ T_{in} = 850 ^\circ C, \ E_{cell} = 1.4 \, V, \ p = 1 \, atm, \ 90/10 \, H_2O/H_2, \ \text{air} \]

\[ Q_{loss} \sim 155 \, W \]

Optimization of SOEC stack performance: Temperature and voltage dependency

**ESC:** Ni-CGO│3YSZ│CGO│LSCF, 150-cell stack

$p = 1$ atm, $90/10 \text{H}_2\text{O}/\text{H}_2$, air, 80% conv.

All simulations are performed considering

- Exothermic operation $> 1.29 \text{ V}$
- Endothermic operation $< 1.29 \text{ V}$
- Thermoneutral operation $\approx 1.29 \text{ V}$

Lower/upper bounds:
- $0.4 \leq \frac{V_{\text{air}}}{V_{\text{H}_2+\text{H}_2\text{O}}} \leq 8$
- $0 \leq |\nabla T|_{\text{max}} \leq 10 \text{ K cm}^{-1}$
Optimization of SOEC stack performance:
Pressure dependency

- For the electrolyte-supported cell design, pressurized operation leads to a performance decrease
- Still, the pressure influence is relatively low

**Optimal steady-state operation conditions** for ESC-based SOEC-stack:
- $T_{\text{stack}} \sim 850 ^\circ \text{C}$
- $E_{\text{cell}} = 1.3-1.35 \text{ V}$ (reaching $\eta_{\text{elec}} > 90\%$, moderate temperature gradients, system scale: heat generation for recuperation)
- conv. $\sim 75-85 \%$
- $p$ not too high
- $X_{\text{in,H}_2\text{O}} \sim 90-95\%$
Summary

- Multi-scale modeling framework applied for SOEC cells and stacks performing steam electrolysis for PtM-application
- Electrochemical/microstructural characterization of two different cell designs
- Scale-up to the **150-cell stack-level**, application-oriented simulations
- Performance discrepancy between cell and stack level revealed
- **Optimized operation conditions** on stack-scale identified

Next steps

- Comparison with CSC-based stack-level results
- Scale-up to the integrated system level (Matlab® Simulink)
  - *How do the optimal operation conditions on integrated system scale differ from the one of the SOEC-stack itself (as identified)?*
  - *How large is the influence of the electrode design on net system performance?*
References

Acknowledgements

Financial support by the federal ministry for economic affairs and energy (Bundesministerium für Wirtschaft und Energie, BMWi) under Grant Numbers 03EIV041D and 03EIV041E in the “MethFuel” group of the collaborative research project “MethQuest” is gratefully acknowledged.

Thank you for your attention!