Electrolysis for conversion of H2O and CO2 into green fuels

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Electrolysis for conversion of H$_2$O and CO$_2$ into green fuels

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Department of Energy Conversion and Storage

Sustainable Fuels from Renewable Energies
IASS Brainstorming Workshop
November 19 and 20, 2013
Outline

1. Introduction
2. Green fuels based on hydrogen and carbon
3. Production of green fuels from synthesis gas
4. Production of synthesis gas
5. Electrolyser status
   a. Hydrogen from water/steam electrolysis
   b. Co-electrolysis of \( \text{H}_2\text{O} \) and \( \text{CO}_2 \) using SOEC
6. Economic considerations
7. Conclusions
Introduction

DTU Energy Conversion = Department of Energy Conversion and Storage

- Sustainable technologies for energy conversion and storage
- Located on two campuses near Copenhagen, Denmark: Lyngby and Risø at Roskilde
- 230+ employees
- Our research span from fundamental investigations to component manufacture
- Focus on industrial collaboration and industrially relevant processes
- Created 2012, bringing together research groups from
  - Risø DTU National Laboratory for Sustainable Energy
  - DTU Chemistry
Energy plan for Denmark

2020: 50 % electricity from Renewables

2035: 0 % fossil energy in power and heat production

2050: 100 % renewable energy

Challenges
1) Increasing share of fluctuating production
2) Biomass is limited; liquid “green” fuels will still be needed in order to avoid the situations below:

Heavy transport

Aviation
Green fuels based on hydrogen and carbon

- **Green fuel** is here defined as: CO$_2$ neutral hydrocarbons from which the amount of CO$_2$ emitted by using the fuel ≤ the amount of CO$_2$ used to make the fuel.

- Biomass or “captured CO$_2$ + renewable energy” or at least CO$_2$ free energy (e.g. nuclear).

- Chemically they may be expressed as H$_x$C$_y$O$_z$ – gas, liquid or solid in principle.

- Liquids are emphasized in this presentation
\[ H_xC_yO_z \]

\((x,y,z)\)

\((2,0,0) = H_2\)

\((0,1,1) = CO\)

\((4,1,0) = H_4C = \text{methane}\)

\((4,1,1) = H_4CO = \text{methanol}\)

\((6,2,1) = H_6C_2O = \text{dimethyl ether (DME)}\)

\((18,8,0) = H_{18}C_8 = \text{octane, an important part of gasoline}\)

Most \(H_xC_yO_z\) (higher than CO and \(H_2\)) can be synthesized from syngas (\(CO + H_2\)) using commercial available catalyst. In particular those that are most interesting for energy purposes can.

\(H_xC_yO_z\) can be made using catalysis if syngas is first made by electrochemical reduction (electrolysis) of \(H_2O\) and \(CO_2\)

Even better if \(H_xC_yO_z\) compounds, e.g. \(CH_3OH\), can be made by electrochemical reduction directly on a cathode.
Why liquid synthetic fuel? The power density argument

• Gasoline (33 MJ/L) filling rate of 20 L/min equivalents 11 MW of power and means it takes 2½ min to get 50 l = 1650 MJ on board.

• For comparison: Li-batteries usually requires 8 h to get recharged. For a 300 kg battery package (0.5 MJ/kg) this means a power of ca. 3.5 kW i.e. it takes 8 h to get 150 MJ on board.

• The ratio between their driving ranges is only ca. 5, because the battery-electric-engine has an efficiency of ca. 70 % - the gasoline engine has ca. 25 %.
Production of syngas from $H_2$ and $CO_2$

The water-gas shift (WGS) reaction:

$$CO_2 + 2\ H_2 \rightleftharpoons CO + H_2 + H_2O$$

By condensation of the water pure syngas is obtained
Production of syngas by SOEC co-electrolysis

Reaction Schemes:
The overall reaction for the electrolysis of steam plus CO₂ is:
\[ \text{H}_2\text{O} + \text{CO}_2 + \text{heat} + \text{electricity} \rightarrow \text{H}_2 + \text{CO} + \text{O}_2 \] (1)

This is composed of three partial reactions. At the negative electrode:
\[ \text{H}_2\text{O} + 2\text{e}^- \rightarrow \text{H}_2 + \text{O}^2^- \] (2)
\[ \text{CO}_2 + 2\text{e}^- \rightarrow \text{CO} + \text{O}^2^- \] (3)

and at the positive electrode:
\[ 2 \text{O}^2^- \rightarrow \text{O}_2 + 4\text{e}^- \] (4)
Methanol and DME synthesis

- $\text{CO} + 2 \text{H}_2 \rightleftharpoons \text{CH}_3\text{OH}$

- $2 \text{CO} + 4 \text{H}_2 \rightleftharpoons (\text{CH}_3)\text{O}_2 + \text{H}_2\text{O}$

- A Cu/ZnO-Al$_2$O$_3$ catalyst

- 200 °C - 300 °C

- 4.5 - 6 MPa, again the electrolyser should be pressurized
Visions for synfuels from electrolysis of steam and carbon dioxide

1. Big off-shore wind turbine parks coupled to a large SOEC – produce CH₄ (synthetic natural gas, SNG) - feed into existing natural gas network (in Denmark).

2. Large SOEC systems – produce DME, gasoline and diesel - Island, Canada, Greenland, Argentina, Australia … geothermal, hydro, solar and wind.

3. Target market: replacement of natural gas and liquid fuels for transportation

4. All the infrastructure exists!!
Vision, Biomass CO$_2$ recycling

Short term realisation - CO$_2$ capture from industrial sources

- CO$_2$ Released to the atmosphere
- H$_2$O Released to the atmosphere
- CO$_2$ Concentrated

Fuel synthesis

$2\text{H}_2 + \text{CO} \rightarrow \text{CH}_2 + \text{H}_2\text{O}$

Synthetic petrol/diesel

Fuel transport

- Consumption
- (Renewable) Electricity

Electrolysis cell

$2\text{H}_2\text{O} \rightarrow 2\text{H}_2 + \text{O}_2$

$2\text{CO}_2 \rightarrow 2\text{CO} + \text{O}_2$

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Vision, co-electrolysis for transport fuels

Long term realisation - CO₂ capture from the atmosphere
Motivation for high temperature electrolysis

• Increasing need for easy and affordable storage of intermittent renewable energy ⇒ Hydrogen and hydrogen carriers ⇒

• Electrolysis ⇒

• Cost: a function of inverse electrode reaction rate

• Kinetics and thermodynamic advantage increase with temperature

• Kinetics usually increase with partial pressure of gaseous reactants
Thermodynamics of H₂O electrolysis

H₂O → H₂ + ½O₂

Energy demand (kJ/mol)

Energy demand (kJ/mol) = Energy (kJ/mol) / 2F

Energy (“volt”) = Energy (kJ/mol) / 2F

E_{tn} = \Delta H / 2F

\Delta H / \Delta G > 1 , \quad \varepsilon = 100 \% \quad \text{at} \quad E = E_{tn} \quad \text{(if no heat loss)}

Price \propto \frac{1}{i} \quad \text{[A/cm²]}, \quad \text{i} \propto E_{cell} - \Delta G / 2F

\frac{1}{i} \propto E_{cell} - \Delta G / 2F

\text{E}_{cell} = E_{tn}
Thermodynamics $\text{CO}_2$

$\text{CO}_2 \rightarrow \text{CO} + \frac{1}{2}\text{O}_2$

![Graph showing energy demand in KJ/mol vs. temperature in °C for the reaction $\text{CO}_2 \rightarrow \text{CO} + \frac{1}{2}\text{O}_2$. The graph includes three lines:

- **Total energy demand ($\Delta H_f$)**: A green line representing the total energy demand.
- **Electrical energy demand ($\Delta G_f$)**: A green line representing the electrical energy demand.
- **Heat demand ($T\Delta S_f$)**: A red line representing the heat demand.](image-url)
Thermodynamics: CO$_2$ and H$_2$O

Electrical energy demand ($\Delta G_f$)

$\text{CO}_2 \rightarrow \text{CO} + \frac{1}{2}\text{O}_2$

$\text{H}_2\text{O} \rightarrow \text{H}_2 + \frac{1}{2}\text{O}_2$

750ºC – 900ºC

$\Delta G_{\text{H}_2\text{O}\rightarrow\text{H}_2+\frac{1}{2}\text{O}_2} = \Delta G_{\text{CO}_2\rightarrow\text{CO}+\frac{1}{2}\text{O}_2}$
Electrolysis Cell Types

1. Simple aqueous electrolytes (e.g. KOH, K$_2$CO$_3$, H$_2$SO$_4$), room temperature to ca. 120 °C, 0.1 - 3 MPa pressure

2. Low temperature “solid” proton conductor membrane (PEM), 70 – 90 °C

3. High temperature PEM 120 - 190 °C.

4. Immobilized aqueous K$_2$CO$_3$, Na$_2$CO$_3$, K$_2$SO$_4$ etc. in mesoporous structures – pressurized 200 – 300 °C, 0.3 – 10 MPa

5. Solid acids (e.g. CsH$_2$PO$_4$), 200 – 250 °C, pressurized?

6. Molten carbonate electrolytes, 800 – 950 °C, 0.1 MPa, only CO$_2$

7. High temperature solid oxide ion conductor (stabilized zirconia), 650 – 950 °C, pressurized 0.5 – 5 MPa
Electrolyzer status

- Few types commercialized but - from an energy conversion and storage point of view - none of them are commercial in today's energy markets.

- The classical alkaline electrolyser was commercialized during the first half of the 20th century.

- If significant amounts of green fuel are to be produced via electrolysis in the near future (the next 1 - 4 years) then only alkaline electrolyser is available on some scale.
Electrolysis, principle of PEM

Anode: $2H_2O \rightarrow 4H^+ + O_2 + 4e^-$

Cathode: $4H^+ + 4e^- \rightarrow 2H_2$

Cell: $2H_2O \rightarrow 2H_2 + O_2$

Polymer electrolyte

Very expensive!

<table>
<thead>
<tr>
<th>Electrolyte</th>
<th>Oxygen electrode</th>
<th>H₂ electrode</th>
<th>Auxiliary mat.</th>
<th>Temp.</th>
</tr>
</thead>
<tbody>
<tr>
<td>LT-PEM Fluoropolymer+ SO₃H, Nafion®</td>
<td>IrO₂</td>
<td>Pt</td>
<td>Ti</td>
<td>80 °C</td>
</tr>
<tr>
<td>HT-PEM PFSA (Aquivion®) with H₃PO₄</td>
<td>IrO₂</td>
<td>Pt</td>
<td>Ta coated steel</td>
<td>150 °C</td>
</tr>
</tbody>
</table>
Oxygen evolution vs pH


The vertical line indicates a diffusion limiting current density.

b – values are Tafel slopes
Oxygen evolution vs pH

The NTNU data is interpreted as follows:

The diffusion limited species is OH$^{-}$

At low current density or high pH the OER is reduction of OH$^{-}$:

\[ 2 \text{OH}^{-} \rightarrow \frac{1}{2} \text{O}_2 + \text{H}_2\text{O} + 2 \text{e}^{-} \]

At high current density (above the OH$^{-}$ limiting diffusion rate) or low pH the OER is reduction of H$_2$O:

\[ \text{H}_2\text{O} \rightarrow \frac{1}{2} \text{O}_2 + 2 \text{H}^{+} + 2 \text{e}^{-} \]

It seems much easier to oxidize OH$^{-}$ to O$_2$ than H$_2$O

The correlation between OER overpotential and Mn$^{3+}$ concentration was probably incidental approximate parallel oxidation reactions
Principle of Alkaline Electrolysis cell (AEC)

Electricity + Heat  \rightarrow  Chemical energy

Anode: \[ 4 \text{OH}^- \rightarrow \text{O}_2 + 2 \text{H}_2\text{O} + 4 \text{e}^- \]
Cathode: \[ 2 \text{H}_2\text{O} + 4 \text{e}^- \rightarrow 2\text{H}_2 + 4 \text{OH}^- \]
Total: \[ 2 \text{H}_2\text{O} \rightarrow 2 \text{H}_2 + \text{O}_2 \]

High costs due to low current density
0.2 A cm\(^{-2}\) at 1.5 V – 0.5 A cm\(^{-2}\) at 1.8 V: cooling system necessary, low efficiency

<table>
<thead>
<tr>
<th>Electrolyte</th>
<th>Ox. electrode</th>
<th>H(_2) electrode</th>
<th>Temp.</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>KOH in water</td>
<td>Ni or Ni/Co-oxides</td>
<td>Ni on steel</td>
<td>80 °C</td>
<td>Low temp: LT-AEC</td>
</tr>
</tbody>
</table>

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## Cell Concepts for Alkaline Electrolysis

<table>
<thead>
<tr>
<th>Conventional cell design</th>
<th>Zero gap design</th>
<th>GDE with immobilized Electrolyte</th>
</tr>
</thead>
</table>

### Conventional cell design
- **Diaphragm**
- **Cathode**
- **Anode**

+ simple design  
- bubble effects  
- ohmic losses


### Zero gap design
- **Diaphragm**
- **Cathode**
- **Anode**

+ decreased ohmic losses  
+ proven technology  
- Assembly relatively difficult


### GDE with immobilized Electrolyte
- **Diaphragm**
- **Cathode**
- **Anode**

+ decreased ohmic losses  
+ minimized bubble effects  
+ Reversible operation  
- simple production method needed to be found
New 200 – 300 °C cell types

As part of the initiative called Catalysis for Sustainable Energy (CASE, www.case.dtu.dk) other types of electrolysis cells are being researched and developed at DTU Risø Campus.

• Solid Acids (CsH₂PO₄)

• Immobilized aqueous K₂CO₃

• Immobilized aqueous KOH

This temperature range seems to be catching interest now, e.g. Andrej Latric et al., Poster049, “Methanol Steam Reformer – High Temperature PEM Fuel Cell System Analysis” at FDFC, April 16-18, 2013, Karlsruhe. High Temperature was given as 250 °C.
New material structure


Scanning electron microscope image showing the open structure of the nickel foam.

Ni foam cost only ca 1/3 of the cost of Ni mesh per m²
New Alkaline Electrolyser

High temperature and pressure alkaline electrolysis

- Electrolyte:
  - aqueous KOH immobilized in a porous structure
- Gas diffusion electrodes:
  - porous Nickel, Raney-Nickel

F. Allebrod, C. Chatzichristodoulou, M. Mogensen, Patent application filed and in process
Cell concept and p


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Production of the cells: Activated electrodes

<table>
<thead>
<tr>
<th>Anode</th>
<th>Co activated</th>
<th>Co activated</th>
<th>Co activated</th>
</tr>
</thead>
<tbody>
<tr>
<td>40 µm</td>
<td>Ni STO</td>
<td>Ni</td>
<td>Co</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Cathode</th>
<th>Mo activated</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inc</td>
<td>Inc STO Mo</td>
</tr>
</tbody>
</table>

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Current Density at 1.5 V and 1.75 V as a Function of Temperature

![Graph showing current density as a function of applied cell voltage for different temperatures and pressures.]

Current density $[\text{A}\cdot\text{cm}^{-2}]$

<table>
<thead>
<tr>
<th>Temperature</th>
<th>1.5 V</th>
<th>1.75 V</th>
</tr>
</thead>
<tbody>
<tr>
<td>102 °C</td>
<td>0.09</td>
<td>0.20</td>
</tr>
<tr>
<td>208 °C</td>
<td>0.64</td>
<td>1.42</td>
</tr>
<tr>
<td>247 °C</td>
<td>0.90</td>
<td>2.00</td>
</tr>
</tbody>
</table>

**Cell composition**

- **Anode**: Ag deposition on Nickel foam
- **Cathode**: Inconel foam

**Cells with 45 wt% KOH (aq.) immobilized in mesoporous ceramic membrane (electrolyte) and with gas diffusion electrodes.**

Performance of Co- and Mo- Activated Cells at 40 bar

Current density [A•cm⁻²]

<table>
<thead>
<tr>
<th>Temperature</th>
<th>1.5 V</th>
<th>1.75 V</th>
</tr>
</thead>
<tbody>
<tr>
<td>200 °C</td>
<td>0.4</td>
<td>1.1</td>
</tr>
<tr>
<td>250 °C</td>
<td>1.1</td>
<td>2.2</td>
</tr>
</tbody>
</table>

Impedance [mΩ cm²] at 1.5 V pol.

<table>
<thead>
<tr>
<th>Temperature</th>
<th>Rₛ</th>
<th>Rₚ</th>
</tr>
</thead>
<tbody>
<tr>
<td>200 °C</td>
<td>186</td>
<td>214</td>
</tr>
<tr>
<td>250 °C</td>
<td>151</td>
<td>77</td>
</tr>
</tbody>
</table>

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Long term galvanostatic measurement

Test conditions

Temperature 250 °C
System pressure 40 bar
Polarization 0.64 A/cm²
pH₂O 15.0 bar
pH₂ 4.0 bar
P_w,KOH 16.5 bar

Cell composition

Anode
- Nickel foam
- Co-oxide catalyst
- Inconel foam
- Mo catalyst

Cathode
The Solid Oxide Electrolyser (SOEC)

- This cell type seems to have the greatest potential of the known electrolyser types, because:
  
  - The cell is fully reversible, i.e. the very same cell may be used as fuel cell as well as electrolyser cell
  
  - High power density
  
  - Contains no noble metals

SOC also has limitations
SOC type

Ni-YSZ supported

[Image of a material cross-section with labels: Ni/YSZ support, Ni/YSZ electrode, YSZ electrolyte, LSM-YSZ electrode, 10 µm, Acc. voltage: 12 kV, SE image, WD = 13 mm, Photo No. = 2165, Time: 14:58:29]
SOEC Cell performance

SOEC

SOFC

World record?

\[ i - V \text{ curves for a Ni-YSZ-supported Ni/YSZ/LSM SOC: electrolyzer (negative current density) and fuel cell (positive current density) at different temperatures and steam or CO}_2 \text{ partial pressures - balance is H}_2 \text{ or CO.} \text{ S.H. Jensen et al., International Journal of Hydrogen Energy, } 32 \text{ (2007) 3253} \]

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Thermodynamic stability of materials and gases

• Often degradation rates are reported as a function of current density even though it most often would be more appropriate to report as a function of electrode potentials and overpotentials.

• Naturally, the overvoltage and current density are directly related for a given of cell, but:

• Thermodynamic stability is directly related to electrode potential (vs. a reference, e.g. 1 atm. O\textsubscript{2} at given temperature) for given cell materials irrespective structural details such as particle size in composite electrodes.
Thermodynamic and mechanical stability of YSZ, Ni-YSZ, LSM-YSZ, CO etc.

- \(\Delta G_f \approx -975 \text{ kJ/mol } \text{ZrO}_2\) at 800 °C ~ a voltage stability range of ca. 2.5 V - the very maximum, but realities are worse

- 1 ppm Zr into Ni at ca. -1.5 V, 850 °C - impurities decrease stability of YSZ; M. Chen et al., J. Electrochem. Soc., 160 (2013) F883

- carbon fiber formation in CO\(_2\)-H\(_2\)O co-electrolysis if high diffusion limitation, Y. Tao et al., submitted to ECST

- O\(_2\) bubble formation with weakening of YSZ near LSM at high oxygen electrode overpotential (>~ 60 mV at 850 °C), R. Knibbe et al., J. Electrochem. Soc., 157 (2010) B1209

- Mechanical strength: limitation for ceramic supported cells!
Fast degradation > ca. 1 A cm\(^{-2}\)

- 850 °C, single cell, steam, -2 A cm\(^{-2}\) for 188 h
- Electrolyte conductivity degradation - near oxygen electrode
- TEM reveals that it is due to O\(_2\) bubble precipitation inside the electrolyte near the O\(_2\) LSM/YSZ-electrode destroying \(\sigma_{O^2}\)


This is to an extent a mechanical problem – O\(_2\) bubble formation is limited by YSZ creep.
Some results from single cell testing

- For higher current densities ($\geq -1 \text{ A/cm}^2$) both types of cell show fast degradation
- To improve the cells the degradation mechanisms must be studied
- This presentation focuses on degradation phenomena in the Ni-YSZ electrode

Degradation: Formation of ZrO_2 particles

TEM analyses of nano-particles

TEM analyses have confirmed that these nano-particles are ZrO_2 with varying crystal structure (monoclinic, tetragonal or cubic) and varying yttria content.

Degradation: Formation of ZrO$_2$ particles

Proposed mechanism for the formation of ZrO$_2$ nano-particles

Possibilities that we see

1. Our state-of-the-art cell can now run stable over more than 2000 h at 800 °C with 1 A cm\(^{-2}\) below thermoneutral voltage of 1.3 V!

2. We are in the process of improving the basis for safe operation though further detailed measurements and modeling

3. The improved understanding makes it possible accelerate the R&D further

4. We are looking into several aspects of mechanical properties and cell design

5. We anticipate significant improvements though further basic materials and electrochemical research
## Economy – “base case” assumptions for H\(_2\) production using SOEC


<table>
<thead>
<tr>
<th>Parameter</th>
<th>Assumption</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electricity</td>
<td>1.3US(\text{c}/\text{kWh})</td>
</tr>
<tr>
<td>Heat</td>
<td>0.3US(\text{c}/\text{kWh})</td>
</tr>
<tr>
<td>Investment</td>
<td>4000 $/m(^2) cell area</td>
</tr>
<tr>
<td>Demineralised Water</td>
<td>2.3 $/m(^3)</td>
</tr>
<tr>
<td>Cell temperature</td>
<td>850 °C</td>
</tr>
<tr>
<td>Heat reservoir temperature</td>
<td>110 °C</td>
</tr>
<tr>
<td>Pressure</td>
<td>1 atm</td>
</tr>
<tr>
<td>Cell voltage</td>
<td>1.29 V (thermo neutral potential)</td>
</tr>
<tr>
<td>Life time</td>
<td>10 years.</td>
</tr>
<tr>
<td>Operating activity</td>
<td>50%</td>
</tr>
<tr>
<td>Interest rate</td>
<td>5%</td>
</tr>
<tr>
<td>Energy loss in heat exchanger</td>
<td>5%</td>
</tr>
<tr>
<td>H(_2)O inlet concentration</td>
<td>95% (5% H(_2))</td>
</tr>
<tr>
<td>H(_2)O outlet concentration</td>
<td>5% (95% H(_2))</td>
</tr>
</tbody>
</table>
**H₂ production – economy estimation**

*Conversion of H₂ to equivalent crude oil price is on a pure energy content (J/kg) basis*
### Economy - “base case” assumptions for CO production by SOEC

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Electricity</td>
<td>1.3US¢/kWh</td>
</tr>
<tr>
<td>Heat</td>
<td>0.3US¢/kWh</td>
</tr>
<tr>
<td>Investment</td>
<td>4000 $/m² cell area</td>
</tr>
<tr>
<td>CO₂</td>
<td>2.3 $/ton</td>
</tr>
<tr>
<td>Cell temperature</td>
<td>850 °C</td>
</tr>
<tr>
<td>Heat reservoir temperature</td>
<td>110 °C</td>
</tr>
<tr>
<td>Pressure</td>
<td>1 atm</td>
</tr>
<tr>
<td>Cell voltage*</td>
<td>1.47 V (thermo neutral potential)</td>
</tr>
<tr>
<td>Life time</td>
<td>10 years.</td>
</tr>
<tr>
<td>Operating activity</td>
<td>50%</td>
</tr>
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<td>Interest rate</td>
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<td>Energy loss in heat exchanger</td>
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<tr>
<td>CO₂ inlet concentration</td>
<td>95% (5% CO)</td>
</tr>
<tr>
<td>CO₂ outlet concentration</td>
<td>5% (95% CO)</td>
</tr>
</tbody>
</table>
Conversion of CO to equivalent crude oil price is on a pure energy content (J/kg) basis.
Conclusions and claims

1. Cost (not efficiency on its own) is most important

2. The reversible SOC has the greatest potential as an energy converter for chemical storage of renewable energy

3. Still many possibilities for further significant improvement of the SOC

4. Put more emphasis on mechanical properties and materials compatibility in SOEC

5. Work more on high temperature alkaline cells

6. Work on other cell types to find even better cells
Acknowledgement

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• Danish Energy Authority
• Energinet.dk
• EU
• Topsoe Fuel Cell A/S
• Danish Programme Committee for Energy and Environment
• Danish Programme Committee for Nano Science and Technology, Biotechnology and IT
• The work of many colleagues over the years

Thank you for your attention!