Complementary analyses of aging in a commercial LiFePO₄/graphite 26650 cell

In this work we investigate the electrode degradation mechanisms in a commercial 2.5 Ah LiFePO₄/graphite 26650 cylindrical cell. Aged and fresh electrode samples were prepared by cycling two cells respectively five and 22 k times. Subsequently the cells were disassembled in a glovebox and the electrode samples were prepared for electrochemical testing in a 3-electrode setup, and for characterization with XRD, XPS and low-kV FIB/SEM tomography. A 1 μm thick CEI (cathode electrolyte interface) layer was observed at the electrode/electrolyte interface of the aged LiFePO₄ electrode. Relative to the fresh LiFePO₄ electrode, the aged electrode exhibited a larger series resistance which indicates the observed degradation layer increases the ionic resistance. In addition, micron-sized agglomerates, probably a mixture of carbonaceous material and decomposition products from the electrolyte, were observed at the electrode/electrolyte interface of the aged graphite electrode. These layers may contribute significantly to the loss of lithium inventory (LLI) in the cell, and to the loss of active material (LAM) in the graphite electrode. Low-voltage FIB/SEM tomography was used to detect local charging effects of graphite particles in the carbon electrode, an effect of poor dissipation of the electric charge to the ground after the sample interaction with the electron beam. The charging effects were primarily observed in the aged electrode and most of the locally charged particles were found to be close to the electrode/electrolyte interface, indicating a poorly percolating graphite network near this interface.
Scopus rating (2016): CiteScore 4.74 SJR 1.355 SNIP 1.177
Web of Science (2016): Impact factor 4.798
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 4.86 SJR 1.321 SNIP 1.324
Web of Science (2015): Impact factor 4.803
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): CiteScore 4.59 SJR 1.378 SNIP 1.456
Web of Science (2014): Impact factor 4.504
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 4.44 SJR 1.427 SNIP 1.587
Web of Science (2013): Impact factor 4.086
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 3.99 SJR 1.644 SNIP 1.574
Web of Science (2012): Impact factor 3.777
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 4.15 SJR 1.615 SNIP 1.788
Web of Science (2011): Impact factor 3.832
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 1.685 SNIP 1.715
Web of Science (2010): Impact factor 3.65
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 1.523 SNIP 1.615
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 1.524 SNIP 1.458
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Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 1.531 SNIP 1.726
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 1.484 SNIP 1.516
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 1.362 SNIP 1.567
Scopus rating (2003): SJR 1.637 SNIP 1.505
Web of Science (2003): Indexed yes
Scopus rating (2002): SJR 1.534 SNIP 1.441
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 1.312 SNIP 1.376
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 0.889 SNIP 1.161
Web of Science (2000): Indexed yes
Scopus rating (1999): SJR 0.936 SNIP 1.183

Original language: English
Keywords: Cathode electrolyte interface, Degradation mechanisms, Electrochemical impedance spectroscopy, Focused ion beam, Li-ion battery
A novel system for large-scale storage of electricity as synthetic natural gas using reversible pressurized solid oxide cells

The ongoing reduction of greenhouse gas emissions entails increased use of intermittent renewable energy technologies such as wind and solar. This raises the need for cost-effective and efficient electricity storage. In particular seasonal variations in supply and demand will require tremendous storage capacity. In this paper we present a truly large-scale electricity storage system which uses pressurized reversible solid oxide cells combined with catalytic reactors to store electricity as synthetic natural gas. By storing the produced gas in existing natural gas grids the system can create a strong and efficient link between the electricity and gas markets. In addition, the system is able to operate reversibly using gas from the grid to satisfy the electric power demand.

The system performance is analyzed with a component-based thermodynamic modeling tool which shows that electricity can be stored as synthetic natural gas with an energy efficiency of 89%. The gas to electricity efficiency is equally high, resulting in a round-trip storage efficiency of 80% (DC-to-DC).

General information
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Contributors: Butera, G., Jensen, S. H., Clausen, L. R.
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Publication date: 2018
Peer-reviewed: Yes

Publication information
Journal: Energy
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Ratings:
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 5.6 SJR 1.99 SNIP 1.923
Web of Science (2017): Impact factor 4.968
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 5.17 SJR 1.974 SNIP 1.823
Web of Science (2016): Impact factor 4.52
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BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 5.03 SJR 2.22 SNIP 2.037
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Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): CiteScore 5.02 SJR 2.458 SNIP 2.556
Web of Science (2013): Impact factor 4.159
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): CiteScore 4.25 SJR 1.935 SNIP 2.214
Web of Science (2012): Impact factor 3.651
Continuous Hydrothermal Flow Synthesis of Gd-doped CeO$_2$ (GDC) Nanoparticles for Inkjet Printing of SOFC Electrolytes

Gd$_{0.2}$Ce$_{0.8}$O$_{2-\delta}$ (GDC) nanoparticles were synthesized using continuous hydrothermal flow synthesis. By varying the synthesis conditions, particle size and morphology could be tailored. Here, particle sizes between 6 to 40 nm with polyhedral or octahedral shape could be obtained. Gd$_{0.2}$Ce$_{0.8}$O$_{2-\delta}$ nanoparticles were further processed into inks for inkjet printing. Despite the small particle size/large surface area, inks with excellent printing behavior were formulated. For proof-of-concept, thin GDC layers were printed on a) green NiO-GDC substrates, and on b) pre-sintered NiO-YSZ substrates. While no dense layers could be obtained on the green NiO-GDC substrates, GDC nanoparticles printed on NiO-YSZ substrates formed a dense continuous layer after firing at 1300 °C.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Ceramic Engineering & Science, Mixed Conductors, Applied Electrochemistry, Imperial College London
Pages: 315-327
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Peer-reviewed: Yes
Diffusion rates of reactants and components in solid oxide cells
The electrochemical performance of solid oxide electrolysis cells (SOECs) is very dependent on diffusion rates of the gaseous reactants through the porous electrodes, and the degradation rate of SOEC Ni/YSZ electrodes can be dependent on Ni-migration. The Ni-migration is dependent on electrode polarization and diffusion rate of Ni-containing minority species impact the migration. The electrode polarization as well as the diffusion rate of minority Ni-species are dependent on the partial pressure and pressure gradient of H2O as well as on electrochemical potential gradients. Thus, the H2O diffusion gradient resulting from the electrical load of the cell is expected to affect the Ni migration.

Therefore, this contribution first evaluates and discusses the diffusion rates of H2/H2O and CO/CO2 in porous Ni-YSZ composites. A 10 kh durability test with electrochemical impedance spectroscopy recorded at the beginning and end of the test show significant signs of change in the diffusion resistance.

Next, the diffusion rate of Ni-species is evaluated based on observed migration of Ni in the electrochemical active Ni-YSZ layer with sub-micron Ni particles. This is compared with catalysis research literature and models for Ni particle diffusivity [1].

Based on the presented results and literature study a modified hypothesis for Ni-migration is provided.
Noise Phenomena in Electrochemical Impedance Spectroscopy of Polymer Electrolyte Membrane Electrolysis Cells

In this study, the origin of noise in electrochemical impedance spectroscopy (EIS) spectra measured on a variety of polymer electrolyte membrane electrolysis cells (PEMECs) has been investigated. EIS was measured during operation at various current densities of seven different PEMECs divided in five different cell types including both acidic PEMECs and alkaline PEMECs. The noise pattern differed between various types of cells and between cells of the same cell type. Integration time had no influence on the EIS noise level, whereas the AC amplitude seems to influence the EIS noise level. Other electrical noise sources influencing the EIS measurements have been studied with oscilloscope. No noise was observed at DC. A hypothesis explaining the relation between bubble formation during electrolysis and EIS noise is proposed based on the experimental findings.
A Decade of Solid Oxide Electrolysis Improvements at DTU Energy

Solid oxide electrolysis cells (SOECs) can efficiently convert electrical energy (e.g. surplus wind power) to energy stored in fuels such as hydrogen or other synthetic fuels. Performance and durability of the SOEC has increased orders of magnitudes within the last decade. This paper presents a short review of the R&D work on SOEC single cells conducted at DTU Energy from 2005 to 2015. The SOEC improvements have involved increasing the of the oxygen electrode performance, elimination of impurities in the feed streams, optimization of processing routes, and fuel electrode structure optimization. All together, these improvements have led to a decrease in long-term degradation rate from ~40 %/kh to ~0.4 %/kh for steam electrolysis at -1 A/cm², while the initial area specific resistance has been decreased from 0.44 Ωcm² to 0.15 Ωcm² at -0.5 A/cm² and 750 °C.

General information
A Physically-Based Equivalent Circuit Model for the Impedance of a LiFePO$_4$/Graphite 26650 Cylindrical Cell

In this work an Equivalent Circuit Model (ECM) is developed and used to model impedance spectra measured on a commercial 26650 LiFePO$_4$/Graphite cylindrical cell. The ECM is based on measurements and modeling of impedance spectra recorded separately on cathode (LiFePO$_4$) and anode (Graphite) samples, harvested from the commercial cell.
Modeling of the single-electrode impedance spectra provided information about the electronic and ionic resistance in the porous composite electrodes, as well as the solid state diffusion. Focused Ion Beam (FIB)/Scanning Electron Microscopy (SEM) of anode and cathode samples was used to make 3-D maps of the electrode microstructures and to obtain microstructural data for the ECM. The complementary analysis was crucial for the resolution of the single electrode impedance parameters and the proposal and validation of a new equivalent circuit used to model the full commercial battery impedance.
Aqueous metal–organic solutions for YSZ thin film inkjet deposition

Inkjet printing of 8% Y2O3-stabilized ZrO2 (YSZ) thin films is achieved by designing a novel water-based reactive ink for Drop-on-Demand (DoD) inkjet printing. The ink formulation is based on a novel chemical strategy that consists of a combination of metal oxide precursors (zirconium alkoxide and yttrium salt), water and a nucleophilic agent, i.e. n-methyldiethanolamine (MDEA). This chemistry leads to metal–organic complexes with long term ink stability and high precision printability. Ink rheology and chemical reactivity are analyzed and controlled in terms of metal–organic interactions in the solutions. Thin dense nanocrystalline YSZ films below 150 nm are obtained by low temperature calcination treatments (400–500 °C), making the deposition suitable for a large variety of substrates, including silicon, glass and metals. Thin films and printed patterns achieve full densification with no lateral shrinkage and high ionic conductivity.
Concentration Impedance in Testing of Solid Oxide Cells Revisited

The concentration impedance originating from diffusion and reactant conversion impedance of the Ni-YSZ supported fuel electrode in solid oxide cell has been treated many times during the latest couple of decades. In spite of this, the separation of the diffusion impedance from the conversion impedance is still not trivial. Therefore, combined theoretical and experimental methods available for breakdown of the concentration impedance are outlined and discussed.
Correction: Large-scale electricity storage utilizing reversible solid oxide cells combined with underground storage of CO₂ and CH₄

Correction for ‘Large-scale electricity storage utilizing reversible solid oxide cells combined with underground storage of CO₂ and CH₄’ by S. H. Jensen et al., Energy Environ. Sci., 2015, 8, 2471–2479.
Investigation of a Spinel-forming Cu-Mn Foam as an Oxygen Electrode Contact Material in a Solid Oxide Cell Single Repeating Unit

A critical issue in state-of-the-art solid oxide cell stacks is the contacting of the oxygen electrode. The commonly used ceramic contact layers are applied in a green state and cannot be sintered properly, due to compliance limitations arising from other stack components like sealing glasses and steels. The consequence is a low layer and interface strength. A metallic copper manganese foam, which is oxidized under operation conditions into a conductive Cu$_{1+x}$Mn$_{2-x}$O$_4$ spinel, is presented in this work as a viable contact solution. The foam has been electrochemically tested in a single repeating unit setup for 350 hours of constant operation, followed by dynamic conditions with thermal cycles. After operation, a micro structural analysis using scanning electron microscopy, energy dispersive X-ray spectroscopy and X-ray diffraction was carried out. It was confirmed that after oxidation/operation the manganese solely formed a CuMn-spinel phase, mixed with a CuO phase. A separate Mn-oxide phase was not found. The conductivity and contacting of the foam was sufficient for > 350 h of SOFC operation. With an initial serial resistance comparable to single cell tests using gold foil as contact material and moderate degradation rates, the CuMn foam presented itself as an interesting cathode contact solution.

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State: Published
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Journal: Fuel Cells
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ISSN (Print): 1615-6846
Ratings:
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 1.88 SJR 0.559 SNIP 0.748
Web of Science (2017): Impact factor 2.149
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 1.79 SJR 0.495 SNIP 0.603
Web of Science (2016): Impact factor 1.706
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 2.02 SJR 0.685 SNIP 0.779
Web of Science (2015): Impact factor 1.769
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 2.05 SJR 0.615 SNIP 0.792
Web of Science (2014): Impact factor 2.08
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 1.99 SJR 0.835 SNIP 0.833
Web of Science (2013): Impact factor 1.546
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 2.76 SJR 1.24 SNIP 0.993
Web of Science (2012): Impact factor 2.364
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 3.31 SJR 1.639 SNIP 1.247
Life cycle assessment of hydrogen production from water electrolysis

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Applied Electrochemistry, Mixed Conductors, Proton conductors
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Publication date: 2017

Host publication information
Title of host publication: Book of Abstracts, Sustain 2017
Publisher: Technical University of Denmark (DTU)
Article number: M-12
Electronic versions:
SustainAbstracts2017c.compressed_123.pdf
Research output: Research - peer-review » Conference abstract in proceedings – Annual report year: 2017

Pressurized reversible operation of a 30-cell solid oxide cell stack using carbonaceous gases
Recent theoretical studies show that reversible electrochemical conversion of H2O and CO2 to CH4 inside pressurized solid oxide cells (SOCs) combined with subsurface storage of the produced gases can facilitate seasonal electricity storage with a round-trip efficiency reaching 70-80% and a storage cost below 3 ¢/kWh. Here we show test results with a 30-cell SOFCMAN 301 stack operated with carbonaceous gases at 18.7 bar and 700 °C in both electrolysis and fuel cell mode. The CH4 content in the stack outlet gas increased from 0.22% at open circuit voltage (OCV) to 18% at -0.17 A cm-2 in electrolysis mode. The degradation rates in both fuel cell and electrolysis mode were comparable to previously reported SOFCMAN stack degradation rates measured at ambient pressure operation with H2/H2O gas mixtures.

General information
State: Published
A Decade of Improvements for Solid Oxide Electrolysis Cells. Long-Term Degradation Rate from 40%/Kh to 0.4 % Kh

Solid oxide electrolysis cells (SOEC) have the potential for efficient large-scale conversion from electrical energy to chemical energy stored in fuels, such as hydrogen or synthetic hydrocarbon fuels by use of well-known catalysis processes. Key issues for the break-through of this technology are to provide inexpensive, reliable, high performing and long-term stable SOEC for stack and system applications. At DTU Energy (formerly Department of Fuel Cells and Solid State Chemistry, Risø National Laboratory), research within SOEC for more than a decade has led to long-term degradation rates on cell level being improved from 40 %/kh to 0.4 %/kh for tests at -1 A/cm² (figure 1). In this paper, we review the key findings and highlight different performance and durability limiting factors that have been discovered, analyzed and addressed over the years to reach the tremendous increase in long-term stability for SOEC as illustrated by the cell tests in figure 1.

A TEM study of morphological and structural degradation phenomena in LiFePO₄-CB cathodes: Morphological and structural degradation in LiFePO₄-CB cathodes

LiFePO₄-based cathodes suffer from various degradation mechanisms, which influences the battery performance. In this paper, morphological and structural degradation phenomena in laboratory cathodes made of LiFePO₄ mixed with carbon black (CB) in a 1 mol/L LiPF₆ in EC:DMC (1:1 by weight) electrolyte are investigated by transmission electron microscopy at various preparation, assembling, storage, and cycling stages. High-resolution transmission electron microscopy imaging shows that continuous SEI layers are formed on the LiFePO₄ particles and that both storage and cycling affect the formation. Additionally, loss of CB crystallinity, CB aggregation, and agglomeration is observed. Charge–discharge curves and impedance spectra measured during cycling confirm that these degradation mechanisms reduce the cathode conductivity and capacity.
Publication information
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Volume: 40
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Ratings:
| BFI (2018) | BFI-level 1 |
| Web of Science (2018): Indexed yes |
| BFI (2017) | BFI-level 1 |
| Scopus rating (2017): CiteScore 2.72 SJR 0.764 SNIP 0.941 |
| Web of Science (2017): Impact factor 3.009 |
| Web of Science (2017): Indexed yes |
| BFI (2016) | BFI-level 1 |
| Scopus rating (2016): CiteScore 2.44 SJR 0.744 SNIP 0.891 |
| Web of Science (2016): Impact factor 2.598 |
| Web of Science (2016): Indexed yes |
| BFI (2015) | BFI-level 1 |
| Scopus rating (2015): CiteScore 2.52 SJR 0.889 SNIP 1.06 |
| Web of Science (2015): Impact factor 2.529 |
| BFI (2014) | BFI-level 1 |
| Scopus rating (2014): CiteScore 2.56 SJR 1.034 SNIP 1.338 |
| Web of Science (2014): Impact factor 2.418 |
| BFI (2013) | BFI-level 1 |
| Scopus rating (2013): CiteScore 2.71 SJR 1.043 SNIP 1.641 |
| Web of Science (2013): Impact factor 2.737 |
| ISI indexed (2013): ISI indexed yes |
| BFI (2012) | BFI-level 1 |
| Scopus rating (2012): CiteScore 2.2 SJR 1.076 SNIP 1.412 |
| Web of Science (2012): Impact factor 1.987 |
| ISI indexed (2012): ISI indexed yes |
| Web of Science (2012): Indexed yes |
| BFI (2011) | BFI-level 1 |
| Scopus rating (2011): CiteScore 2.24 SJR 1.012 SNIP 1.349 |
| Web of Science (2011): Impact factor 2.122 |
| ISI indexed (2011): ISI indexed yes |
| BFI (2010) | BFI-level 1 |
| Scopus rating (2010): SJR 1.114 SNIP 1.325 |
| Web of Science (2010): Impact factor 1.86 |
| BFI (2009) | BFI-level 1 |
| Scopus rating (2009): SJR 1.027 SNIP 1.208 |
| BFI (2008) | BFI-level 1 |
| Scopus rating (2008): SJR 0.589 SNIP 0.778 |
| Web of Science (2008): Indexed yes |
| Scopus rating (2007): SJR 0.584 SNIP 1.012 |
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| Scopus rating (2005): SJR 1.043 SNIP 0.84 |
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| Scopus rating (2004): SJR 0.872 SNIP 0.939 |
| Scopus rating (2003): SJR 0.582 SNIP 0.616 |
Characterisation of a Planar Solid Oxide Cell Stack Operated at Elevated Pressure

As the global and local energy production becomes more dependent on intermittent renewable sources like wind and solar, efficient and reversible conversion of electricity to storable fuels becomes increasingly important. Solid oxide cells (SOCs) are interesting in this context since they can be operated either as electrolyzers (SOEC) to convert electricity to fuels such as hydrogen or methane, and as fuel cells (SOFC) to convert fuels to electricity. Both productivity and conversion efficiency can be improved if the SOC operation pressure can be increased from ambient pressure to 10–30 bar.

Here we characterize an SOC stack operated at pressures from ambient pressure to 10 bar without fluctuations in the steam supply. The pressure dependency of stack temperature, cell area specific resistance (ASR), current-voltage (iV) curves, stack impedance spectra and pressure drop across the stack and heat exchangers is analyzed in this paper. Additionally, the expected impact on the hydrogen production efficiency and cost is discussed.
Electron microscopy investigations of changes in morphology and conductivity of LiFePO4/C electrodes

In this work we study the structural degradation of a laboratory Li-ion battery LiFePO4/Carbon Black (LFP/CB) cathode by various electron microscopy techniques including low kV Focused Ion Beam (FIB)/Scanning Electron Microscopy (SEM) 3D tomography. Several changes are observed in FIB/SEM images of fresh and degraded cathodes, including cracks in the LFP particles, secondary disconnected particles, and agglomeration of CB. Low voltage (1 kV) SEM images show that the CB agglomerates have a different brightness than the fresh CB, due to charging effects. This suggests that the electronic conductivity of the CB agglomerates is low compared to that of the fresh CB particles. HRTEM analysis shows that fresh CB particles are quasi crystalline, whereas the LFP/CB interface in the degraded electrode shows amorphous carbon surrounding the LFP particles. The presence of the amorphous carbon is known to impede the electronic conductivity and thereby decreasing percolation in the cathode and reducing the electrode capacity.

General information
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Organisations: Department of Energy Conversion and Storage, Applied Electrochemistry, Imaging and Structural Analysis, Atomic scale modelling and materials, Northwestern University
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Publication date: 2016
Peer-reviewed: Yes

Publication information
Journal: Journal of Power Sources
Volume: 307
ISSN (Print): 0378-7753
Ratings:
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Performance and Lifetime Limiting Effects in Li-Ion Batteries

Lithium-ion batteries (LIBs) find widespread use for electricity storage, from portable devices such as smart phones to electric vehicles (EV), because of their high energy density and design flexibility. However, limited lifetime is still a challenge for several LIB materials. Specifically, the detailed coupling between degradation mechanisms and battery usage is not fully understood, which impede lifetime improvements. To understand the degradation mechanisms and increase the performance of these materials, the development of improved characterization methods is crucial. This PhD thesis focuses on the thorough analysis of degradation mechanism in LIBs, trying to relate morphological and structural changes in Lithium-ion battery electrodes to performance degradation observed during electrode cycling. Degradation mechanisms in laboratory scale LFP cathodes were correlated with the degradation mechanisms observed in commercial LIBs. The structural and morphological changes in cycled laboratory LFP cathodes were studied by low-kV FIB/SEM Tomography and TEM analysis and related to the electrode performance using Electrochemical Impedance Spectroscopy (EIS). The two main degradation processes observed by microscopy analysis in the aged electrode were cracking of LFP particles and agglomeration of carbon black (CB) additive. The increased heterogeneity of the CB network reduces the electron percolation throughout the porous electrode, thereby decreasing the amount of electrochemically active LFP particles. The electron resistivity was quantified with the EIS analysis using a Transmission Line Model (TLM) developed for porous LFP electrodes. Similar TLM models were applied for the analysis of the polarization processes in a commercial LFP and graphite electrodes. The microscopy analysis of the electrodes showed the presence of carbonaceous agglomerates on the electrode/electrolyte interfaces. The agglomerates are expected to increase the ionic resistance and be related to loss of lithium inventory (LLI).

Pressurized Operation of a Planar Solid Oxide Cell Stack

Solid oxide cells (SOCs) can be operated either as fuel cells (SOFC) to convert fuels to electricity or as electrolyzers (SOEC) to convert electricity to fuels such as hydrogen or methane. Pressurized operation of SOCs provide several benefits on both cell and system level. If successfully matured, pressurized SOEC based electrolyzers can become more efficient both energy- and cost-wise than PEM and Alkaline systems. Pressurization of SOFCs can significantly increase the cell power density and reduce the size of auxiliary components. In the present study, a SOC stack was successfully operated at pressures up to 25 bar. The pressure dependency of the measured current-voltage (I-V) curves and impedance spectra on the SOC stack are analyzed and the relation between various system parameters and pressure is derived. With increasing pressure the open circuit voltage (OCV) and the reaction kinetics (electrode performance) increases for thermodynamic and kinetic reasons, respectively. Further, the summit frequency of the gas concentration impedance arc and the pressure difference across the stack and heat exchangers is seen to decrease with increasing pressure following a power-law expression. Finally a durability test was conducted at 10 bar.
Reversible Operation of Solid Oxide Cells for Sustainable Fuel Production and Solar/Wind Load-Balancing

The solid oxide electrochemical cell (SOC) is a promising candidate for large-scale energy storage. In electrolysis mode it stores renewable electricity as chemical energy in the form of fuels like hydrogen and hydrocarbons, and the same cell can be operated in the reversedirection to produce electricity from fuels – either previously stored fuels or from an external supply e.g. natural gas or biogas. This reversibility combined with fuel-flexibility is unique among energy storage technologies like closed-system batteries and single-direction electrolyzers. However, few studies have been conducted with focus on fundamentals or applications of bi-directional operation. This presentation will highlight our recent developments in applying reversible SOCs (RSOCs) for renewable energy storage with respect to cell and stack testing, cell and system design, and techno-economic analysis. At the cell level, long-term testing has shown that improved stability can be achieved by reversible operation compared with steady-state electrolysis operation. Further, we have developed novel Ni-free fuel-electrodes that both outperform conventional Ni-based electrodes and do not catalyze carbon deposition, which opens the door to advanced applications of RSOCs that utilize carbonaceous fuels. At the stack level, we have demonstrated operation that follows real-world time-series electricity supply and demand data, considering a 100% renewable energy scenario where wind power is the only power supply. When the wind power supply exceeds demand, the RSOC stack produces syngas via co-electrolysis of CO2 and H2O. Part of the syngas is converted to methanol downstream in the system to meet the demand of transportation vehicles, and the rest is stored for electrical load balancing by conversion back to electricity in fuel cell mode when electricity demand exceeds the wind power supply. At the system level, techno-economic analyses and system designs for different scales and applications have been realized. A simulation of an RSOC system that uses real-world time-series market prices for electricity and natural gas in Denmark to decide when to operate in electrolysis mode (buying electricity and selling methane) or fuel-cell mode (buying gas and selling electricity) shows the advantage of a reversible system and the changing operating profile as the fraction of wind power supply grows. Finally, we discuss the potential for systems with novel chemistries and components to compete with state-of-the-art rechargeable batteries with respect to cost and round-trip efficiency.

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Elektrolyse gør al energi fra vindmøller værdifuld

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Alkali Metal-O2 Batteries. Performance and Lifetime Limiting Effects
The rechargeable Na-O2 and Li-O2 batteries are attractive battery technologies as they potentially are very cheap and as they theoretically possess about 3 and 10 times higher energy density than the current Li-ion technologies. This PhD thesis is dedicated to studying the effects that limit cell performance of these two technologies.

The Li-O2 battery was first introduced in 1996 and focus in the field is still on understanding the fundamental mechanisms controlling discharge and charge. This PhD thesis was mainly dedicated to the Li-O2 battery and initially charge conduction through the discharge product, Li2O2, was investigated. This was done by using of a conventional three electrode cell in which the heterogeneous electron transfer rate of three different redox couples were studied on Li2O2 coated glassy carbon electrodes to provide a measure of the conductivity of the Li2O2 layers. Charge transport through Li2O2 gives further evidence that hole transport dominates charge-transfer through Li2O2. Electrochemical impedance spectroscopy was also used to conduct detailed investigations of surface capacitance, ion transport, and chargetransfer reactions in the cathode of the Li-O2 cell. The capacitance of the cathode was shown to be sensitive to the thickness of the deposited Li2O2 layer. These investigations also explored the influence of the composition of the electrolyte and conditions, which favors a solution mediated Li2O2 deposition mechanism. On charge, an electrochemical “safe” operating voltage was identified until 3.30 V were an interface layer was formed, which activates side reactions and further increases the cell potential. A number of ionic liquids were also investigated for their oxygen diffusivity and solubility and while these were in the order of currently employed aprotic electrolytes as the ionic liquids significantly decompose under electrochemical operation. Last, the influence of CO2 was investigated and it was suggested that CO2 blocks the step valleys of the deposited Li2O2 forcing Li2O2 growth away from the electrode surface hereby increasing cell capacity, as the discharge becomes less limited by the cathode surface area.

The Na-O2 battery is an even newer technology as it was first introduced to the scientific community in 2010. The two batteries are experimentally quite similar as the only difference is the choice of anode. However, when one studies the two systems, the mechanisms controlling each type of battery are quite different. The discharge and charge processes of the non-aqueous Na-O2 battery were studied in this thesis. On discharge, the deposition mechanism of NaO2 was shown to be highly dependent on the current density and cell limitations could be correlated to the depositions mechanisms. On charge, three regions of NaO2 oxidation were identified, each corresponding to a different type of NaO2 oxidation.
Degradation Studies on LiFePO$_4$ cathode

In this paper we examine a laboratory LiFePO$_4$ (LFP) cathode and propose a simple model that predicts the electrode capacity as function of C-rate, number of cycles and calendar time. Microcracks were found in Li$_{1-x}$FePO$_4$ particles in a degraded LFP electrode and low-acceleration voltage (1 kV) FIB/SEM analysis allowed us to obtain phase contrast between FePO$_4$ and LiFePO$_4$. The evolution of micro-cracks is expected to increase the concentration of LFP particles which are not electronically accessible and thus cause a loss in capacity.

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Eliminating degradation in solid oxide electrochemical cells by reversible operation

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Fabrication of thin yttria-stabilized-zirconia dense electrolyte layers by inkjet printing for high performing solid oxide fuel cells

In this work, we present how a low-cost HP Deskjet 1000 inkjet printer was used to fabricate a 1.2 mm thin, dense and gas tight 16 cm² solid oxide fuel cells (SOFC) electrolyte. The electrolyte was printed using an ink made of highly diluted (<4 vol.%) nanometric yttria stabilized zirconia (YSZ) powders (50 nm in size) in an aqueous medium. The ink was designed to be a highly dispersed, long term stable colloidal suspension, with optimal printability characteristics. The electrolyte was made by a multiple printing procedure, which ensures coverage of the several flaws occurring in a single printing pass. Together with an optimized sintering procedure this resulted in good adhesion and densification of the electrolyte. The SOFC exhibited a close-to-theoretical open circuit voltage and a remarkable peak power density above 1.5 W cm⁻² at 800 °C.
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| Web of Science (2015): Indexed yes |
| BFI (2014): BFI-level 1 |
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In Situ Studies of Fe$^{4+}$ Stability in β-Li$_3$Fe$_2$(PO$_4$)$_3$ Cathodes for Li Ion Batteries

In commercial Fe-based batteries the Fe$^{2+}$/Fe$^{3+}$ oxidation states are used, however by also utilizing the Fe$^{4+}$ oxidation state, intercalation of up to two Li ions per Fe ion could be possible. In this study, we investigate whether Fe$^{4+}$ can be formed and stabilized in β-Li$_3$Fe$_2$(PO$_4$)$_3$. The work includes in situ synchrotron X-ray powder diffraction studies (XRPD) during charging of β-Li$_3$Fe$_2$(PO$_4$)$_3$ up to 5.0 V vs. Li/Li$.+$ A novel capillary-based micro battery cell for in situ XRPD has been designed for this. During charge, a plateau at 4.5 V was found and a small contraction in volume was observed, indicating some Li ion extraction. The volume change of the rhombohedral unit cell is anisotropic, with a decrease in the a parameter and an increase in the c parameter during the Li ion extraction. Unfortunately, no increased discharge capacity was observed and Mössbauer spectroscopy showed no evidence of Fe$^{4+}$ formation. Oxidation of the organic electrolyte is inevitable at 4.5 V but this alone cannot explain the volume change. Instead, a reversible oxygen redox process (O$^{2−}→$O−) could possibly explain and charge compensate for the reversible extraction of lithium ions from β-Li$_3$Fe$_2$(PO$_4$)$_3$. 

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Integration of CO₂ air capture and solid oxide electrolysis for methane production

This work studied the production of substitute natural gas (SNG) from CO₂ captured from the atmosphere followed by co-electrolysis with H₂O in solid oxide electrolyzer cells (SOEC) and downstream catalytic methane production. Over the coming 20 years, Denmark is on a track to remove fossil fuels from all sectors of the energy system except for transportation. In the recently published Energikoncept 2035 [1], the Danish grid operator, Energinet.dk lays out a scenario based on 72% wind power and 21% biomass and waste in the electricity grid mix. In this scenario, biogas and electrolysis gasses are projected to be used for production of process heat, peak-load power generation and on the longer term to replace hydrocarbons in the most energy intensive parts of the transportation sector; especially aviation. As a prerequisite for the scenario, no biomass can be imported to enhance the supply of combustible resources. In such an energy system, technologies for production of CO₂ neutral hydrocarbons for easy storage and use in the existing infrastructure; especially in the natural gas grid; may be of great value. The studied technology fulfills those demands. The main goal of the work was to design a plant and develop a thermodynamic model of the plant operation, enabling analyses related to selection of operating parameters; analysis and optimization of internal heat recovery and integration between the main technological subsystems. Finally to identify the main areas of technological development through economic analyses. The work included experimental work on an example of a system for capture of CO₂; the humidity swing (HS) system, qualitatively evaluating the H₂O uptake and CO₂ desorption characteristics of the sorbent material, especially in relation to the supply of H₂O to the sorbent. It was found that H₂O supplied in the gas phase resulted in slow uptakes and desorption rates of CO₂, whereas supplying liquid water to the sorbent resulted in fast desorption in the first hours, after which the rate dropped sharply. A method was developed and used to characterize the impurities present in CO₂ stream from the HS system in addition to the temperature vacuum swing (TVS) system under development by Climeworks Ltd. The method relied on adsorption of impurities on a filter consisting of nickel-yttria-stabilised-zirconia (Ni/YSZ), similar to the material used in the fuel electrodes of SOECs followed by elemental analysis by glow discharge mass spectrometry. The method had a sub-ppm detection limit. Across the tested systems, a range of elements known to be detrimental to solid oxide cell (SOC) operation were detected in the range from tens of ppb to 20 ppm. The SNG plant was modelled using the process integration software package PRO/II alongside the design process, and a series of minor studies using PRO/II and thermodynamic analysis software FactSage® aided the design process. This included studying a long range of questions such as alternative strategies for CO₂ compression; the structure of the methanation plant; and the risk of carbon formation in both SOEC and methanation reactors, etc. The model was based on a thermodynamic 0-dimensional model of the electrolyzer sub-system, developed to technological specifications from the thermodynamic SOEC model published by Sun et al. [2] This model was used for a study of operating parameters, and two design cases were identified for the full plant based on these results. The two cases both operated at 80 atm, and had SOEC operating temperatures of 850 °C and 600 °C. The area specific resistance (ASR) of the SOECs were extrapolated to high pressure and low temperatures based on data for standard DTU Energy Ni-YSZ based cells, and the pressure dependency of the individual cell processes. With the full plant model finished, the potential for internal recovery of surplus heat was analyzed, and a network of heat exchangers synthesized in order to minimize the requirements for external heating and cooling services. Based on the process flow sheets and the heat exchanger network, the dimensions and costs of the equipment of the plant were calculated and additional cost components such as installation of equipment, land use, labor costs, operation and maintenance, etc. were estimated according to standard methods. The plant had a yearly production capacity of 575,000 Nm³ of SNG with a methane content above 98.5% which resulted in a Wobbe index of 49 MJ/Nm³ which is sufficient for injection into the natural gas grid. The SOEC stack power was around 700 kW, and the plant operated a ten energy efficiency of 65% (HHV) and 58% (LHV). An economic analysis based on guidelines from the Danish energy agency and standard methods was conducted accounting for interest rates, taxes, depreciation etc. at a minimum acceptable rate of return set to the minimum of 4%. The economic analysis resulted in SNG production prices of 1.88 €/Nm³ and 2.94 €/Nm³ based on an electricity price of 18.6 €/MJ, a price of process heat at 120 °C of 11.9 €/MJ and a price of cell area of 0.23 €/cm². The main cost drivers were identified as the capital costs of the SOEC and air capture systems and the heat exchanger documenting the modelling and design process. For operating costs, the electricity price had a significant impact, whereas the dependency of the SNG price on the heat price was minor. The technical issues where discussed in separate chapters interspersed by chapters documenting the modelling and design process. Finally, a comprehensive discussion at the end treats the technical issues of the plant in the light of the economic analysis.
Large-scale electricity storage utilizing reversible solid oxide cells combined with underground storage of CO₂ and CH₄

Electricity storage is needed on an unprecedented scale to sustain the ongoing transition of electricity generation from fossil fuels to intermittent renewable energy sources like wind and solar power. Today pumped hydro is the only commercially viable large-scale electricity storage technology, but unfortunately it is limited to mountainous regions and therefore difficult to expand. Emerging technologies like adiabatic compressed air energy storage (ACAES) or storage using conventional power-to-gas (P2G) technology combined with underground gas storage can be more widely deployed, but unfortunately for long-term to seasonal periods these technologies are either very expensive or provide a very low round-trip efficiency. Here we describe a novel storage method combining recent advances in reversible solid oxide electrochemical cells with sub-surface storage of CO₂ and CH₄, thereby enabling large-scale electricity storage with a round-trip efficiency exceeding 70% and an estimated storage cost around 3 b/kW h⁻¹, i.e., comparable to pumped hydro and much better than previously proposed technologies.

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Low - voltage FIB/SEM Tomography for 3D Microstructure Evolution of LiFePO4/C Electrode

Li-ion batteries find widespread use in many electricity storage applications, from portable devices to electric vehicles [1-3], and LiFePO4 (LFP) is one of the most common cathodes because of its long durability and high safety [4, 5] but, since its low ionic and electronic conduction, it is always mixed with carbon black (CB) additives to increase electronic percolation in the electrode. Focused Ion Beam (FIB)/Scanning Electron Microscopy (SEM) Tomography is one of the most used techniques for the study of the three-dimensional microstructure of porous electrodes [6-8]. Imaging at low-kV has been shown to be an excellent technique for studying electron percolation in Ni-network in solid oxide fuel cells [9]. In this work we study the degradation process that occurs in a LFP/C electrode by Low-kV FIB/SEM Tomography, using the low-voltage percolation technique to identify compositional changes in the CB network in three-dimensions. FIB/SEM images of a fresh and degraded cathode are compared and LFP grains are seen to crack with cycling, resulting in the formation of secondary disconnected particles with increased ionic resistivity; CB particles are instead observed to agglomerate, reducing the electrochemically active surface area. Using low voltage imaging (1 kV) a significant fraction of the large carbon agglomerates found in the aged electrode show a higher secondary electron yield compared to the fresh CB particles at low accelerating voltage. This suggests that degradation occurs both due to morphological changes and due to amorphous-crystalline phase transitions in the carbon network, resulting in non-percolating CB agglomerates.

Figure 1 shows lateral Everhart – Thornley (E-T) and in-lens detector images of fresh (a, b) and degraded cathode (c, d) collected after FIB slicing. White grains are LFP, black particles are CB additive and grey regions are pores infiltrated with silicon resin to improve phase contrast [7]. The CB network appears entirely dark in the in-lens detector image of the fresh electrode (Fig. 1b). It is possible to notice some charging effects from the insulating silicon resin. The in-lens detector image of the degraded electrode (Fig. 1d) is instead characterized by the presence of big carbon agglomerates (red rings) which are brighter because they charge as the electron beam hit them. This indicates a lower electric conductivity. 3D reconstruction of the entire network (Fig. 2) revealed that the carbon phase with lower electric conductivity accounted for approximately 25% of the volume of the total carbon in the sample. This of course affects electrode capacity since a reduced electron percolation in the CB network impedes (de)lithiation process of LFP particles. References - M. Armand, J. M. Tarascon, Nature, 451, 652-657 (2008). - B. Scrosati, J. Hassoun, and Y.K. Sun, Energy Environ. Sci., 4, 3287-3295 (2011). - J.M. Tarascon, M. Armand, Nature, 414, 359-367 (2001). - A.K. Padhi, K.S. Nanjundaswamy, and J. B. Goodenough, J. Electrochem. Soc., 144(4), 1188-1194 (1997). - Y. Wang, P. He, and H. Zhou, Energy Environ. Sci., 4, 805-817 (2010). - T. Hutzenlaub et al., Electrochemical and Solid-State Letters, 15 (3), A33-A36 (2012). - M. Ender et al, J. Electrochem. Soc., 159(7), A972-A980 (2012). - Z. Liu et al., J. Power Sources, 227, 267-274 (2013). - K. Thydén, Y.L. Liu, and J.B. Bilde-Sørensen, Solid State Ionics, 178, 1984-1989 (2008). [Figure]

General information

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Low-voltage FIB/SEM Tomography for 3D Microstructure Evolution of LiFePO4/C Electrode

This work presents an investigation of the degradation mechanisms that occur in LiFePO4/C battery electrodes during charge/discharge cycling. Impedance spectra were measured on a fresh electrode and an electrode aged by cycling. The spectra were modeled with an equivalent circuit which indicates that both the ionic and electronic pathways in the electrode were negatively affected by the cycling. Focused Ion Beam/Scanning Electron Microscopy (FIB/SEM) tomography of both electrodes shows that cycling causes agglomerations of Carbon black (CB). In addition to this, Low-voltage FIB/SEM revealed non-conductive CB in the aged electrode.

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Performance Characterization of Solid Oxide Cells Under High Pressure

In this work, recent pressurized test results of a planar Ni-YSZ (YSZ: Yttria stabilized Zirconia) supported solid oxide cell are presented. Measurements were performed at 800 C in both fuel cell and electrolysis mode at different pressures. A comparison of the electrochemical performance of the cell at 1 and 3 bar shows a significant and equal performance gain at higher pressure in both fuel cell mode and electrolysis mode. Electrochemical impedance spectroscopy revealed that the serial resistance was not affected by the operation pressure; all the other processes that are dependent on partial pressures (oxygen, steam and hydrogen) were affected by increasing the pressure. In electrolysis mode at low current density, the performance improvement was counteracted by the increase in open circuit voltage, but it has to be borne in mind that the pressurized gas contains higher molar free energy.

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Redox Probing Study of the Potential Dependence of Charge Transport Through Li₂O₂

In the field of energy storage devices the pursuit for cheap, high energy density, reliable secondary batteries is at the top of the agenda. The Li−O₂ battery is one of the possible technologies that, in theory, should be able to close the gap, which exists between the present state-of-the-art Liion technologies and the demand placed on batteries by technologies such as electrical vehicles. Here we present a redox probing study of the charge transfer across the main deposition product lithium peroxide, Li₂O₂, in the Li−O₂ battery using outer-sphere redox shuttles. The change in heterogeneous electron transfer exchange rate as a function of the potential and the Li₂O₂ layer thickness (∼depth-of-discharge) was determined using electrochemical impedance spectroscopy. The attenuation of the electron transfer exchange rate with film thickness is dependent on the probing potential, providing evidence that hole transport is the dominant process for charge transfer through Li₂O₂ and showing that the origin of the sudden death observed upon discharge is due to charge transport limitations.
Degradation Studies on LiFePO₄ Cathode

Lithium-ion batteries are a promising technology for automotive application, but limited performance and lifetime is still a big issue. The aim of this work is to study and address degradation processes which affect LiFePO₄ (LFP) cathodes - one of the most common cathodes in commercial Li-ion batteries. In order to evaluate how the LFP cathode is affected by C-rate a LFP working electrode, Lithium metal foil counter electrode and Lithium metal reference electrode was tested in a 3-electrode setup with a standard 1M LiPF6 in 1:1 EC/DMC electrolyte and glass fiber separator. The working electrode/counter electrode was subjected to several charge/discharge cycles between 3.0 V and 4.0 V at different discharge rates. Figure 1 shows the voltage profile of the LFP electrode (solid line) and full battery (dotted line) during charge/discharge process. It is seen that the higher the C-rate, the higher is the polarization furnished by the counter electrode which reduces the capacity. In Figure 2, the discharge capacity [mAh/g] is plotted vs the number of charge/discharge cycles. Series of 10 cycles at a given C-rate was applied to the battery. Each series was followed by a C/10 cycle (green points). A linear fit has been applied to the first series (omitting first two cycles where instability of the system is observed), in order to calculate the degradation rates. High C-rates are seen to affect the discharge capacity, but the capacity is almost completely recovered (green points) and only a limited degradation occurs. Impedance spectroscopy has been also applied to investigate the LFP cathode degradation. Figure 3 shows the imaginary part of the impedance measured at 50% State-of-Charge after each series of cycles. The relative increase in the impedance arc around 1 KHz (assumed to be associated with charge transfer resistance at the LFP particle surfaces) is seen to gradually decrease with increasing number of series. This indicates that more cycles per series is needed to establish a convincing relation between C-rate and degradation. The degradation studies will be coupled with FIB/SEM analysis in order to observe changes in the pore structure or micro cracks that would affect electronic percolation. Figure 4 displays an example of a fresh LFP cathode after FIB cutting. White particles are LFP grains while the black area contains carbon particles and pores, which are difficult to distinguish from each other. Substitution of the epoxy resin with a silicon resin increases the contrast between pores and carbon particles [1] and this will be used in the forthcoming FIB/SEM analysis.

Performance characterization of solid oxide cells under high pressure

Solid oxide electrolysis cells (SOECs) offer a great potential for large scale conversion of renewable electrical energy into chemical energy via electrolysis of H2O and CO2 to produce syngas (H2 + CO). The produced syngas can be further catalytically converted into various gaseous or liquid hydrocarbon fuels, which is normally performed at high pressure to achieve a high yield. Operation of SOECs at elevated pressure will therefore facilitate integration with the downstream fuel synthesis and is furthermore advantageous as it increases the cell performance. In this work, recent pressurised test results of a planar Ni-YSZ (YSZ: Yttria stabilized Zirconia) supported solid oxide cell are presented. The test was performed at 800 °C at pressures up to 15 bar. A comparison of the electrochemical performance of the cell at 1 and 3 bar shows a significant and equal performance gain at higher pressure in both fuel cell mode and electrolysis mode. In electrolysis mode at low current density, the performance improvement was counteracted by the increase in open circuit voltage, but it has to be born in mind that the pressurised gas contains higher molar free energy. Operating at high current density is in particular more beneficial when operating the SOEC at increased pressure.

Storing electricity and CO2 as synthetic hydrocarbon fuels by high temperature electrolysis

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Durability of Solid Oxide Electrolysis Cells for Syngas Production
Performance and durability of Ni-YSZ supported solid oxide electrolysis cells (SOECs) for co-electrolysis of H₂O and CO₂ at high current density was investigated. The cells consist of a Ni-YSZ support, a Ni-YSZ electrode, a YSZ electrolyte, and an LSM-YSZ electrode. The durability was examined at 800°C and electrolysis current densities of −1 or −1.5 A/cm² with 60% reactant (H₂O + CO₂) utilization. The cell voltage degradation showed a strong dependence on the electrolysis current density. Electrochemical characterization of the cells showed that the degradation was mainly related to the LSM-YSZ electrode when operated at −1 A/cm², whereas at increased current density (−1.5 A/cm²), both the Ni-YSZ and the LSM-YSZ electrode degraded. The initial degradation (0 - 200 h) is mostly due to increasing of polarization resistance, while the long term degradation is mainly caused by increasing of serial resistance.

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In Situ Study of High Voltage Performance of Li$_3$Fe$_2$(PO$_4$)$_3$ Cathodes for Li Ion batteries

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Modeling degradation in SOEC impedance spectra
Solid oxide cell (SOC) performance is limited by various processes. One way to investigate these processes is by electrochemical impedance spectroscopy. In order to quantify and characterize the processes, an equivalent circuit can be used to model the SOC impedance spectra (IS). Unfortunately, the optimal equivalent circuit is often unknown and to complicate matters further, several processes contribute to the SOC impedance - making detailed process characterization difficult. In this work we analyze and model a series of IS measured during steam electrolysis operation of an SOC. During testing, degradation is only observed in the Ni/YSZ electrode and not in the electrolyte or the LSM/YSZ electrode. A batch fit of the differences between the IS shows that a modified Gerischer element provides a better fit to the NiYSZ electrode impedance than the frequently used RQ element - albeit neither equivalent circuit provides a perfect fit. However, modeling with the Gerischer element indicates that the Ni/YSZ electrode performance decrease, relates to an electrochemical reaction resistance increase at the electrode triple phase boundaries. © 2013 The Electrochemical Society.

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Operation and Degradation Aspects of EV Batteries

In this chapter we analyse the relation between EV battery operation and degradation. This is done through measurements and modelling of the performance, degradation effects and thermal effects of two types of lithium batteries. The batteries investigated, lithium iron phosphate and lithium–nickel–manganese–cobalt, represent two promising chemistries for electric vehicle operation. The methods used to obtain an electric equivalent-circuit model for the batteries are explained and the effects of battery degradation on the model parameters are evaluated. The analysis leads to results regarding the influence of battery operation on expected lifetime and in particular the influence that vehicle-to-grid operation might have.
Measurements of Electric Performance and Impedance of a 75 Ah NMC Lithium Battery Module

Detailed characterization of battery modules is necessary to construct reliable models that incorporate performance related aspects of the modules such as thermodynamics, electrochemical reaction kinetics and degradation mechanisms. Charge-discharge curves, temperature and battery impedance measurements can provide information about these aspects. Charge-discharge curves can be used to measure the battery open circuit voltage and the internal resistance. Temperature measurements provide information about the thermodynamic reactions and impedance spectra yield detailed information about the reaction kinetics. In this paper we present the measurement methods used to examine the internal resistance, the capacity and the impedance of a 75 Ah NMC battery module.

In order to measure the impedance of the battery module and of the individual cells in the module, we combine the single sine technique and the Laplace transformed excitation signal technique which each have pros and cons. By combining the two impedance measurement techniques we are able to reduce the measurement time by a factor of 20 relative to ordinary single-sine measurements.

Further we use the impedance measurements to calculate the overvoltage as a function of state of charge and the difference between charging overvoltage and discharging overvoltage and compare it with measurements.

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Performance and Durability of Solid Oxide Electrolysis Cells for Syngas Production

Performance and durability of Ni/YSZ based solid oxide electrolysis cells (SOECs) for co-electrolysis of H2O and CO2 at high current density were investigated. The cells consist of a Ni/YSZ support, a Ni/YSZ fuel electrode, a YSZ electrolyte, and a LSM-YSZ oxygen electrode. The cell durability was examined at 800°C and electrolysis current density of -1 or -1.5 A/cm² with 60% reactant (H2O+CO2) utilization. The cell voltage degradation showed a strong dependence on the electrolysis current density, with an overall cell voltage degradation rate of 0.24 mV/h at -1 A/cm² and of 0.82 mV/h at -1.5 A/cm². Electrochemical characterization of the cells showed that the degradation was mainly related to the LSM/YSZ electrode when operated at -1 A/cm², whereas at increased current density (-1.5 A/cm²), both the Ni/YSZ and LSM/YSZ electrodes showed degradation.

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Publication date: 2012

Production of "Green Natural Gas" Using Solid Oxide Electrolysis Cells (SOEC): Status of Technology and Costs
This paper gives arguments in favour of using green natural gas (GNG) as storage media for the intermittent renewable energy sources. GNG is here defined as being CH4, i.e. methane, often called synthetic natural gas or substitute natural gas (SNG), produced using renewable or at least CO2 neutral energy sources only. Also dimethyl ether (DME = (CH3)2O), which might be called Liquefied Green Gas, LGG, in analogy to Liquefied Petroleum Gas, LPG, because DME has properties similar to LPG. It further gives a short review of the state of the art of electrolysis in general and SOEC in particular. Production of synthesis gas (H2 + CO) from CO2 and H2O using SOEC technology is evaluated. GNG and LGG can be produced from synthesis gas (or short: syngas) by means of well established commercially available catalysis technology. Finally, estimations of costs and efficiencies are presented and the relative importance of cost and efficiency is briefly discussed.

General information
State: Published
Pages: 2314-2320
Publication date: 2012

Thermodynamic analysis of synthetic hydrocarbon fuel production in pressurized solid oxide electrolysis cells
A promising way to store wind and solar electricity is by electrolysis of H2O and CO2 using solid oxide electrolysis cells (SOECs) to produce synthetic hydrocarbon fuels that can be used in existing fuel infrastructure. Pressurized operation decreases the cell internal resistance and enables improved system efficiency, potentially lowering the fuel production cost significantly. In this paper, we present a thermodynamic analysis of synthetic methane and dimethyl ether (DME) production using pressurized SOECs, in order to determine feasible operating conditions for producing the desired hydrocarbon fuel and avoiding damage to the cells. The main parameters of cell operating temperature, pressure, inlet gas composition and reactant utilization are varied to examine how they influence cell thermoneutral and reversible potentials, in situ formation of methane and carbon at the Ni–YSZ electrode, and outlet gas composition. For methane production, low temperature and high pressure operation could improve the system efficiency, but might lead to a higher capital cost. For DME production, high pressure SOEC operation necessitates higher operating temperature in order to avoid carbon formation at higher reactant utilization. Optimal operating conditions are dependent on the total system design.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Applied Electrochemistry, Mixed Conductors, Fundamental Electrochemistry
Pages: 17101-17110
Publication date: 2012
Peer-reviewed: Yes
Performance and durability of solid oxide electrolysis cells for syngas production

General information
State: Published
Organisations: Electrochemical Evaluation, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electroceramics, Microstructures and Interfaces, Electrochemistry
Publication date: 2011

Host publication information
Title of host publication: Proceedings
Electronic versions:
Performance and durability.pdf
Source: orbit
Source-ID: 316447
Research output: Research - peer-review › Conference abstract in proceedings – Annual report year: 2011

Synthetic fuel production using pressurized solid oxide electrolysis cells

General information
State: Published
Organisations: Risø National Laboratory for Sustainable Energy, Fuel Cells and Solid State Chemistry Division, Electrochemistry
Contributors: Jensen, S. H.
Publication date: 2011
Peer-reviewed: No
Event: Paper presented at HyFC Academy Workshop, Roskilde, Denmark.
Source: orbit
Source-ID: 316012
Research output: Research › Paper – Annual report year: 2011

Hydrogen and synthetic fuel production using pressurized solid oxide electrolysis cells

Wind and solar power is troubled by large fluctuations in delivery due to changing weather. The surplus electricity can be used in a Solid Oxide Electrolyzer Cell (SOEC) to split CO2 + H2O into CO + H2 (+O2). The synthesis gas (CO + H2) can subsequently be catalyzed into various types of synthetic fuels using a suitable catalyst. As the catalyst operates at elevated pressure the fuel production system can be simplified by operating the SOEC at elevated pressure. Here we present the results of a cell test with pressures ranging from 0.4 bar to 10 bar. The cell was tested both as an SOEC and as a Solid Oxide Fuel Cell (SOFC). In agreement with previous reports, the SOFC performance increases with pressure. The SOEC performance, at 750 °C, was found to be weakly affected by the pressure range in this study, however the internal resistance decreased significantly with increasing pressure.

General information
State: Published
Contributors: Jensen, S. H., Sun, X., Ebbesen, S., Knibbe, R., Mogensen, M. B.
Poisoning of Solid Oxide Electrolysis Cells by Impurities

Electrolysis of H2O, CO2, and co-electrolysis of H2O and CO2 was studied in Ni/yttria-stabilized zirconia (YSZ) electrode supported solid oxide electrolysis cells (SOECs) consisting of a Ni/YSZ support, a Ni/YSZ electrode layer, a YSZ electrolyte, and an lanthanum strontium manganite (LSM)/YSZ oxygen electrode. When applying the gases as received, the cells degraded significantly at the Ni/YSZ electrode, whereas only minor (and initial) degradation was observed for either the Ni/YSZ or LSM/YSZ electrode. Application of clean gases to the Ni/YSZ electrode resulted in operation without any long-term degradation, in fact some cells activated slightly. This shows that the durability of these SOECs is heavily influenced by impurities in the inlet gases. Cleaning the inlet gases to the Ni/YSZ electrode may be a solution for operating these Ni/YSZ-based SOECs without long-term degradation.

General information
State: Published
Pages: B1419-B1429
Publication date: 2010
Peer-reviewed: Yes

Publication information
Journal: Journal of The Electrochemical Society
Volume: 157
Issue number: 10
ISSN (Print): 0013-4651
Ratings:
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 3.48 SJR 1.267 SNIP 1.009
Web of Science (2017): Impact factor 3.662
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 2.97 SJR 1.222 SNIP 0.963
Web of Science (2016): Impact factor 3.259
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 3.17 SJR 1.115 SNIP 1.066
Web of Science (2015): Impact factor 3.014
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 3.36 SJR 1.213 SNIP 1.25
Prospects for large scale electricity storage in Denmark

In a future power systems with additional wind power capacity there will be an increased need for large scale power management as well as reliable balancing and reserve capabilities. Different technologies for large scale electricity storage provide solutions to the different challenges arising with high wind power penetration. This paper presents a review of the electricity storage technologies relevant for large power systems. The paper also presents an estimation of the economic feasibility of electricity storage using the west Danish power market area as a case.

General information
State: Published
Organisations: Composites and Materials Mechanics, Materials Research Division, Risø National Laboratory for Sustainable Energy, Electrochemistry, Fuel Cells and Solid State Chemistry Division
Contributors: Krog Ekman, C., Jensen, S. H.
Pages: 1140-1147
Publication date: 2010
Peer-reviewed: Yes

Publication information
Journal: Energy Conversion and Management
Volume: 51
Issue number: 6
ISSN (Print): 0196-8904
Ratings:
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 6.85 SJR 2.537 SNIP 2.233
Web of Science (2017): Impact factor 6.377
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 6.04 SJR 2.232 SNIP 2.109
Web of Science (2016): Impact factor 5.589
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 5.24 SJR 2.023 SNIP 2.079
Web of Science (2015): Impact factor 4.801
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 5.35 SJR 1.789 SNIP 2.791
Web of Science (2014): Impact factor 4.38
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 4.49 SJR 1.613 SNIP 2.534
Web of Science (2013): Impact factor 3.59
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 3.72 SJR 1.674 SNIP 2.242
Web of Science (2012): Impact factor 2.775
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 3.03 SJR 1.24 SNIP 1.82
Web of Science (2011): Impact factor 2.216
Studies on solid oxide electrolysis cells - aspect of synthetic hydrocarbon fuel production

General information
State: Published
Organisations: Electrochemical Evaluation, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy; Electrochemistry
Contributors: Sun, X., Jensen, S. H., Ebbesen, S., Mogensen, M. B.
Publication date: 2010

Host publication information
Title of host publication: Conference proceedings
Publisher: AIChE, American Institute of Chemical Engineers
Keywords: Electrolysis
Source: orbit
Source-ID: 275064
Research output: Research → Article in proceedings – Annual report year: 2011

Advanced Test Method of Solid Oxide Cells in a Plug-Flow Setup

This paper describes a case study of two electrolysis tests of solid oxide cells [Ni/yttria-stabilized zirconia (YSZ)-YSZ-lanthanum strontium manganite (LSM)/YSZ] tested in a plug-flow setup. An extensively instrumented cell test setup was used, and the tests involved measurements of the cell impedance at open-circuit voltage and under current load, the cell voltage, and the in-plane voltage in the electrodes. From the cell-voltage measurements it was evident that a significant passivation of the cells occurred over the first similar to 10 days. Thereafter, the cells reactivated at constant electrolysis conditions. From measurements of the in-plane voltages in the electrodes and impedance spectra obtained during the electrolysis operation, we derive information about the resistance distributions in the Ni electrodes and describe how these distributions evolve over time. Impedance spectra at open-circuit voltage before and after electrolysis testing at various gas compositions were used to show that the Ni electrode was affected by the electrolysis operation, whereas the LSM electrode was not.
Kan vi få CO₂-neutral benzin?
General information
State: Published
Contributors: Ebbesen, S., Jensen, S. H., Smith, A., Mogensen, M. B.
Pages: 28-30
Publication date: 2009
Peer-reviewed: Unknown

Publication information
Journal: Dansk Kemi
Volume: 90
Issue number: 12
ISSN (Print): 0011-6335
Ratings:
ISI indexed (2013): ISI indexed no
ISI indexed (2012): ISI indexed no
ISI indexed (2011): ISI indexed no
Web of Science (2007): Indexed yes
Web of Science (2004): Indexed yes
Original language: Danish
Keywords: Fuel Cells and hydrogen
Source: orbit
Source-ID: 253841
Research output: Communication › Journal article – Annual report year: 2009

SOEC potential economics, peak shaving

General information
State: Published
Organisations: Rise National Laboratory for Sustainable Energy, Fuel Cells and Solid State Chemistry Division, Electrochemistry
Contributors: Jensen, S. H.
Publication date: 2009
Peer-reviewed: No
Event: Abstract from HyFC Academy Workshop, Rise, Denmark.
Keywords: Fuel Cells and hydrogen, Electrolysis
Source: orbit
Source-ID: 255369
Research output: Research › Conference abstract for conference – Annual report year: 2009

Electrolysis of steam and CO2

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Contributors: Mogensen, M. B., Jensen, S. H., Ebbesen, S., Hauch, A.
Publication date: 2008
Peer-reviewed: No
Event: Paper presented at Electroceramics XI, Manchester, United Kingdom.
Source: orbit
Source-ID: 232186
Research output: Research › Paper – Annual report year: 2008

Gas Concentration Impedance in an SOFC Plug Flow Setup

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Department of Chemistry
Contributors: Jensen, S. H., Mogensen, M. B., Jacobsen, T.
Pages: Abstract 431
Separation of process contributions in impedance spectra by variation of test conditions

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Physical Chemistry, Department of Chemistry
Contributors: Jensen, S. H., Jacobsen, T., Mogensen, M. B.
Publication date: 2008

Host publication information
Title of host publication: Meeting Abstracts - Electrochemical Society
Publisher: The Electrochemical Society
Electronic versions: jensen.pdf

Bibliographical note
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Source: orbit
Source-ID: 264284
Research output: Research - peer-review › Conference abstract in proceedings – Annual report year: 2008

SOFC Ni-electrode Resistance Distribution Investigated by Gas Concentration Impedance in a Plug-Flow Setup

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electroceramics, Physical Chemistry, Department of Chemistry
Contributors: Jensen, S. H., Hendriksen, P. V., Mogensen, M. B., Jacobsen, T.
Pages: 307-315
Publication date: 2008
Peer-reviewed: Yes

Publication information
Journal: ECS Transactions
Volume: 13
Issue number: 26
ISSN (Print): 1938-5862
Ratings:
BFI (2018): BFI-level 1
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 0.44 SJR 0.225 SNIP 0.252
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 0.4 SJR 0.228 SNIP 0.253
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 0.36 SJR 0.211 SNIP 0.244
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 0.36 SJR 0.212 SNIP 0.234
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 0.27 SJR 0.192 SNIP 0.231
Solid Oxide Electrolysis Cells: Microstructure and Degradation of the Ni/Yttria-Stabilized Zirconia Electrode

Solid oxide fuel cells produced at Risø DTU have been tested as solid oxide electrolysis cells for steam electrolysis by applying an external voltage. Varying the sealing on the hydrogen electrode side of the setup verifies that the previously reported passivation over the first few hundred hours of electrolysis testing was an effect of the applied glass sealing. Degradation of the cells during long-term galvanostatic electrolysis testing [850°C, -1/2 A/cm2, p(H2O)/p(H2)=0.5/0.5] was analyzed by impedance spectroscopy and the degradation was found mainly to be caused by increasing polarization resistance associated with the hydrogen electrode. A cell voltage degradation of 2%/1000 h was obtained. Postmortem analysis of cells tested at these conditions showed that the electrode microstructure could withstand at least 1300 h of electrolysis testing, however, impurities were found in the hydrogen electrode of tested solid oxide electrolysis cells. Electrolysis testing at high current density, high temperature, and a high partial pressure of steam [-2 A/cm2, 950°C, p(H2O)=0.9 atm] was observed to lead to significant microstructural changes at the hydrogen electrode-electrolyte interface.

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Contributors: Hauch, A., Ebbesen, S., Jensen, S. H., Mogensen, M. B.
Pages: B1184-B1193
Publication date: 2008
Peer-reviewed: Yes

Publication information
Journal: Journal of the Electrochemical Society
Volume: 155
Issue number: 11
ISSN (Print): 0013-4651
Ratings:
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 3.48 SJR 1.267 SNIP 1.009
Web of Science (2017): Impact factor 3.662
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 2.97 SJR 1.222 SNIP 0.963
Web of Science (2016): Impact factor 3.259
Solid Oxide Electrolyser Cell

Solid oxide fuel cells (SOFCs) produced at Risø National Laboratory were tested as steam electrolyser under various current densities, operating temperatures and steam partial pressures. At 950 °C and a cell voltage of 1.48 V the current density was -3.6 A/cm² with approx. 30% H₂ + 70% H₂O in the inlet gas and a H₂O utilization of approx. 40%. The tested SOECs were also used for CO₂ electrolysis. Economy studies of CO and H₂ production show that especially H₂ production can be competitive in areas with cheap electricity. Assuming the above described initial performance and a lifetime of 10 years it is possible to achieve a production price of 0.7 US$/kg H₂ with an electricity price of 1.3 US¢/kWh.

The cell voltage was measured as function of time. In test of about two month of duration a long-term degradation was observed. At 850 °C, -0.5 A/cm² with 50 vol% H₂ the degradation rate was approx. 20 mV/1000h. It was shown that the degradation happens at Ni/YSZ-electrode. The long term degradation is probably caused by coarsening of the Ni-particles. After onset of electrolysis operation a transient passivation/reactivation phenomena with duration of several days was observed. It was shown that the phenomenon is attributed to the SiO₂ contamination at the Ni/YSZ-electrode-electrolyte interface. The SiO₂ arises from the albite glass sealing (NaAlSi₃O₈) that surrounds the electrode. Si may enter the Ni/YSZ electrode via the reaction Si(OH)₄(g) « SiO₂(l) +2H₂O(g). At the active sites of the Ni/YSZ electrode steam is reduced via the reaction H₂O + 2e⁻ ® H₂ + O₂-. This shifts the equilibrium of the first reaction to form SiO₂(l) at the active sites. After a certain time the sealing crystallizes and the SiO₂(l) evaporates from the active sites and the cell reactivates. The passivation is shown to relate to a build up of a diffusion-type impedance arc that converge towards (jω)⁻½ for the frequency ω converging towards infinity.

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Contributors: Jensen, S. H.
Number of pages: 135
Publication date: Feb 2007

Publication information
Place of publication: Kgs. Lyngby, Denmark
Publisher: Technical University of Denmark (DTU)
Original language: English
(Risø-PhD; No. 29(EN)).
Keywords: Risø-PhD-29(EN), Risø-PhD-29, Risø-PhD-0029
Electronic versions:
Søren.pdf
Source: orbit
Source-ID: 267863
Research output: Research › Ph.D. thesis – Annual report year: 2007

A method to separate process contributions in impedance spectra by variation of test conditions

Many processes contribute to the overall impedance of an electrochemical cell, and these may be difficult to separate in the impedance spectrum. Here, we present an investigation of a solid oxide fuel cell based on differences in impedance spectra due to a change of operating parameters and present the result as the derivative of the impedance with respect to ln(f). The method is used to separate the anode and cathode contributions and to identify various types of processes.

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electroceramics, Physical Chemistry, Department of Chemistry
Contributors: Jensen, S. H., Hauch, A., Hendriksen, P. V., Mogensen, M. B., Bonanos, N., Jacobsen, T.
Scopus rating (2006): SJR 1.608 SNIP 1.535
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 1.523 SNIP 1.481
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 1.712 SNIP 1.7
Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 1.971 SNIP 1.677
Web of Science (2003): Indexed yes
Scopus rating (2002): SJR 2.036 SNIP 1.618
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 1.663 SNIP 1.729
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 1.632 SNIP 1.7
Web of Science (2000): Indexed yes
Scopus rating (1999): SJR 1.6 SNIP 1.846

Original language: English

Electronic versions:
Søren.pdf
DOIs:
10.1149/1.2790791
URLs:
http://dx.doi.org/10.1149/1.2790791

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Source: orbit
Source-ID: 210249
Research output: Research - peer-review › Journal article – Annual report year: 2007

Durability of solid oxide electrolysis cells for hydrogen production

General information
State: Published
Organisations: Risø National Laboratory for Sustainable Energy, Fuel Cells and Solid State Chemistry Division, Electrochemistry
Contributors: Hauch, A., Jensen, S. H., Ebbesen, S., Mogensen, M. B.
Pages: 327-338
Publication date: 2007

Host publication information
Title of host publication: Energy solutions for sustainable development. Proceedings
Publisher: Risø National Laboratory
Editors: Sønderberg Petersen, L., Larsen, H.
ISBN (Print): 978-87-550-3603-1
(Denmark. Forskningscenter Risoe. Risoe-R; No. 1608(EN)).
Keywords: Risø-R-1608, Risø-R-1608(EN)
Electronic versions:
ris_r_1608.pdf
Source: orbit
Source-ID: 216410
Research output: Research - peer-review › Article in proceedings – Annual report year: 2007

EM investigations of impurities in a fuel cell H2 electrode

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electroceramics
Hydrogen and synthetic fuel production from renewable energy sources

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Ceramic processing
Contributors: Jensen, S. H., Larsen, P. H., Mogensen, M. B.
Pages: 3253-3257
Publication date: 2007
Peer-reviewed: Yes

Publication information
Volume: 32
ISSN (Print): 0360-3199
Ratings:
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 4.1 SJR 1.116 SNIP 1.267
Web of Science (2017): Impact factor 4.229
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 3.74 SJR 1.145 SNIP 1.315
Web of Science (2016): Impact factor 3.582
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 3.46 SJR 1.27 SNIP 1.314
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): CiteScore 3.54 SJR 1.207 SNIP 1.484
Web of Science (2014): Impact factor 3.313
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): CiteScore 3.38 SJR 1.265 SNIP 1.449
Web of Science (2013): Impact factor 2.93
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): CiteScore 3.96 SJR 1.499 SNIP 1.708
Web of Science (2012): Impact factor 3.548
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): CiteScore 4.42 SJR 1.443 SNIP 1.828
New method in resolving process, responses in impedance spectra

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Contributors: Jensen, S.
Publication date: 2007
Peer-reviewed: No
Source: orbit
Source-ID: 215891
Research output: Research › Paper – Annual report year: 2007

New method in resolving process responses in impedance spectra. Postdoc presentation

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Contributors: Jensen, S.
Publication date: 2007
Peer-reviewed: No
Event: Paper presented at SERC biannual meeting, Risø, Denmark.
Source: orbit
Source-ID: 216096
Passivation and activation of SOFC nanostructured cathodes

**General information**
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electroceramics
Contributors: Wang, W., Bentzen, J. J., Jensen, S. H., Bonanos, N., Hendriksen, P. V., Mogensen, M. B.
Pages: 1243-1250
Publication date: 2007

**Host publication information**
Title of host publication: Proceedings
Volume: Pt. 1
Place of publication: Pennington, NJ
Publisher: The Electrochemical Society
Editors: Eguchi, K., Singhal, S., Yokokawa, H., Mizusaki, J.
ISBN (Print): 978-1-56677-554-0
(ÉCS Transactions; No. 1, Vol. 2007-07).
DOIs: 10.1149/1.2729225
Source: orbit
Source-ID: 215685
Research output: Research - peer-review › Article in proceedings – Annual report year: 2007

Reversible solid oxide cells

**General information**
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Contributors: Mogensen, M. B., Jensen, S., Hauch, A., Chorkendorff, I., Jacobsen, T.
Publication date: 2007
Peer-reviewed: No
Event: Paper presented at 32. International Cocoa Beach conference and exposition on advanced ceramics and composites, Cocoa Beach, FL (US), .
URLs:
Source: orbit
Source-ID: 216358
Research output: Research › Paper – Annual report year: 2007

Reversible Solid Oxide Cells: Performance of Reversible Solid Oxide Cells: 301-MogensenPerformance of Reversible Solid Oxide Cells: A Review

**General information**
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Experimental Surface and Nanomaterials Physics, Department of Physics, Department of Chemistry
Pages: 91-101
Publication date: 2007

**Host publication information**
Title of host publication: Ceramic Engineering and Science Proceedings : Advances in Solid Oxide Fuel Cells III
Volume: Volume 28
Publisher: American Ceramic Society
Editor: Bansal, N.
Source: orbit
Source-ID: 194479
Research output: Research - peer-review › Article in proceedings – Annual report year: 2007
Silica segregation in the Ni/YSZ electrode

Solid oxide fuel cells were tested as solid oxide electrolysis cells used for high-temperature steam electrolysis. The cells were tested at a variety of operation temperatures, current densities, and gas flows to the electrodes. The cell voltages monitored during the electrolysis operation increased significantly during the first few days of testing. Impedance spectroscopy obtained during electrolysis shows that it is the Ni/yttria-stabilized zirconia (YSZ) electrode that passivates. Reference cells and tested cells were examined in a scanning electron microscope after testing. These postmortem analyses reveal the reason for the observed passivation, because results from energy-dispersive spectroscopy clearly show evidence that silica-containing impurities have segregated to the hydrogen electrode/electrolyte interface during electrolysis testing. Examples of different microstructures and amounts of Si-containing impurities in the electrolyte/hydrogen electrode interface are presented and related to the electrolysis test conditions and the passivation histories of the electrolysis cells. (C) 2007 The Electrochemical Society.

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Department of Chemistry, Risø National Laboratory for Sustainable Energy, Nano-Microstructures in Materials, Materials Research Division
Contributors: Hauch, A., Jensen, S. H., Bilde-Sørensen, J., Mogensen, M. B.
Pages: A619-A626
Publication date: 2007
Peer-reviewed: Yes

Publication information
Journal: Journal of The Electrochemical Society
Volume: 154
Issue number: 7
ISSN (Print): 0013-4651
Ratings:
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 3.48 SJR 1.267 SNIP 1.009
Web of Science (2017): Impact factor 3.662
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 2.97 SJR 1.222 SNIP 0.963
Web of Science (2016): Impact factor 3.259
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 3.17 SJR 1.115 SNIP 1.066
Web of Science (2015): Impact factor 3.014
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 3.36 SJR 1.213 SNIP 1.25
Web of Science (2014): Impact factor 3.266
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 2.92 SJR 1.169 SNIP 1.309
Web of Science (2013): Impact factor 2.859
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 2.61 SJR 1.329 SNIP 1.281
Web of Science (2012): Impact factor 2.588
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 2.74 SJR 1.331 SNIP 1.335
Solid oxide electrolysis for fuel production

General information
State: Published
Organisations: Risø National Laboratory for Sustainable Energy, Fuel Cells and Solid State Chemistry Division, Electrochemistry
Contributors: Ebbesen, S., Hauch, A., Jensen, S., Mogensen, M. B.
Publication date: 2007
Peer-reviewed: No

Bibliographical note
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Source: orbit
Source-ID: 216424
Research output: Research - peer-review › Journal article – Annual report year: 2007

Electronic versions:
Jensen.pdf

DOIs:
10.1149/1.2733861

URLs:
http://dx.doi.org/10.1149/1.2733861
Performance and durability of solid oxide electrolysis cells

Solid oxide fuel cells produced at Riso National Laboratory have been tested as electrolysis cells by applying an external voltage. Results on initial performance and durability of such reversible solid oxide cells at temperatures from 750 to 950 degrees C and current densities from -0.25 A/cm(2) to -0.50 A/cm(2) are reported. The full cells have an initial area specific resistance as low as 0.27 Omega cm(2) for electrolysis operation at 850 degrees C. During galvanostatic long-term electrolysis tests, the cells were observed to passivate mainly during the first similar to 100 h of electrolysis. Cells that have been passivated during electrolysis tests can be partly activated again by operation in fuel cell mode or even at constant electrolysis conditions after several hundred hours of testing.

General information
State: Published
Organisations: Department of Chemistry, Ceramic processing, Fuel Cells and Solid State Chemistry Division, Riso National Laboratory for Sustainable Energy, Electrochemistry
Contributors: Hauch, A., Jensen, S. H., Ramousse, S., Mogensen, M. B.
Pages: A1741-A1747
Publication date: 2006
Peer-reviewed: Yes
Performance and stability of barium strontium cobaltite composite cathodes for SOFC

General information
State: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Wang, W., Jensen, S. H., Mogensen, M. B.
Publication date: 2006

Host publication information
Title of host publication: Proceedings (cd-rom)
Performance of reversible solid oxide cells: A review

General information
State: Published
Organisations: Risø National Laboratory for Sustainable Energy
Publication date: 2006

Host publication information
Title of host publication: Proceedings (cd-rom)
Place of publication: Oberrohrdorf
Publisher: European Fuel Cell Forum
Editor: Bossel, U.
URLs:
Source: orbit
Source-ID: 309645
Research output: Research › Article in proceedings – Annual report year: 2006

Ni/YSZ-electrode passivation at cathodic current

General information
State: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Jensen, S. H., Hauch, A., Mogensen, M. B.
Pages: 247-252
Publication date: 2005

Host publication information
Title of host publication: Solid state electrochemistry. Proceedings
Place of publication: Roskilde
Publisher: Risø National Laboratory
ISBN (Print): 87-550-3455-1
Source: orbit
Source-ID: 308383
Research output: Research › Article in proceedings – Annual report year: 2005

Ni/YSZ electrodes in solid oxide electrolyser cells

General information
State: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Hauch, A., Jensen, S. H., Mogensen, M. B.
Pages: 203-208
Publication date: 2005

Host publication information
Title of host publication: Solid state electrochemistry. Proceedings
Place of publication: Roskilde
Publisher: Risø National Laboratory
ISBN (Print): 87-550-3455-1
High temperature electrolysis of steam and carbon dioxide

General information
State: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Jensen, S. H., Høgh, J., Barfod, R., Mogensen, M. B.
Pages: 204-215
Publication date: 2003

Host publication information
Title of host publication: Energy technologies for Post Kyoto targets in the medium term. Proceedings
Editors: Sønderberg Petersen, L., Larsen, H.
ISBN (Print): 87-550-3203-6
(Denmark. Forskningscenter Risoe. Risoe-R; No. 1405(EN)).
Keywords: Risø-R-1405(EN), Risø-R-1405
Electronic versions: ris_r_1405.pdf

Bibliographical note
(CD-ROM)
Source: orbit
Source-ID: 306026
Research output: Research - peer-review › Article in proceedings – Annual report year: 2003

Projects:

Modeling of Large-Scale Electricity Storage Systems based on Pressurized Reversible Solid Oxide Cells
Master of Science Thesis
Butera, G., Project Participant, Department of Mechanical Engineering, Thermal Energy
Clausen, L. R., Main Supervisor, Department of Mechanical Engineering, Thermal Energy
Jensen, S. H., Supervisor, Risø National Laboratory for Sustainable Energy, Department of Energy Conversion and Storage, Applied Electrochemistry
Campanari, S., Supervisor, Politecnico di Milano
01/10/2016 → 28/02/2017
Keywords: Natural Gas, Electricity Storage, Natural gas grid, Pressurized Solid Oxide Cells, Highly Efficient Storage, Bio-Syngas Upgrade
Collaborators: Politecnico di Milano
Documents:
Giacomo_Butera_Master_Thesis_2015_2016
Project: Research

Biofuel production based on Integrated Systems combining Biomass Gasification and Solid Oxide Cells
Butera, G., PhD Student, Department of Mechanical Engineering
Clausen, L. R., Main Supervisor, Department of Mechanical Engineering
Ahrenfeldt, J., Supervisor, Department of Chemical and Biochemical Engineering
Jensen, S. H., Supervisor, Department of Energy Conversion and Storage
Samfinansieret - Andet
01/09/2017 → 31/08/2020
Award relations: Biofuel production based on Integrated Systems combining Biomass Gasification and Solid Oxide Cells
Project: PhD

Højtemperatur-elektrolyse
Jensen, S. H., PhD Student, Department of Energy Conversion and Storage
Chorkendorf, I., Main Supervisor, Department of Physics
Hendriksen, P. V., Supervisor, Department of Energy Conversion and Storage
Mogensen, M. B., Supervisor, Department of Energy Conversion and Storage
Skaarup, S., Examiner, Department of Chemistry
Ivers-Tiffée, E., Examiner
Skou, E. M., Examiner
Risø (Løn)
01/10/2003 → 08/02/2007
Award relations: Højtemperatuurelektrolyse
Project: PhD

Co-electrolysis SOEC
Rao, M., PhD Student, Department of Energy Conversion and Storage
Hagen, A., Main Supervisor, Department of Energy Conversion and Storage
Jensen, S. H., Supervisor, Department of Energy Conversion and Storage
Hendriksen, P. V., Supervisor, Department of Energy Conversion and Storage
Anden EU-finansiering
15/08/2016 → 14/08/2019
Award relations: Co-electrolysis SOEC
Project: PhD

Modeling Energy Supply for Future Cities
Dominkovic, D. F., PhD Student, Department of Applied Mathematics and Computer Science
Pedersen, A. S., Main Supervisor, Department of Energy Conversion and Storage
Nielsen, P. S., Supervisor, Department of Management Engineering
Serensen, M. P., Supervisor, Department of Applied Mathematics and Computer Science
Jensen, S. H., Examiner, Department of Energy Conversion and Storage
Ahigren, E., Examiner, Department of Chemistry
Lund, H., Examiner
Lund, H., Examiner
Samfinansieret - Andet
15/09/2015 → 30/09/2018
Award relations: Modeling Energy Supply for Future Cities
Project: PhD

Development og highly efficient solid oxide electrolyzer cell systems
Duhn, J. D., PhD Student, Department of Management Engineering
Jensen, A. D., Main Supervisor, Department of Chemical and Biochemical Engineering
Wedel, S., Supervisor, Department of Chemical and Biochemical Engineering
Wix, C., Supervisor
Jensen, S. H., Examiner, Department of Energy Conversion and Storage
Deutschmann, O., Examiner
Mortensen, P. M., Examiner, Department of Chemical and Biochemical Engineering
Deutschmann, O., Examiner
Industrial PhD
01/12/2013 → 30/09/2017
Award relations: Development og highly efficient solid oxide electrolyzer cell systems
Project: PhD

Investigation of performance and lifetime limiting effects in Li-air battery cells
Knudsen, K. B., PhD Student, Department of Energy Conversion and Storage
Hjelm, J., Main Supervisor, Department of Energy Conversion and Storage
Jensen, S. H., Supervisor, Department of Energy Conversion and Storage
Hagen, A., Examiner, Department of Energy Conversion and Storage
Adelhelm, P., Examiner
Edström, E. K., Examiner
Adelhelm, P., Examiner
Edström, E. K., Examiner
Forskningsrådssamfinansiering
01/12/2012 → 22/02/2016
Award relations: Investigation of performance and lifetime limiting effects in Li-air battery cells
Project: PhD

Electrochemical Removal of NOx and Hydrocarbons
Friedberg, A. Z., PhD Student, Department of Energy Conversion and Storage
Kammer Hansen, K., Main Supervisor, Department of Chemistry
Jensen, S. H., Examiner, Risø National Laboratory for Sustainable Energy
Management of Lithium-air batteries - safety, reliability and performance
Christensen, A. E., PhD Student, Department of Energy Conversion and Storage
Norby, P., Main Supervisor, Department of Energy Conversion and Storage
Larsen, E., Supervisor, Department of Electrical Engineering
Vestin, K., Supervisor
Jensen, S. H., Examiner, Department of Energy Conversion and Storage
Monroe, C. W., Examiner
Møller, P. J., Examiner
Forskningsrådstipendium
01/12/2012 → 20/04/2016
Award relations: Management of Lithium-air batteries - safety, reliability and performance
Project: PhD

Performance and lifetime limiting effects in Li-ion batteries
Scipioni, R., PhD Student, Department of Energy Conversion and Storage
Jensen, S. H., Main Supervisor, Department of Energy Conversion and Storage
Hjelm, J., Supervisor, Department of Energy Conversion and Storage
Norby, P., Supervisor, Department of Energy Conversion and Storage
Bowen, J. R., Examiner, Department of Energy Conversion and Storage
Bowen, J. R., Examiner, Department of Energy Conversion and Storage
Lindbergh, G., Examiner
Lindbergh, G., Examiner
Choi, J. W., Examiner
Lindbergh, G., Examiner
Offentlig finansiering
01/06/2013 → 20/09/2016
Award relations: Performance and lifetime limiting effects in Li-ion batteries
Project: PhD

Udvikling og karakterisering af avancerede Li-batterier
Christiansen, A. S., PhD Student, Department of Chemistry
Holtappels, R., Main Supervisor, Risø National Laboratory for Sustainable Energy
Jensen, S. H., Supervisor, Risø National Laboratory for Sustainable Energy
Norby, P., Supervisor, Risø National Laboratory for Sustainable Energy
Pedersen, A. S., Examiner, Risø National Laboratory for Sustainable Energy
Dahl, S., Examiner, Department of Physics
Dominko, R., Examiner
Dominko, R., Examiner
Institut, samfinansiering
01/03/2011 → 02/09/2015
Award relations: Udvikling og karakterisering af avancerede Li-batterier
Project: PhD

Fabrication of Green Hydrocarbon Fuels via Capture and Electrolysis of CO2
Ebbehøj, S. L., PhD Student, Department of Energy Conversion and Storage
Mogensen, M. B., Main Supervisor, Department of Energy Conversion and Storage
Jensen, S. H., Supervisor, Department of Energy Conversion and Storage
Risager, A., Supervisor, Department of Chemistry
Hendriksen, P. V., Examiner, Department of Energy Conversion and Storage
Hansen, J. B., Examiner
Hartvigsen, J. J., Examiner
Bøgild Hansen, J., Examiner
Hartvigsen, J. J., Examiner
Activities:

Reversible Operation using Carbonaceous Gasses of a 30-cell Solid Oxide Cell Stack

Period: 12 Dec 2017 → 15 Dec 2017
Søren Højgaard Jensen (Guest lecturer)
Hendrik Langnickel (Guest lecturer)
Nils Hintzen (Other)
Ming Chen (Guest lecturer)
Xiufu Sun (Guest lecturer)
Anne Hauch (Guest lecturer)
Giacomo Butera (Guest lecturer)
Lasse Røngaard Clausen (Guest lecturer)
Department of Energy Conversion and Storage
Applied Electrochemistry
Mixed Conductors
Department of Mechanical Engineering
Thermal Energy

Description
Recent theoretical studies show that reversible electrochemical conversion of H2O and CO2 to CH4 inside novel pressurized solid oxide cells (SOCs) combined with subsurface storage of the produced gasses can facilitate seasonal electricity storage with a round-trip efficiency 70-80% and a storage cost below 3 ¢/kWh. Here we show test results from a 30-cell SOC stack operated with carbonaceous gasses at 18.7 bar at 700 °C in both electrolysis and fuel cell mode. The GC data from the electrolysis test results show 18% methane in the dry outlet gas, i.e. substantial methane formation inside the SOC stack. Further we observed degradation rates comparable to that of ambient pressure operation with H2/H2O gas mixtures.

Links:

Related event

7th European Fuel Cell Piero Lunghi Conference
12/12/2017 → 16/12/2017
Naples, Italy
Activity: Talks and presentations › Conference presentations

Prizes:

Third Price EFC17 Best Paper Awards
Søren Højgaard Jensen (Recipient), Hendrik Langnickel (Recipient), Nils Hintzen (Recipient), Ming Chen (Recipient), Xiufu Sun (Recipient), Anne Hauch (Recipient), Giacomo Butera (Recipient) & Lasse Røngaard Clausen (Recipient)
Department of Energy Conversion and Storage, Applied Electrochemistry, Mixed Conductors, Department of Mechanical Engineering, Thermal Energy

Description
Third price for the paper: "Reversible Operation using Carbonaceous Gasses of a 30-cell Solid Oxide Cell Stack"
The awards are given to the best papers submitted to the EFC17 conference and that report the most important insights and progress within the broad field of hydrogen and fuel cell technologies. The awards are sponsored by the EFC17 conference. All nominations are judged by an independent Best Paper Selection Committee.

Details
Awarded date: 13 Dec 2017
Degree of recognition: International
event: 7th European Fuel Cell Piero Lunghi Conference
Prize: Prizes, scholarships, distinctions

Press clippings:

Danska bränsleceller ger konstgjord bensin
Søren Højgaard Jensen
02/11/2011
Electrochemistry, Risø National Laboratory for Sustainable Energy, Fuel Cells and Solid State Chemistry Division

Media contribution (1)

Danska bränsleceller ger konstgjord bensin
02/11/2011
Klotet, En blogg och ett program av Vetenskapsradion, Sveriges Radio 2. November, Radio
DOC-QA
Søren Højgaard Jensen
Risø National Laboratory for Sustainable Energy, Fuel Cells and Solid State Chemistry Division, Electrochemistry
Press/Media: Press / Media