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.
Electrochemical impedance of an alkaline organic flow battery

Flow batteries have in recent years been recognized as a technology with high potential for use in grid-scale electrical energy storage. However, the system costs are currently impeding widespread utilisation of the technology in the energy grid. A price reduction could be achieved by changing from vanadium-based to organic-based electrolytes [1], and by increasing the system performance. The latter requires identification and quantification of the processes that contribute to voltage losses in the system. This can be done using electrochemical impedance spectroscopy (EIS), a technique that enables the resolution of the different contributions to the internal resistance within an electrochemical system. The resistance contributions are identified and resolved by fitting a model to the experimentally obtained spectra, which takes into account the various electrochemical and physico-chemical processes occurring in the system. Studies of flow battery impedance have previously been done on the vanadium system [2–4] and on an organic/organometallic system [5]. In this work, we investigate the losses in the alkaline 2,6-dihydroxyanthraquinone/ferrocyanide system, originally proposed by Lin et al. [6], through EIS. The impedance response of a full cell is measured and resolved into the series, charge-transfer, and finite diffusion resistances.

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.

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Reactivating the Ni-YSZ electrode in solid oxide cells and stacks by infiltration

The solid oxide cell (SOC) could play a vital role in energy storage when the share of intermittent electricity production is high. However, large-scale commercialization of the technology is still hindered by the limited lifetime. Here, we address this issue by examining the potential for repairing various failure and degradation mechanisms occurring in the fuel electrode, thereby extending the potential lifetime of a SOC system. We successfully infiltrated the nickel and yttria-stabilized zirconia cermet electrode in commercial cells with Gd-doped ceria after operation. By this method we fully reactivated the fuel electrode after simulated reactant starvation and after carbon formation. Furthermore, by infiltrating after 900h of operation, the degradation of the fuel electrode was reduced by a factor of two over the course of 2300h. Lastly, the scalability of the concept is demonstrated by reactivating an 8-cell stack based on a commercial design.
The present invention relates to a composite for an electrode, a composite precursor, a method of manufacturing a composite, and the composite obtained by said method. The invention further relates to an electrode comprising the composite, as well as a solid state electrochemical cell comprising the composite. The invention also relates to the use of the composite as a fuel electrode, solid oxide fuel cell, and/or solid oxide electrolyser. The invention discloses a composite for an electrode, comprising a three-dimensional network of dispersed metal particles, stabilised zirconia particles and pores, wherein the size of the pores is smaller than the size of the metal particles, wherein the size of the metal particles is

**Durable fuel electrode**

The present invention relates to a composite for an electrode, a composite precursor, a method of manufacturing a composite, and the composite obtained by said method. The invention further relates to an electrode comprising the composite, as well as a solid state electrochemical cell comprising the composite. The invention also relates to the use of the composite as a fuel electrode, solid oxide fuel cell, and/or solid oxide electrolyser. The invention discloses a composite for an electrode, comprising a three-dimensional network of dispersed metal particles, stabilised zirconia particles and pores, wherein the size of the pores is smaller than the size of the metal particles, wherein the size of the metal particles is
essentially equal to or smaller than the size of the stabilised zirconia particles, wherein the porosity is below 33, 30, or 29 vol%, more preferably below 26 or 24 vol%, and most preferably below 23, 22, 21, 18, 15, or 13 vol%, and/or wherein the pores are essentially exclusively generated from the volume created by reducing a corresponding metal oxide to the metal particles.

**A Physically-Based Equivalent Circuit Model for the Impedance of a LiFePO₄/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 LiFePO₄/Graphite cylindrical cell. The ECM is based on measurements and modeling of impedance spectra recorded separately on cathode (LiFePO₄) 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.

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Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
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Carbon deposition and sulfur poisoning during CO2 electrolysis in nickel-based solid oxide cell electrodes

Reduction of CO2 to CO and O2 in the solid oxide electrolysis cell (SOEC) has the potential to play a crucial role in closing the CO2 loop. Carbon deposition in nickel-based cells is however fatal and must be considered during CO2 electrolysis. Here, the effect of operating parameters is investigated systematically using simple current-potential experiments. Due to variations of local conditions, it is shown that higher current density and lower fuel electrode porosity will cause local carbon formation at the electrochemical reaction sites despite operating with a CO outlet concentration outside the thermodynamic carbon formation region. Attempts at mitigating the issue by coating the composite nickel/yttria-stabilized zirconia electrode with carbon-inhibiting nanoparticles and by sulfur passivation proved unsuccessful. Increasing the fuel electrode porosity is shown to mitigate the problem, but only to a certain extent. This work shows that a typical SOEC stack converting CO2 to CO and O2 is limited to as little as 15–45% conversion due to risk of carbon formation. Furthermore, cells operated in CO2-electrolysis mode are poisoned by reactant gases containing ppb-levels of sulfur, in contrast to ppm-levels for operation in fuel cell mode.
**Challenges and Possibilities of EIS on PEMEC**

Electrochemical impedance spectroscopy (EIS) has been proven a very strong electrochemical characterization tool in electrochemical research in general and in the areas of fuel cell and battery research in particular. However, this is not the case for polymer electrolyte membrane electrolysis cells (PEMEC), for which relatively few reports on the application of systematic EIS studies are available. Asking experienced researchers in the field about why, the answer has often been that these cells reveal too much electrical noise to obtain EIS with acceptable quality due to O$_2$ and H$_2$ bubble formation. Our view of the ideal structure of a PEMEC is that there ought not to be any effect of gas bubbles on the EIS as the current paths should not be disturbed by bubbles. However, we also see noise in our spectra, but the level of noise varies very much from one cell type to another. We have studied noise on three types of PEMEC and two type of alkaline electrolysis cell (AEC) for comparison. A characteristic feature of the studied PEMEC is that there is no or very little noise seen in the EIS in the frequency range above ca. 500 Hz and again not much noise below 5 Hz.

Our hypothesis is that this phenomenon is related to bubbles that are adhering to active sites of the electrocatalyst. When the catalyst layer is subjected to alternating current (AC) during the EIS then, in the PEMEC case, the O$_2$ pressure and volume of the bubbles growing on the catalyst layer will oscillate with the frequency of the AC. The volume change will naturally change with the frequency. The longer the wave period (the lower the frequency) is, the bigger is the change in the amount of O$_2$ production during an AC period. In other words, a vibration of the O$_2$ bubble size and internal pressure must be induced by the AC current. Thus, we imagine that at some low frequency, the bubbles adhering to the catalytic layer get more unstable and detach with an uneven rate from the catalyst surface. This causes the noise observed. Presumably, the structure and the properties of the interface of the catalyst to the liquid aqueous phase as well as the operation parameter will affect the frequency range and the size of noise in the EIS.
Chemical and Electrochemical Properties of La$_{0.58}$Sr$_{0.4}$Fe$_{0.8}$Co$_{0.2}$O$_{3-\delta}$ (LSCF) Thin Films upon Oxygen Reduction and Evolution Reactions

The Oxygen Evolution and Oxygen Reduction Reactions (OER/ORR), occurring at the oxygen electrode of Solid Oxide Cells (SOCs) in the two possible ways of operation, require substantial overpotentials, therefore lowering the operating efficiency of the cells. The reaction mechanisms occurring at these electrodes are still not completely understood due to their complexity and localized character at the interfaces between different materials or between the gas atmosphere and the electrocatalyst, and need in situ techniques with very high chemical sensitivity, with the additional difficulty of probing the materials as close as possible to their realistic operating conditions. In addition, the properties of LSCF are, despite numerous studies, still unclear in many aspects, despite LSCF being one of the state-of-the-art electrocatalysts used for SOCs. It is understood that the surface chemical composition deviates from the nominal bulk composition, and that secondary phases can segregate at the surfaces and interfaces during operation. Furthermore, the electrochemical properties such as Area Specific Resistance (ASR), oxygen exchange coefficient ($k_{\text{ex}}$), ASR activation energy ($E_a$) and $pO_2$-exponents for LSCF reported in the literature vary considerably. This study aims to better understand the properties of LSCF, by combining the results of Electrochemical Impedance Spectroscopy (EIS) and Near-Ambient Pressure X-ray Photoelectron Spectroscopy (NAP-XPS) on model electrodes, both in polarized and unpolarized conditions. In particular, NAP-XPS studies of the surface chemistry evolution under operation, as well as the correlation between surface potential changes in relation to the applied overpotential are addressed, in an attempt to determine the real driving force for the oxygen reactions. For this purpose, thin films of LSCF are deposited by Pulsed Laser Deposition (PLD) through shadow masks, in order to obtain well-defined electrode geometries with low risk of contamination, and subsequently tested both in highly clean EIS measuring setups and at the synchrotron beamline. The results of both kinds of experiments are correlated, the goal being a better understanding of the material’s properties under operation, as well as possible degradation phenomena.

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Chemical and Electrochemical Properties of La$_{0.58}$Sr$_{0.4}$Fe$_{0.8}$Co$_{0.2}$O$_{3-\delta}$ (LSCF) Thin Films upon Oxygen Reduction and Evolution Reactions
Determination of the bonding strength in solid oxide fuel cells' interfaces by Schwickerath crack initiation test

An adaptation of the Schwickerath crack initiation test (ISO 9693) was used to determine the bonding strength between an anode support and three different cathodes with a solid oxide fuel cell interconnect. Interfacial elemental characterization of the interfaces was carried out by SEM/EDS analysis on fracture surfaces to investigate the bonding mechanisms. SEM/EDS of fresh fractures were also performed to determine the cohesion/adhesion mechanism of bonding. Calculations of the residual stresses were determined by finite element simulation using ANSYS, based on thermo-mechanical properties of the materials obtained by measurement, calculation or literature.
Electrochemical Characterization of a PEMEC Using Impedance Spectroscopy

In this study, electrochemical impedance spectroscopy (EIS) is applied in combination with cyclic voltammetry (CV) and current density - cell voltage curves (iV-curves) to investigate the processes contributing to the total impedance of a polymer electrolyte membrane electrolysis cell (PEMEC). iV-curves were linear above 0.35 A cm\(^{-2}\) implying ohmic processes to be performance limiting, however the impedance spectra showed three arcs indicating three electrochemical reactions at these conditions not to be purely ohmic, but also to have capacitive properties. A hypothesis that the composite IrO\(_x\)/Nafion anode catalyst layer causes two of these arcs with a constant sum of resistance and current constrictions cause the third arc, is suggested. This hypothesis implies that the total differential cell resistance at current densities above 0.35 A cm\(^{-2}\) is purely ascribed to protonic resistance in Nafion in this type of PEMEC.
Lifetime limiting effects in pre-commercial solid oxide cell devices

The solid oxide electrochemical cell technology is promising for efficient energy storage, especially when the share of intermittent renewable electricity production is high. The technology is being commercialized in niche markets, but large-scale employment is still hindered by limited durability of the devices. The lifetime limiting mechanisms are addressed in this work.

A general introduction into mechanisms limiting the durability is presented. A database of more than 50 parameters from 150 publications and 1,000,000 hours of accumulated testing was established, and a quantitative analysis of degradation and lifetime was conducted. It is shown that the technology is approaching the official targets required for commercialization, but that work remains to be done.

It is further recognized that targeting niche applications initially will allow for employment of economies of scale, which will bring down costs and facilitate entry into larger markets. Here, we examine electrochemical reduction of CO2 to CO and one of the main failure mechanisms related to it. Carbon formation on the nickel electrocatalyst can be detrimental to the microstructural integrity of the cell. It is found that the possible operating window is severely limited due to gradients of temperature, gas concentration, and overpotential across the electrode. These affects also apply to stack- and system-level, and the results obtained are combined with modeling and stack testing experiences. Thus, on account of this mechanism, the possible outlet CO concentration is limited by up to 50% below the thermodynamic carbon deposition threshold based on the inlet temperature, depending on design and operating strategy.

Replacement of the Ni electrocatalyst would increase the stability towards this issue and may improve the robustness in other ways as well. Ceria has been reported as a potential candidate in such endeavors. Thin film electrodes of nickel and ceria are therefore studied as model systems using near-ambient pressure x-ray photoelectron spectroscopy to further the fundamental understanding of the carbon formation mechanism. The reaction occurs further from the thermodynamic threshold on ceria, and fundamental mechanisms for electrochemically driven carbon growth are suggested based on observed adsorbate species.

By infiltrating ceria after degradation has already occurred, the robustness and lifetime of the cells are increased. Complete reactivation of the fuel electrode is achieved after otherwise detrimental failure mechanisms have occurred, such as reactant starvation and carbon formation. Moreover, the degradation of the electrode over the course of nearly 2500 hours is essentially eliminated by infiltrating after microstructural stabilization had occurred. Lastly, the method is scaled up by replicating the positive effects of post-degradation infiltration on an 8-cell stack.

General information
An Ag based brazing system with a tunable thermal expansion for the use as sealant for solid oxide cells

An Ag-Al2TiO5 composite braze was developed and successfully tested as seal for solid oxide cells. The thermo-mechanical properties of the Ag-Al2TiO5 system and the chemical compatibility between this composite braze and relevant materials used in stacks were characterized and the leak rates as a function of the operation temperature were measured. The thermal expansion coefficient in the Ag-Al2TiO5 system can be tailored by varying the amount of the ceramic filler. The brazing process can be carried out in air, the joining partners showed a good chemical stability and sufficient low leak rates were demonstrated. Furthermore, the long-term stability of the Ag-Al2TiO5 composite braze was studied under relevant SOFC and SOEC conditions. The stability of brazed Crofer/Ag-Al2TiO5/NiO-YSZ assemblies in reducing atmosphere and in pure oxygen was investigated over 500 h at 850 °C. Additionally, a cell component test was performed to investigate the durability of the Ag-Al2TiO5 seal when exposed to dual atmosphere. The seals performed well over 900 h under electrolysis operation conditions (~0.5 A cm², 850 °C), and no cell degradation related to the Ag-Al2TiO5 sealing was found, indicating that the developed braze system is applicable for the use in SOFC/SOEC stacks.

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An Electrochemical Impedance Spectroscopy Study on the Effects of the Surface- and Solution-Based Mechanisms in Li-O2 Cells

The maximum discharge capacity in non-aqueous Li-O2 batteries has been limited to a fraction of its theoretical value, largely due to a conformal deposition of Li2O2 on the cathode surface. However, it has recently been established that additives that increase the shielding of either O2 or Li+ will activate the formation of toroidal shaped Li2O2, thereby dramatically increasing cell capacity. Here we apply porous electrode theory to electrochemical impedance measured at the Li-O2 cathode to investigate changes in the surface- and ionic resistance within the pores under conditions where either the surface-mechanism or the solution-mechanism is favored. Our experimental observations show that (i) an additional charge transfer process is observed in the impedance spectrum where the solution-based mechanism is favored; (ii) the changes in the ionic resistance in the cathode during discharge (related to Li2O2 build up) is much greater in cells where the solution-based mechanism is activated and can qualitatively determine the extent of discharge product deposited within the pores of the cathode versus the deposition extent at the electrode/electrolyte interface; and (iii) that the observed “sudden-death” during discharge is a consequence of the increasing charge transfer resistance.
regardless of whether Li$_2$O$_2$ forms predominantly through either the surface- or solution-based mechanism.
An Electrochemical Impedance Study of the Capacity Limitations in Na–O2 Cells

Electrochemical impedance spectroscopy, pressure change measurements, and scanning electron microscopy were used to investigate the nonaqueous Na–O2 cell potential decrease and rise (sudden deaths) on discharge and charge, respectively. To fit the impedance spectra from operating cells, an equivalent circuit model was used that takes into account the porous nature of the positive electrode and is able to distinguish between the electrolyte resistance in the pores and the charge-transfer resistance of the pore walls. The results obtained indicate that sudden death on discharge is caused by, depending on the current density, either accumulation of large NaO2 crystals that eventually block the electrode surface and/or a thin film of NaO2 forming on the cathode surface at the end of discharge, which limits charge-transfer. The commonly observed sudden rise in potential toward the end of charge may be caused by a concentration depletion of NaO2 dissolved in the electrolyte near the cathode surface and/or an accumulation of degradation products on the cathode surface.

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Electrochemical Characterization of PEMECs Operating at Various Current Densities
This conference contribution touches upon electrochemical characterization of operating polymer electrolyte membrane electrolysis cells (PEMECs) by the application of electrochemical impedance spectroscopy (EIS). Analysis of differences in impedance spectra (ADIS) (Jensen et al., 2007) can be applied to gain insight into the relative magnitudes of resistance contributions from the two electrodes, the electrolyte and of mass transfer limitations and can help identifying the time scale of the respective processes. The gained knowledge may facilitate further development of the PEMECs.

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Electrochemical Impedance Spectroscopy on Industrially-Relevant Solid Oxide Electrolyzer Cell Stacks: A Powerful Tool for in-Situ Investigations of Degradation Mechanisms
In the current efforts of moving energy production to renewable sources, wind and solar energy are widely considered as the key technologies to cover our growing demands. However, the fluctuating nature of these sources requires a flexible energy system and storage technologies to ensure that energy services can be covered in a stable and affordable manner. One promising solution is the synthetic fuel production by solid oxide electrolyzers. Electricity can be stored in a power-to-gas process during times of excess electricity production and then further converted to liquid fuels for e.g. transportation, or at high demands converted back to electricity by either conventional power plants or fuel cells.

One of today’s biggest hurdles for a successful commercialization of solid oxide electrolyzers is the stack’s lifetime with current industry targets in the order of five to ten years. To identify and quantify the different degradation mechanisms sensitive in-situ analysis tools are needed. On single cell level, electrochemical impedance spectroscopy (EIS) is a well-established diagnostics tool. On stack level EIS has been shown to be more difficult because of geometrical restrictions of the stack design and significantly lower resistances due to the larger active cell area. Nevertheless, it is becoming a more and more important technique for stack diagnostics.

Here we present impedance spectroscopy results of two solid oxide stacks provided by Haldor Topsoe A/S. The first stack was a 14-cell stack (Delta design) specifically optimized for EIS measurements, while the other stack was an 8-cell stack (TSP-1 design), where impedance measurements were carried out without major modifications to the stack. The individual cell voltages were monitored simultaneously by EIS during up to 2000 hours in electrolysis within the ForskEL project 2015-1-12270 “Towards Solid Oxide Electrolysis Plants in 2020”, funded by Danish Energet.dk. The analysis provides valuable insight into the degradation processes which could not have been obtained by current-voltage-data alone.

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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.
Investigation of the bonding strength and bonding mechanisms of SOFCs interconnector-electrode interfaces

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Kinetic Studies on State of the Art Solid Oxide Cells: A Comparison between Hydrogen/Steam and Reformate Fuels

Electrochemical reaction kinetics at the electrodes of Solid Oxide Cells (SOCs) were investigated at 700 °C for two cells with different fuel electrode microstructures as well as on a third cell with a reduced active electrode area. Three fuel mixtures were investigated – hydrogen/steam and reformate fuels hydrogen/carbon-dioxide and hydrogen/methane/steam. It was found that the kinetics at the fuel electrode were exactly the same in both reformates. The hydrogen/steam fuel displayed slightly faster kinetics than the reformate fuels. Furthermore the gas conversion impedance in the hydrogen/steam fuel split into two processes with opposing temperature behavior in the reformate fuels. An 87.5 % reduction in active electrode area diminishes the gas conversion impedance in the hydrogen/steam fuel at high fuel flow rates. In both reformates, the second and third lowest frequency processes merged into a single process as the gas conversion was reduced. The SOC with finer electrode microstructure displayed improved kinetics.
Original language: English
Keywords: Continuously stirred tank reactor, Electrochemical impedance spectroscopy, Electrode potential, Gas conversion, Kinetics, Plug flow, Pseudo-gas conversion, Reformate fuel, Solid oxide fuel cell
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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 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).

Quantitative review of degradation and lifetime of solid oxide cells and stacks

A comprehensive review of degradation and lifetime for solid oxide cells and stacks has been conducted. Based on more than 50 parameters from 150 publications and 1,000,000 hours of accumulated testing, this paper presents a quantitative analysis of the current international status of degradation and lifetime in the field. The data is used to visualize specific trends regarding choice of materials, operating conditions and degradation rates. The average degradation rate reported is decreasing and is quickly approaching official targets. The database is published online for open-access and a continued updating by the community is encouraged. Furthermore, the commonly reported test parameters and degradation indicators are discussed. The difficulty in standardizing testing due to variations in cell and stack design, materials and intended purpose of the system is acknowledged. A standardization of reporting of long-term single-cell and stack tests is proposed.
**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 charge-transfer 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.

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**Analysis of the Interphase on Carbon Black Formed in High Voltage Batteries**

Carbon black (CB) additives commonly used to increase the electrical conductivity of electrodes in Li-ion batteries are generally believed to be electrochemically inert additives in cathodes. Decomposition of electrolyte in the surface region of CB in Li-ion cells at high voltages up to 4.9 V is here studied using electrochemical measurements as well as structural and surface characterizations. LiPF₆ and LiClO₄ dissolved in ethylene carbonate:diethylene carbonate (1:1) were used as the electrolyte to study irreversible charge capacity of CB cathodes when cycled between 4.9 V and 2.5 V. Synchrotron-based soft X-ray photoelectron spectroscopy (SOXPES) results revealed spontaneous partial decomposition of the electrolytes on the CB electrode, without applying external current or voltage. Depth profile analysis of the electrolyte/cathode interphase indicated that the concentration of decomposed species is highest at the outermost surface of the CB. It is concluded that carboxylate and carbonate bonds (originating from solvent decomposition) and LiF (when LiPF₆ was used) take part in the formation of the decomposed species. Electrochemical impedance spectroscopy measurements and transmission electron microscopy results, however, did not show formation of a dense surface layer on CB particles.
Analysis of the Interphase on Carbon Black Formed in High Voltage Batteries

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An Electrochemical Impedance Spectroscopy Investigation of the Overpotentials in Li−O2 Batteries
Lithium−O2 (Li−O2) batteries are currently limited by a large charge overpotential at practically relevant current densities, and the origin of this overpotential has been heavily debated in the literature. This paper presents a series of electrochemical impedance measurements suggesting that the increase in charge potential is not caused by an increase in the internal resistance. It is proposed that the potential shift is instead dictated by a mixed potential of parasitic reactions and Li2O2 oxidation. The measurements also confirm that the rapid potential loss near the end of discharge (“sudden death”) is explained by an increase in the charge transport resistance. The findings confirm that our theory and conclusions in ref 1, based on experiments on smooth small-area glassy carbon cathodes, are equally valid in real Li−air batteries with porous cathodes. The parameter variations performed in this paper are used to develop the understanding of the electrochemical impedance, which will be important for further improvement of the Li−air battery.
A Study of $e^-$ Transport through Li$_2$O$_2$, the Main Discharge Product in the Li-O$_2$ Battery

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 Li-ion technologies and the demand placed on batteries by technologies such as electrical vehicles [1]. However, the Li-O2 battery still suffers greatly from high overpotentials during oxygen reduction and evolution reactions (discharge and charge, respectively), poor rechargeability, and decomposition of salts and solvents etc. [2] [3]. In order to improve the electrochemical performance of the Li-O2 batteries; it is crucial to understand the fundamental mechanisms that governs and limits the system during electrochemical operation. Here we present a redox probing study of the charge transfer across the deposition product lithium peroxide, Li$_2$O$_2$, using outer-sphere redox shuttles: cobaltocene, ferrocene, and decamethylferrocene. The change in heterogeneous electron transfer exchange rate as a function of the potential and the Li$_2$O2 layer thickness (~state of charge) was determined using electrochemical impedance spectroscopy. The attenuation of the electron transfer exchange rate with film thickness is dependent on the probing potential, indicating that hole tunneling is the dominant process for charge transfer through Li$_2$O$_2$ supporting previous work by Luntz et al. [4]. Additionally, this work includes the determination of diffusion coefficients and concentrations of the redox shuttles and the superoxide ion, in a 1,2-dimethoxyethane based electrolyte. References [1] S. A. Freunberger, P. G. Bruce, L. J. Hardwick et J.-M. Tarascon, «Li-O2 and Li-S batteries with high energy storage,» Nature Materials, vol. 11, pp. 19-29, 2012. [2] B. D. McCluskey, A. Valery, A. C. Luntz, S. R. Gowda, G. M. Wallraff, J. M. Garcia, T. Mori et L. E. Krupp, «Combining Accurate O2 and Li2O2 Assays to Separate Discharge and Charge stability Limitations in Nonaqueous Li-O2 Batteries,» J. Phys. Chem. Lett., vol. 4, pp. 2989-2993, 2013. [3] R. Younesi, M. Hahlin, F. Björefors, P. Johansson et K. Edström, «Li-O2 Battery Degradation by Lithium Peroxide (Li2O2): A Model Study,» Chem. Mater., vol. 25, pp. 77-84, 2013. [4] A. C. Luntz, V. Viswanathan, J. Voss, J. B. Varley, J. K. Nørskov, R. Scheffler and A. Speidel, “Tunneling and Polaron Charge Transport through Li$_2$O$_2$ in Li-O2 Batteries,” J. Phys. Chem. Lett., vol. 4, pp. 3494-3499, 2013.
reasons for these observations will be discussed in detail.

In an attempt to mitigate the degradation due to carbon deposition, the Ni-YSZ electrode was infiltrated with a gadolinium doped ceria (CGO) solution. Initial results indicate that the coking tolerance was not enhanced, but it is still unclear whether infiltrated cells degrade less. However, infiltrated cells display a significant performance enhancement before coking, especially under electrolysis current.

The investigation thus indicated carbon formation in the Ni containing fuel electrode before the thermodynamically calculated threshold for average measurements of the cell was reached. The deviation from the average threshold was reproduced on several cells and quantified (figure 1). The observation of carbon formation on a local, microstructural level before the expected thermodynamic threshold for average measurements on the cell level, is of crucial importance when choosing operating conditions for commercial systems. The effect would be even more severe on stack level, where the gas diffusion and temperature gradients are more pronounced. Initial results of the mitigation strategy of infiltrating CGO are negative, but increased performance prior to coking was observed.

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Carbon Deposition during CO2 Electrolysis in Ni-Based Solid-Oxide-Cell Electrodes
The carbon formation threshold in an operating cell was investigated during electrolysis of an idealized reactant atmosphere of CO and CO2. The electrolysis current was gradually increased in steps until the cell voltage spontaneously increased, thereby indicating cell degradation and possibly the onset of carbon deposition. The outlet gas composition at each current step was estimated based on the inlet gas composition and the reactant conversion using Faraday's law. The increase in voltage was observed at lower outlet pCO/pCO2 ratios than that corresponding to the expected thermodynamic threshold for carbon formation. The degradation observed was related to the fuel electrode, as confirmed by electrochemical impedance spectroscopy. Mitigation of the degradation mechanism was attempted by infiltrating gadolinium doped ceria. The onset of carbon deposition was largely unaffected, but the polarization resistance of the electrode during electrolysis was significantly decreased.

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Degradation Studies on LiFePO₄ cathode

In this paper we examine a laboratory LiFePO₄ (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 Li₁₋ₓFePO₄ particles in a degraded LFP electrode and low-acceleration voltage (1 kV) FIB/SEM analysis allowed us to obtain phase contrast between FePO₄ and LiFePO₄. 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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Electrochemical Impedance Spectroscopy as a Tool for PEMECs Development

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Electrode Kinetics and Gas Conversion in Solid Oxide Cells
The solid oxide fuel cell (SOFC) converts hydrogen, carbon monoxide and hydrocarbon fuels (directly) into electricity with very high efficiencies and has demonstrated almost comparable performance when operated in reverse mode as a solid oxide electrolysis cell (SOEC). In this case electrical (and thermal) energy is stored as chemical energy of reaction products. To this end, the cells are fed with steam (H2O electrolysis), carbon dioxide (CO2 electrolysis) or a mixture of both (H2O/CO2 co-electrolysis) and of course electrical (ΔG) and thermal (TΔS) energies for the splitting of reactant compounds. Hydrogen, carbon monoxide or both (synthesis gas) are produced at the fuel electrode meanwhile oxygen is produced at the oxygen electrode. In reversible or cyclic mode the solid oxide cell is operated alternatingly as fuel cell or electrolysis cell depending on the needs of the end user.

Upon polarization of the solid oxide cell (SOC) and independent of polarization mode (fuel cell mode or electrolysis mode), the current flowing through the cell is limited by processes such as adsorption and desorption of reactants or products, diffusion through the porous electrodes, activation or charge transfer at the reaction sites, gas conversion at the reaction sites and flow fields and ohmic drop across the electrolyte. These processes occur in both electrodes and often their characteristic frequencies overlap, rendering characterization of a given mechanism particularly challenging. To optimize the SOCs for operation in the different fuels, operation temperature and operation modes it is important to understand the kinetics of the SOC electrodes. This thesis was aimed at understanding the kinetics of the SOC under different operation conditions of temperature, polarization, and fuel mixture. For investigations on full cells, electrochemical impedance
spectroscopy and distribution of relaxation times techniques were used to investigate kinetics of the Ni/YSZ fuel electrode in three fuel mixtures – hydrogen/steam and reformate fuels hydrogen/carbon-dioxide and hydrogen/methane/steam. It was found that the kinetics at the fuel electrode were exactly the same in both reformates. This means that chemical equilibrium reactions were much faster than the electrochemical reactions. The electrode displayed slightly faster kinetics in hydrogen/steam fuel than in the reformate fuels.

To minimize the influence of (i) joule heating effects as a result of current flow across the electrolyte, (ii) concentration-related effects like gas diffusion, and (iii) overlapping of the characteristic frequencies of processes, the investigations were extended from full cell geometries to a novel pseudo-three electrode cell geometry with working-electrode areas of ca. 1 mm² that enabled isolated investigation of the fuel and oxygen electrodes. In a 50/50 H2/H2O fuel mixture, the Ni8 mol % yttria-stabilized zirconia (Ni/YSZ) fuel electrode showed slower reaction kinetics operating under cathodic polarisation than anodic— the same finding had been reported in literature from investigations on full cells whereby the LSCF electrode showed a larger increase in polarization resistance together with the local pH2O, substrate diffusion (specifically Knudsen diffusion) was identified as one of the causes of asymmetry between anodic and cathodic mode polarization. Obtained charge-transfer coefficients compared well with those reported in literature and their evolution with temperature was similar to that reported in literature based on porous Ni/YSZ fuel electrodes. From the two investigated oxygen electrodes, the higher performing (La0.6Sr0.4)0.99CoO3/2-δ (LSCF) oxygen electrode showed slower reaction kinetics under cathodic mode operation at 50 mV overvoltage than in anodic mode. The trend was opposite for the lower performing La0.58Sr0.4Co0.2Fe0.8O3 (LSFC) oxygen electrode. However, with decreasing oxygen partial pressure both electrodes displayed increasing asymmetry between anodic and cathodic modes. It could be shown that surface exchange kinetics were the major cause of the decreasing kinetics with decreasing pO2 and that the cathodic mode kinetics were slowed down much more than the anodic branch kinetics thus increasing the asymmetry. Independent of operation mode, commercialization of the SOC technology requires a guarantee of longevity as well as predictability of the SOC performance under desired operation conditions.

The performance is generally evaluated through the current/voltage (C/V) curve. As such, a deviation from the expected/predicted performance curve can serve to identify the presence of ageing or an ageing inducing process. A 0-D stationary model was previously developed at the Institut für Angewandte Materialien - Werkstoffe der Elektrotechnik (IAM-WET) in Germany to predict the C/V curve of a SOC for fuel cell operation mode. In this thesis, the applicability of this model was verified for electrolysis mode operation, the model was extended to accommodate temperature changes under polarization in fuel cell and electrolysis mode operation, and the model was further extended to cover operation in reformate fuels H2/H2O/CO/CO2. The latter was accomplished by including a new concentration-related overpotential contribution in the model to account for the CO/CO2 diffusion to the reaction sites as a result of the water gas shift equilibrium reactions.

The long-term stability of the system depends on whether the system is operated solely in fuel-, electrolysis-, reversible or dynamic mode. Optimization of the cells for high performance and/or durability in each of these operation modes requires a thorough understanding of the processes and mechanisms affecting the kinetics and ageing of the systems. In five tests with varying durations between 1000 h and 2500 h the long-term stability of the SOCs was investigated for constant electrolysis, cyclic and dynamic operation modes in a symmetric binary fuel of 50/50 H2/H2O at 800 °C and 700 °C. The SOCs investigated under constant electrolysis mode aged more than those investigated under cyclic mode with the fuel electrode dominating the ageing in constant electrolysis mode and the oxygen electrode dominating that in cyclic mode. During dynamic cycling, the SOCs aged less at 800 °C than at 700 °C. It was observed that for cycles with equal durations in SOEC and SOFC modes whereby the cycle lengths were less than or equal to 2 x 5 h the voltage ageing was almost symmetrical for both SOFC and SOEC modes. For longer cycle lengths SOEC mode voltage ageing was at least double the SOFC mode voltage ageing. This result is consistent with suggestions in literature that intermittent operation of SOCs in fuel cell mode slows down or even reverses SOC ageing that occurs during long-term electrolysis operation. Based on the finding that the fuel electrode dominated the ageing under constant electrolysis operation, it was speculated based on literature, to be caused by precipitation of nickel oxide that had diffused into the YSZ matrix of the fuel electrode during sintering. Constant electrolysis operation provided enhanced conditions for the precipitation of the nickel oxide as metallic nickel. Furthermore, Zr, Ni, Y, and O containing nano-particles were found on the Ni particles. In literature during electrolysis of H2O/CO2 under same conditions of temperature and current density nano particles were also found on Ni particles, identified as ZrO2 and attributed the major cause of fuel electrode ageing. In cyclic operation these enhancing conditions were not maintained long-enough for severe nickel precipitation. It is known and was verified that the LSCF/CGO electrode is better performing than the LSCF electrode. To compare the stability of these two state-of-the-art (s.t.a.) oxygen electrodes, 1000 h tests under non-polarized or open circuit voltage (OCV) conditions were carried out using symmetric cell geometry. Both electrodes displayed a two-step ageing trend with rapid initial ageing within the first 400 h followed by relaxation to slower ageing rates. The LSCF electrode showed a larger increase in polarization resistance especially within the first 400 h in which it aged by factor 6 faster than the LSC/CGO electrode. The rapid ageing of the LSCF electrode within the first 300 – 400 h of operation has also been reported in literature.

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Enhancement of the chemical stability in confined δ-Bi₂O₃

Bismuth-oxide-based materials are the building blocks for modern ferroelectrics, multiferroics, gas sensors, light photocatalysts, and fuel cells. Although the cubic fluorite δ-phase of bismuth oxide (δ-Bi₂O₃) exhibits the highest conductivity of known solid-state oxygen ion conductors, its instability prevents use at low temperatures. Here we demonstrate the possibility of stabilizing δ-Bi₂O₃ using highly coherent interfaces of alternating layers of Er₂O₃-stabilized δ-Bi₂O₃ and Gd₂O₃-doped CeO₂. Remarkably, an exceptionally high chemical stability in reducing conditions and redox cycles at high temperature, usually unattainable for Bi₂O₃-based materials, is achieved. Even more interestingly, at low oxygen partial pressure the layered material shows anomalous high conductivity, equal or superior to pure δ-Bi₂O₃ in air. This suggests a strategy to design and stabilize new materials that are comprised of intrinsically unstable but high-performing component materials.
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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Influence of hydroxyl content of binders on rheological properties of cerium-gadolinium oxide (CGO) screen printing inks

The influence of hydroxyl content of binders on rheological properties of screen printing inks is investigated. The actual amount of hydroxyl groups is correlated to the level of hyper-entanglement that characterizes the binders in solution. Three of the most used binders (ethyl cellulose, and two vinyl resins) were selected and characterized in solution via viscosimetry method. A high degree of hyper-entanglement was observed for ethyl cellulose polymers, whereas a mitigated effect characterized the two vinyl resins. Cerium-gadolinium oxides (CGO)-based inks, prepared using the selected binders, were investigated by means of rheology. The vinyl resin at higher hydroxyl content and low level of hyper-entanglement was demonstrated to impart superior printability properties.

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Kinetic Studies on Ni-YSZ Composite Electrodes

Introduction Polarization of the Solid Oxide Cell (SOC) causes current to flow. If the fuel electrode is anodically polarized, the cell operates in fuel cell mode, oxidizing a fuel like hydrogen, carbon monoxide or hydrocarbons. In cathodic polarization the cell operates in electrolysis mode, reducing steam, carbon dioxide or both at the fuel electrode. Independent of polarization direction, the current flowing through the electrodes of an SOC is limited by processes such as adsorption and desorption of reactants or products, diffusion through the porous electrodes, activation or charge transfer at the reaction sites gas conversion at the flow fields, and ohmic drop across the electrolyte. Since these processes occur in both electrodes and some of them with overlapping characteristic frequencies, it is particularly challenging to isolate and characterize a particular mechanism. Furthermore, when polarized, the cell heats up due to joule heating of the electrolyte but also the electrodes either heat or cool due to exothermic oxidation or endothermic reduction of gaseous reactant species. Kinetic investigation of SOC electrodes independent of the above effects thus requires a carefully chosen cell geometry, methodology and operation conditions. Experimental The investigated cells consist of porous Ni/8YSZ composite working-electrodes with an active area between 0.8 and 1 mm² and ~100 mm² counter electrodes of the same...
material screen-printed on a special shaped 8YSZ electrolyte pellet. The electrodes are sintered in air at 1350 °C. Details of the cell geometry are given elsewhere. The cells were characterized by electrochemical impedance spectroscopy using a Gamry Reference 600TM potentiostat. Current/voltage characteristics were recorded at different temperatures and gas compositions using the same instrument. The tests are carried out in a single gas atmosphere with maximum flow rate of 6 L/h. Results and Discussion Current density vs working electrode overpotential curves recorded in the temperature range 800 – 650°C in a 50/50 H2/H2O fuel mixture are displayed in figure 1(a). The curve at 700°C shows that for a current density of 100 mA/cm2 in cathodic polarization, an overpotential of ca. 150 mV is required, compared with 100 mV in anodic polarization. This reflects asymmetry in the kinetics of hydrogen oxidation and steam reduction. By recording current density vs overpotential curves at H2/H2O ratios of 30/70, 50/50 and 70/30 as displayed in figure 1(b) it could be shown that in the potential window investigated herein the dependence of kinetics on H2/H2O ratio is not significant. At any given potential in the investigated window, and independent of operation mode, there is a slight increase in current density with increasing steam content consistent. This translates to a decreasing area specific resistance of the fuel electrode electrochemistry with pH2O. A power law dependency of ~0.33 is reported in literature. Outlook In this work experimental results of kinetic investigations on state of the art solid oxide cell electrodes carried out using a novel solid oxide cell geometry, allowing, for the very first time, determination of kinetic parameters void of influences such as temperature or reactant starvation will be presented. The results will provide a basis for discussion of existing analytical descriptions of the current/overpotential relations of SOC electrodes. References: 1. C. Graves, T. L. Skafe, B. R. Sudireddy, J. Nielsen, M. Mogensen, in preparation. 2. T. Kawada et al., J. Electrochem. Soc., 137, 3042–3047 (1990). 3. J. Mizusaki et al., Solid State Ionics, 70-71, 52–58 (1994). 4. C. R. Graves, S. D. Ebbesen, and M. Mogensen, in ECS Transactions,, vol. 25, p. 1945–1955, ECS (2009). 5. P. Hottappels, L. G. J. de Haart, and U. Stimming, J. Electrochem. Soc., 146, 1620–1625 (1999). 6. J. Njodzefon, D. Klotz, A. Weber, and E. Ivers-Tiffée, J. Electrochem. Soc., 160(2013). 7. A. Leonide, Y. Apel, and E. Ivers-Tiffée, in ECS Transactions,, vol. 19, p. 81–109, ECS (2009). Figures: Figure 1: Current density vs overpotential curves recorded (a) in the temperature range 800- to 650°C in a 50/50 H2/H2O ratio and (b) at 800°C in H2/H2O ratios 30/70, 50/50 and 70/30. [Figure]
Kinetic Studies on State of the Art Solid Oxide Cells – A Comparison between Hydrogen/Steam and Reformate Fuels

Electrochemical reaction kinetics at the electrodes of Solid Oxide Cells (SOCs) were investigated at 700 °C for two cells with different fuel electrode microstructures as well as on a third cell with a reduced active electrode area. Three fuel mixtures were investigated – hydrogen/steam and reformate fuels hydrogen/carbon-dioxide and hydrogen/methane/steam. It was found that the kinetics at the fuel electrode were exactly the same in both reformates. The hydrogen/steam fuel displayed slightly faster kinetics than the reformate fuels. Furthermore the gas conversion impedance in the hydrogen/steam fuel split into two processes with opposing temperature behavior in the reformate fuels. An 87.5% reduction in active electrode area diminishes the gas conversion impedance in the hydrogen/steam fuel at high fuel flow rates. In both reformates, the second and third lowest frequency processes merged into a single process as the gas conversion was reduced. The SOC with finer electrode microstructure displayed improved kinetics.

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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,
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.
Reactions and SEI Formation during Charging of Li-O₂ Cells

In this letter we combine detailed electrochemical impedance measurements with quantitative measurements of O₂ evolution and Li₂O₂ oxidation to describe the charge mechanisms during charge of Li-O₂ batteries with porous carbon electrodes. We identify Li₂O₂ oxidation at 3.05 V and an apparent chemical formation of a solid electrolyte interface (SEI) layer as the first monolayer of Li₂O₂ is oxidized, leading to a voltage increase. The first electrochemical degradation reaction is identified between 3.3 V and 3.5 V, and the chemical degradation is limited above 3.5 V, suggesting that a chemically stable SEI layer has been formed.

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Redox Probing Study of the Potential Dependence of Charge Transport Through Li$_2$O$_2$

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−O$_2$ 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, Li$_2$O$_2$, in the Li−O$_2$ battery using outer-sphere redox shuttles. The change in heterogeneous electron transfer exchange rate as a function of the potential and the Li$_2$O$_2$ 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 Li$_2$O$_2$ and showing that the origin of the sudden death observed upon discharge is due to charge transport limitations.
Advanced impedance modeling of solid oxide electrochemical cells

Impedance spectroscopy is a powerful technique for detailed study of the electrochemical and transport processes that take place in fuel cells and electrolysis cells, including solid oxide cells (SOCs). Meaningful analysis of impedance measurements is nontrivial, however, because a large number of modeling parameters are fit to the many processes which often overlap in the same frequency ranges. Also, commonly used equivalent circuit (EC) models only provide zero-dimensional (0-D) approximations of the processes of the two electrodes, electrolyte and gas transport. Employing improved analytical techniques to provide good guesses for the modeling parameters, like transforming the impedance data to the distribution of relaxation times (DRT), together with experimental parameter sensitivity studies, is the state-of-the-art approach to achieve good EC model fits.

Here we present new impedance modeling methods which advantageously minimize the number of modeling parameters...
and the parameters used have direct physicochemical meaning. This is accomplished by (i) employing an improved cell model where the representative 0-D resistive-capacitive type EC elements are replaced by analytical 1-D porous electrode and 2-D gas transport models which have fewer unknown parameters for the same number of processes, (ii) use of a new model fitting algorithm, “multi-fitting”, in which multiple impedance spectra are fit simultaneously with parameters linked based on the variation of measurement conditions, (iii) constraining the parameter values during fitting to ranges of physically reasonable values.

Using these methods, the number of fitting parameters for four impedance spectra measured with isolated changes to the fuel and oxidant gas compositions, has been reduced from 80 to 21-34 depending on the model. The obtained results include structural parameters like porosity and tortuosity; or if those characteristics are known, use of even fewer fitting parameters is possible. The methods have been implemented in a software package written by one of the authors, which also implements many previously used impedance analysis methods and integrates the analysis process in a modular workflow – data validation (Kramers-Kronig), clean-up, visualization (DRT and others), modeling (nonlinear least-squares fitting), and final plotting for publication.

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Communication: The influence of CO₂ poisoning on overvoltages and discharge capacity in non-aqueous Li-Air batteries
The effects of Li₂CO₃ like species originating from reactions between CO₂ and Li₂O₂ at the cathode of non-aqueous Li-air batteries were studied by density functional theory (DFT) and galvanostatic charge-discharge measurements. Adsorption energies of CO₂ at various nucleation sites on a stepped (1 ̅100) Li₂O₂ surface were determined and even a low concentration of CO₂ effectively blocks the step nucleation site and alters the Li₂O₂ shape due to Li₂CO₃ formation. Nudged elastic band calculations show that once CO₂ is adsorbed on a step valley site, it is effectively unable to diffuse and impacts the Li₂O₂ growth mechanism, capacity, and overvoltages. The charging processes are strongly influenced by CO₂ contamination, and exhibit increased overvoltages and increased capacity, as a result of poisoning of nucleation sites: this effect is predicted from DFT calculations and observed experimentally already at 1% CO₂. Large capacity losses and overvoltages are seen at higher CO₂ concentrations. © 2014 AIP Publishing LLC.

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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. References [1] M. Ender et al, Journal of The Electrochemical Society, 159 (7) A972-A980 (2012) [Formula]

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Fuel flow distribution in SOFC stacks revealed by impedance spectroscopy

As SOFC technology is moving closer to a commercial break through, methods to measure the “state-of-health” of operating stacks are becoming of increasing interest. This requires application of advanced methods for detailed electrical and electrochemical characterization during operation. An operating stack is subject to compositional gradients in the gaseous reactant streams, and temperature gradients across each cell and across the stack, which complicates detailed analysis.

An experimental stack with low ohmic resistance from Topsoe Fuel Cell A/S was characterized using Electrochemical Impedance Spectroscopy (EIS). The stack measurement geometry was optimized for EIS by careful selection of the placement of current feeds and voltage probes in order to minimize measurement errors.

It was demonstrated that with the improved placement of current feeds and voltage probes it is possible to separate the
loss contributions in an ohmic and a polarization part and that the low frequency response is useful in detecting mass transfer limitations. This methodology can be used to detect possible minor changes in the supply of gas to the individual cells, which is important when going to high fuel utilizations. The fuel flow distribution provides important information about the operating limits of the stack when high electrical efficiency is required.

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**Impedance of SOFC electrodes: A review and a comprehensive case study on the impedance of LSM:YSZ cathodes**

It was shown through a comprehensive impedance spectroscopy study that the impedance of the classic composite LSM:YSZ (lanthanum strontium manganite and yttria stabilized zirconia) solid oxide fuel cell (SOFC) cathode can be described well with porous electrode theory. Furthermore, it was illustrated through a literature review on SOFC electrodes that porous electrode theory not only describes the classic LSM:YSZ SOFC cathode well, but SOFC electrodes in general. The extensive impedance spectroscopy study of LSM:YSZ cathodes consisted of measurements on cathodes with three different sintering temperatures and hence different microstructures and varying degrees of LSM/YSZ solid state interactions. LSM based composite cathodes, where YSZ was replaced with CGO was also studied in order to acquire further knowledge on the chemical compatibility between LSM and YSZ. All impedance measurements were acquired in the very broad temperature range of 200–900°C for complete elucidation of the impedance. All impedance spectra were analyzed in terms of porous electrode theory. Physical materials parameters were extracted from the analysis, which were in excellent accordance with literature values. Valuable insight about the dissolution of Mn in the cathode composite material YSZ during preparation was furthermore provided along with valuable engineering characteristics such as the electrochemical utilization thickness. From the combined impedance study and literature review, it is clear, that porous electrode theory is the most suitable framework for any type of porous composite SOFC electrode evaluation.

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Impedance perspectives on Li-air battery overpotentials

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Abstract

Kinetic Studies on State of the Art Solid Oxide Cells
Introduction The Solid Oxide Fuel Cell (SOFC), which converts hydrogen as well as hydrocarbon fuels directly into electricity, has demonstrated almost comparable performance when operated reversely as Solid Oxide Electrolyser Cell (SOEC) for electrical energy storage as fuels. In both applications of the technology, cell optimization and eventual commercialisation requires a sound understanding of the mechanisms that affect performance and stability. These mechanisms depend on operation conditions like temperature, gas composition, fuel utilisation and current load as well as on gradients along cell and stack and cell microstructure. This increases the complexity of the systems, such that deconvolution and analytical description of the involved mechanisms becomes a major challenge, especially if both macroscopic trends as well as fundamental chemistry are to be accommodated. The electrode reaction kinetics is one such mechanism. Experimental The investigated single cells with a 4 x 4 cm² active electrode area are anode supported cells with Ni/3YSZ substrates (ca. 0.3 mm), Ni/8YSZ anodes (ca. 10 μm), 8YSZ electrolytes (ca. 10 μm), Ce0.9Gd0.1O2−δ buffer layers (ca. 1 μm) and La0.6Sr0.4CoO3−δ/Ce0.9Gd0.1O2−δ cathodes (ca. 30 μm). The cells were characterised through electrochemical impedance spectroscopy at open circuit voltage and under current load. Current/voltage characteristics were recorded at different temperatures and gas compositions. The fuel utilization and current density range were chosen to emulate practical conditions. Figure 1 displays the Distribution of Relaxation Times (DRT) of spectra recorded at varying current densities in SOFC mode at 700- and at 800°C ensuring a maximum fuel utilization of 60% in three different fuel electrode gas mixtures. Considering that the area beneath a peak represents the Area Specific Resistance (ASR) of the given dynamic process, it is clear that depending on operation temperature and fuel composition the processes make varying contributions to the overall cell resistance. Current vs. Overpotential Relation The ASRs of the processes were obtained through Complex Non-linear Least Squares fit of the spectra with an equivalent circuit model. The DRT was used for pre-identification of initial fit parameters. By integration of the ASRs vs current density, the corresponding overpotential curves were obtained, as displayed in Figure 2 for the electrochemical oxidation of H2 at the Ni/YSZ electrode. Figures 2 reveals that the overpotential associated with the electrochemical oxidation of H2 (and CO) at the Ni/YSZ electrode at all current densities increases from H2/H2O to H2/CO2 and CH4/H2/H2O fuel compositions. This is probably because in the reformate cases, H2 has to be replenished through the water gas shift reaction and also at equilibrium the H2/H2O case contains more H2O. The cooling effect of the endothermic steam reforming may contribute to a higher overpotential in CH4/H2/H2O mixture. Relative to the overall potential drop in the investigated current density range, it can be claimed that under these conditions and within the limits of accuracy, the Ni/YSZ electrode displays comparable performance in the three different fuel gas compositions. Outlook In this work experimental results of investigations on cells with varying microstructure of the fuel electrode operated in H2/H2O and reformate mixtures will be presented. Detailed impedance analysis will be used to determine the current/overpotential relationships and corresponding partial pressure dependencies. This will provide a solid basis for discussion of the highly debated analytical description of the electrochemical reaction overpotential of the Ni/YSZ electrodes. References 1. H. Schichlein A.C. Müller, M. Voigts, A. Krügel, and E. Ivers-Tiffée, J. Appl. Electrochem., 32, 875 (2002). 2. A. Kromp, A. Leonide, A. Weber, E. Ivers-Tiffée, J. Electrochem. Soc., 158 (8), B980-B986 (2011). [Formula]
Rheological analysis of stabilized cerium-gadolinium oxide (CGO) dispersions

The objective of the present work is to generate general rheological criteria to investigate high solid loading dispersions suitable for the shaping of homogeneous ceramic bodies. Systematic analysis of the rheological properties of moderately low specific surface area (SSA) Ce0.9Gd0.1O3-δ (CGO10) dispersions was performed in rotational and oscillatory modes. The dispersant content was optimized to attain fully stabilized dispersions. A critical upper limit for the ceramic content was introduced and denoted ϕh. It defines the limit to non-Newtonian flow and corresponds to the highest feasible volume fraction to which reproducible dispersions are achieved. The method proposed for its determination is based on the analysis of the flow index as function of the ceramic volume fraction. For the CGO dispersions formulated in this work, ϕh was found to be around (0.34 ± 0.04). The maximum volume fraction (ϕm) was also estimated and found to be (0.55 ± 0.01).
Solid Oxide Fuel Cell Stack Diagnostics

As SOFC technology is moving closer to a commercial breakthrough, methods to measure the “state-of-health” of operating stacks are becoming of increasing interest. This requires application of advanced methods for detailed electrical and electrochemical characterization during operation. An operating stack is subject to compositional gradients in the gaseous reactant streams, and temperature gradients across each cell and across the stack, which complicates detailed analysis. Several experimental stacks from Topsoe Fuel Cell A/S were characterized using Electrochemical Impedance Spectroscopy (EIS). The stack measurement geometry was optimized for EIS by careful selection of the placement of current feeds and voltage probes in order to minimize measurement errors. It was demonstrated that with the improved placement of current feeds and voltage probes it is possible to separate the loss contributions in an ohmic and a polarization part and that the low frequency response is useful in detecting mass transfer limitations. A sequential and a parallel measurement setup was developed for acquisition of impedance measurements. From the sequential to the parallel measurement setup the acquisition time was cut down significantly and it was demonstrated parallel acquisition of 16 repeating units (cells) and the total stack impedance could be made fully automated. The performance and degradation of a 13-cell cross-flow stack was monitored for more than 2500 hours at steady operating conditions using the sequential impedance measurement setup. Impedance measurements were used to examine the long-term behavior and monitor the
evolution of the series and polarization resistances for four out of the 13 repeating units during the first 1400 hours of operation. The losses for the four selected repeating units are reported and discussed. The performance and degradation of a 14-cell co-flow stack was monitored for more than 667 hours at steady operating conditions using the sequential impedance measurement setup. The stack was tested galvanostatically (at constant current) with 50% steam in the hydrogen fuel gas supplied to the stack. EIS was used to examine the long-term behavior and monitor the evolution of the impedance of each of the repeating units and the whole stack. The observed impedance was analyzed in detail for one of the repeating units and the whole stack and the losses reported and discussed in the following. Parallel acquisition using electrochemical impedance spectroscopy can be used to detect possible minor differences in the supply of gas to the individual cells, which is important when going to high fuel utilizations. The fuel flow distribution was determined and provides important information about the operating limits of the stack when high electrical efficiency is required. Furthermore, the measured gas concentration impedance was in between the impedances predicted by two different gas concentration impedance models. Total harmonic distortion, THD, experiments were carried out on an experimental 14-cell SOFC stack at varying frequencies and fuel utilizations. The results illustrated that THD can be used to detect increasing non-linearities in the current-voltage characteristics of the stack when the stack suffers from fuel starvation by monitoring the stack sum voltage only.

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Structural instability and electrical properties in epitaxial Er2O3-stabilized Bi2O3 thin films
Bismuth oxide based materials exhibit the highest oxygen ion conductivities, making them of great interest for use in energy conversion devices such as solid oxide fuel cells. However, these materials exhibit chemical and thermal instabilities and understanding and their stabilization is an actively pursued research goal. In this study, we investigate the structural and electrical properties of erbium oxide stabilized bismuth oxide (Er0.4Bi1.6O3 − δ) as thin films. These are deposited by pulsed laser deposition onto several single crystal substrates (MgO, Al2O3 and SrTiO3). The films show new forms of instabilities, both upon aging treatments in air and even under conductivity measurements, with remarkable changes in the film composition and microstructure. © 2014 Published by Elsevier B.V.

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Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 2.64 SJR 0.856 SNIP 0.952
Web of Science (2017): Impact factor 2.751
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 2.41 SJR 0.75 SNIP 0.909
Web of Science (2016): Impact factor 2.354
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 2.5 SJR 0.802 SNIP 1.016
Web of Science (2015): Impact factor 2.38
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 2.62 SJR 0.837 SNIP 1.282
Web of Science (2014): Impact factor 2.561
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 2.35 SJR 0.903 SNIP 1.269
Web of Science (2013): Impact factor 2.112
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 2.31 SJR 1.051 SNIP 1.253
Web of Science (2012): Impact factor 2.046
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 2.96 SJR 1.376 SNIP 1.615
Web of Science (2011): Impact factor 2.646
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 1.46 SNIP 1.498
Web of Science (2010): Impact factor 2.496
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 1.508 SNIP 1.483
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 1.515 SNIP 1.617
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 1.292 SNIP 1.384
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 1.239 SNIP 1.541
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 1.093 SNIP 1.423
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 1.18 SNIP 1.55
Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 1.473 SNIP 1.389
Web of Science (2003): Indexed yes
Scopus rating (2002): SJR 1.253 SNIP 1.36
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 1.121 SNIP 1.213
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 1.161 SNIP 1.312
Web of Science (2000): Indexed yes
Scopus rating (1999): SJR 1.08 SNIP 1.254

Original language: English
Sulfur Poisoning of SOFC Anodes: Effect of Overpotential on Long-Term Degradation

Sulfur impurities in carbon containing fuels for solid oxide fuel cells (SOFC), e.g. natural gas and biogas, typically lead to significant losses in performance due to the sulfur sensitivity of Ni/yttria-stabilized-zirconia (YSZ) anodes for SOFC. Full cells having Ni/YSZ anodes have been characterized during long-term galvanostatic operation in internal reforming gas mixture (\(\text{CH}_4/\text{H}_2\text{O}/\text{H}_2:30/60/10\)), with 2 ppm H2S exposure to the anode for 500 hours at 850°C, at different current densities. This work focuses on the long-term effect of H2S exposure over a few hundreds of hours; and describes and correlates the observed evolution of anode performance, over hundreds of hours, with sulfur exposure at low cell overpotential (low current density) and at high overpotential (high current density) with and without H2S exposure. For tests at low overpotential with H2S exposure only a reversible loss in performance was observed and post-mortem SEM analysis showed an intact Ni/YSZ anode microstructure. For tests at high cell overpotential the H2S exposure caused both a reversible loss in performance and an irreversible long-term degradation. Post-mortem SEM analysis of the Ni/YSZ anode from these tests showed increased porosity and lack of percolating Ni in the few microns of the anode closest to the anode/electrolyte interface.

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Contributors: Hauch, A., Hagen, A., Hjelm, J., Ramos, T.
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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
The performance and durability of solid oxide fuel cell (SOFC) cathodes consisting of a porous Ce\textsubscript{0.9}Gd\textsubscript{0.1}O\textsubscript{1.95} (CGO) infiltrated with nitrates corresponding to the nominal compositions La\textsubscript{0.6}Sr\textsubscript{0.4}Co\textsubscript{1.05}O\textsubscript{3-δ} (LSC), LaCoO\textsubscript{3-δ} (LC), and Co\textsubscript{3}O\textsubscript{4} are discussed. At 600°C, the polarization resistance, R\textsubscript{p}, varied as: LSC (0.062Ωcm\textsuperscript{2})<LC (0.079Ωcm\textsuperscript{2})<Co\textsubscript{3}O\textsubscript{4} (0.27Ωcm\textsuperscript{2}). High temperature X-ray diffraction revealed a number of different phases in LSC and LC. The electrochemical performance of the LSC-infiltrated CGO cathode was found to depend on the infiltrate firing temperature and is suggested to originate from a complex interplay between the formation, percolation, and surface area of
electronically conducting and catalytically active phases. Simplified models that predict the $R_p$ of LSC-infiltrated CGO were applied and showed that the performance is not only characterized by the nanoscale size of the infiltrate but also from a better surface exchange property. A 450h test of an LSC-infiltrated CGO cathode showed an $R_p$ with final degradation rate of only 11mΩcm$^{-2}$kh$^{-1}$. An SOFC with an LSC-infiltrated CGO cathode tested for 1,500h at 700°C and 0.5Acm$^{-2}$ (60% fuel, 20% air utilization) revealed no measurable degradation. © 2013 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

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Journal: Fuel Cells
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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
Web of Science (2011): Impact factor 3.149
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 1.623 SNIP 1.236
Electrochemical Characterization and Degradation Analysis of Large SOFC Stacks by Impedance Spectroscopy

As solid oxide fuel cell (SOFC) technology is moving closer to a commercial breakthrough through lifetime limiting factors, and methods to measure the “state-of-health” of operating cells and stacks are becoming of increasing interest. This requires application of advanced methods for detailed electrochemical characterization during operation.

An experimental stack with low ohmic resistance from Topsoe Fuel Cell A/S was characterized in detail using electrochemical impedance spectroscopy (EIS). An investigation of the optimal geometrical placement of the current feeds and voltage probes was carried out in order to minimize measurement errors caused by stray impedances. Three different stack geometries were investigated by impedance spectroscopy and the stack geometry with the minimum effect of stray impedances was selected.

A 13-cell experimental SOFC stack was tested during 2,500 h of operation with hydrogen as fuel with 52% fuel utilization and constant current load (0.2 A cm\(^{-2}\)) at 750 \(^\circ\)C. Stack interconnects were coated with six different coatings to prevent chromium poisoning on the cathode side. Four repeating units (RUs) with different coatings were selected for detailed impedance analysis. EIS allowed a distinction to be made in terms of the degradation between the four RU types that is not possible from IV-data only.

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Organisations: Department of Energy Conversion and Storage, Applied Electrochemistry, Mixed Conductors, Haldor Topsoe AS
Contributors: Mosbæk, R. R., Hjelm, J., Barfod, R., Høgh, J. V. T., Hendriksen, P. V.
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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
High Temperature and Pressure Alkaline Electrolysis

The production of energy from renewable sources has the possibility to fulfill the worldwide energy demand. Electricity generation from wind energy converters and photovoltaic systems will be implemented within the European Union to a large extent. The fluctuation of the wind speed and solar radiation raises the necessity to store the produced energy. Hydrogen production by water electrolysis is one of the most promising ways to do so. Alkaline electrolyzers have proven to operate reliably for decades on a large scale (up to 160 MW), but in order to become commercially attractive and compete against conventional technologies for hydrogen production, such as natural gas reforming, the production and investment costs have to be reduced. A reduction of the investment costs may be achieved by increasing the operational pressure and temperature of the electrolyzer, as this will result in: 1) production of pressurized hydrogen and oxygen, 2) improved electrical efficiencies and 3) increased current density, i.e. increased hydrogen production rate for a given electrolyser cell area. This thesis describes an exploratory technical study mainly in order to examine the possibility to produce hydrogen and oxygen with a new type of alkaline electrolysis cell at high temperatures and pressures. To perform measurements under high pressure and at elevated temperatures it was necessary to build a measurement system around an autoclave which could stand high temperatures up to 250 °C and pressures up to 200 bar as well as extremely caustic environments. Based on a literature study to identify resistant materials for these conditions, Inconel 600 was selected among the metals which are available for autoclave construction. An initial single atmosphere high temperature and pressure measurement setup was build comprising this autoclave. A second high temperature and pressure measurement setup was build based on experiences from the first setup in order to perform automated measurements. The conductivity of aqueous KOH at elevated temperatures and high concentrations was investigated using the van der Pauw method in combination with electrochemical impedance spectroscopy (EIS). Conductivity values as high as 2.7 S cm-1 for 35 wt%, 2.9 S cm-1 for 45 wt%, and 2.8 S cm-1 for 55 wt% concentrated aqueous solutions were measured at 200 °C. The conductivity of immobilized KOH was determined by the same method in the same temperature and concentration range. Conductivity values as high as 0.67 S cm-1 for 35 wt%, 0.84 S cm-1 for 45 wt%, and 0.73 S cm-1 for 55 wt% concentrated immobilized aqueous solutions were determined at 200 °C. A new type of an alkaline electrolysis cell was developed in order to operate at high temperatures and pressures. Aqueous potassium hydroxide immobilized electrolyte in porous SrTiO3 was used in those cells. Electrolysis cells with metal foam based gas diffusion electrodes and the immobilized electrolyte were successfully demonstrated at temperatures up to 250 °C and 40 bar. Different electro-catalysts were tested in order to reduce the oxygen and hydrogen overpotentials. Current densities of 1.1 A cm-2 and 2.3 A cm-2 have been measured at a cell voltage of 1.5 V and 1.75 V, respectively, without using expensive noble metal catalysts. Electrical efficiencies of almost 99 % at 1.1 A cm-2 and 85 % at 2.3 A cm-2 combined with relatively small production costs may lead to both reduced investment and operating costs for hydrogen and oxygen production. One of the produced electrolysis cells was operated for 350 h. Based on the successful results a patent application covering this novel cell was filed. Based on the successful results a patent application covering this novel cell was filed. Assuming that the developed cells will be scaled up and successfully tested for some thousand hours, they may offer an important role in future energy storage scenarios.

Investigation of novel solid oxide fuel cell cathodes based on impregnation of SrTixFe1-xO3-δ into ceria-based backbones

Solid oxide fuel cell (SOFC) cathodes were prepared by impregnating the nitrates corresponding to SrTixFe1-xO3-δ (STF), x = 0; 0.1; 0.2; 0.3; 0.4 and 0.5, into a porous backbone of Ce 0.9Gd0.1O2-δ (CGO). STF was chosen as very high oxygen surface exchange rate, high ionic conductivity and electrochemical stability as a thin film electrode have been reported for these materials. XRD measurements showed a high degree of secondary phase formation in the infiltrate as well as reaction with the CGO backbone. Microstructural analysis showed that the STF infiltrate had formed a coating on the CGO backbone. All prepared electrodes were characterized as symmetric cells using impedance spectroscopy. Within the investigated series the infiltrate with x = 0.1 (STF10) showed the best performance with an area specific resistance (ASR) of ASR ≈ 6.4 Ω cm2 (STF10) at 600°C in air. The relatively poor performance is believed to originate from poor electronic conduction in the electrodes and possibly also reactions between Sr-containing compounds and CGO. To circumvent the low electronic conductivity, backbones of a composite cathode containing LaCo0.4Ni0.6O 3-δ (LCN60) and CGO were also tried infiltrated with STF. The ASR of the backbone was 8.7 Ω cm2 prior to infiltration and this decreased
Rheology of stabilized cerium-gadolinium oxide (CGO) colloidal system
Achievement of stable dispersion with high solid loadings and low viscosity is crucial issue in ceramic films processing. In this work, systematic analysis of the rheological properties of CGO colloidal suspension was performed. The study aimed to define methods for evaluating fully stabilized conditions and critical parameters. The influences of dispersant and inorganic contents on suspensions properties were investigated. The optimization of dispersant content was achieved by studying flow behaviour of suspensions at different amounts of dispersant. Rotational and oscillatory tests were performed to study the inorganic loading impact on the suspension rheology. To identify a realistic load limit, the parameter $h$ was proposed. It defines the highest volume fraction to meet the processing requirements. A simple method for its determination was also proposed. The maximum solid volume fraction ($m$) was evaluated using a simple linear method. An extensive comparison with the commonly used non-linear physical models was performed.
The Influence of CO2 Poisoning on Overvoltages and Discharge Capacity in Non-aqueous Li-Air Batteries

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Electronic versions:
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Source-ID: u::9953
Research output: Research - peer-review › Conference article – Annual report year: 2013

Three-dimensional Nanofiber Cathode for Low Temperature and High Temperature Fuel Cells

General information
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Organisations: Department of Energy Conversion and Storage, Ceramic Engineering & Science, Imaging and Structural Analysis, Applied Electrochemistry, Proton conductors, Fundamental Electrochemistry, Technical University of Denmark
Contributors: Zhang, W. (., Zhang, W., Simonsen, S., Esposito, V., Hjelm, J., Ramousse, S., Jensen, J. O., Shao, J.
Number of pages: 1
Use of Intrinsic Viscosity for evaluation of polymer-solvent affinity

The objective of the current paper was to define a rheological method for the study of the solvent/binder affinity. The adopted strategy involves the study of the intrinsic viscosity [$\eta$] of polymer solutions. [$\eta$] was estimated via an extrapolation procedure using the Huggins and Kramer equations. The effects of chemical structure and molecular weight on the polymer/solvent affinity were investigated. Rheological properties of Ethyl Cellulose (EC), Polyvinyl Butyral (PVB) and Polyvinyl Acetal (PVAc) resins in a solution of a high boiling point glycol ether solvent were explored and compared. The intrinsic viscosity and the Mark-Houwink shape parameter were estimated for the three polymers and used as criteria for estimating the polymer/solvent affinity.

Detailed Electrochemical Characterisation of Large SOFC Stacks

As solid oxide fuel cell (SOFC) technology is moving closer to a commercial break through, lifetime limiting factors, determination of the limits of safe operation and methods to measure the “state-of-health” of operating cells and stacks are becoming of increasing interest. This requires application of advanced methods for detailed electrochemical characterisation during operation. An operating stack is subject to steep compositional gradients in the gaseous reactant streams, and significant temperature gradients across each cell and across the stack, which makes it a complex system to analyse in detail. Today one is forced to use mathematical modelling to extract information about existing gradients and cell resistances in operating stacks, as mature techniques for local probing are not available. This type of spatially resolved information is essential for model refinement and validation, and helps to further the technological stack development.

Further, more detailed information obtained from operating stacks is essential for developing appropriate process
monitoring and control protocols for stack and system developers. An experimental stack with low ohmic resistance from Topsoe Fuel Cell A/S was characterised in detail using electrochemical impedance spectroscopy. An investigation of the optimal geometrical placement of the current probes and voltage probes was carried out in order to minimise measurement errors caused by stray impedances. Unwanted stray impedances are particularly problematic at high frequencies. Stray impedances may be caused by mutual inductance and stray capacitance in the geometrical set-up and do not describe the fuel cell. Three different stack geometries were investigated by electrochemical impedance spectroscopy. Impedance measurements were carried out at a range of ac perturbation amplitudes in order to investigate linearity of the response and the signal-to-noise ratio. Separation of the measured impedance into series and polarisation resistances was possible.

**Durable and Robust Solid Oxide Fuel Cells**
The solid oxide fuel cell (SOFC) is an attractive technology for the generation of electricity with high efficiency and low emissions. Risø DTU (now DTU Energy Conversion) works closely together with Topsoe Fuel Cell A/S in their effort to bring competitive SOFC systems to the market. This 2-year project had as one of its’ overarching goals to improve durability and robustness of the Danish solid oxide fuel cells. The project focus was on cells and cell components suitable for SOFC operation in the temperature range 600 – 750 °C. The cells developed and/or studied in this project are intended for use within the CHP (Combined Heat and Power) market segment with stationary power plants in the range 1 – 250 kWe in mind. Lowered operation temperature is considered a good way to improve the stack durability since corrosion of the interconnect plates in a stack is lifetime limiting at T > 750 °C. The fact that degradation and robustness is not very well explored or understood at operating temperatures below 750 °C, provides motivation for focussing on materials and cells suitable for, and operated in this temperature range.

A significant part of this project was concerned with improved understanding of degradation and failure mechanisms. Improved understanding of performance and lifetime limiting factors will make it possible to develop strategies for countering degradation and improving the power density of SOFC based systems, both necessary to advance towards the goals set out in the national plan for SOFC implementation.
Electrochemical Switching of Conductance with Diarylethene-Based Redox-Active Polymers

Reversible switching of conductance using redox triggered switching of a polymer-modified electrode is demonstrated. A bifunctional monomer comprising a central electroswitchable core and two bithiophene units enables formation of a film through anodic electropolymerization. The conductivity of the polymer can be switched electrochemically in a reversible manner by redox triggered opening and closing of the diarylethene unit. In the closed state, the conductivity of the modified electrode is higher than in the open state.

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  BFI (2018): BFI-level 1
  Web of Science (2018): Indexed yes
  BFI (2017): BFI-level 1
  Scopus rating (2017): CiteScore 4.58 SJR 2.135 SNIP 1.147
  Web of Science (2017): Impact factor 4.484
  Web of Science (2017): Indexed yes
  BFI (2016): BFI-level 1
  Scopus rating (2016): CiteScore 4.48 SJR 1.964 SNIP 1.195
  Web of Science (2016): Impact factor 4.536
  Web of Science (2016): Indexed yes
  BFI (2015): BFI-level 1
  Scopus rating (2015): CiteScore 4.68 SJR 1.886 SNIP 1.26
  Web of Science (2015): Impact factor 4.509
  Web of Science (2015): Indexed yes
  BFI (2014): BFI-level 1
  Scopus rating (2014): CiteScore 5.08 SJR 2.032 SNIP 1.447
  Web of Science (2014): Impact factor 4.772
  Web of Science (2014): Indexed yes
  BFI (2013): BFI-level 1
  Scopus rating (2013): CiteScore 5.14 SJR 2.143 SNIP 1.445
  Web of Science (2013): Impact factor 4.835
  ISI indexed (2013): ISI indexed yes
  Web of Science (2013): Indexed yes
  BFI (2012): BFI-level 1
  Scopus rating (2012): CiteScore 4.98 SJR 2.529 SNIP 1.461
  Web of Science (2012): Impact factor 4.814
  ISI indexed (2012): ISI indexed yes
  Web of Science (2012): Indexed yes
  BFI (2011): BFI-level 1
  Scopus rating (2011): CiteScore 4.92 SJR 2.339 SNIP 1.465
  Web of Science (2011): Impact factor 4.805
  ISI indexed (2011): ISI indexed yes
  Web of Science (2011): Indexed yes
  BFI (2010): BFI-level 1
Highly durable anode supported solid oxide fuel cell with an infiltrated cathode

An anode supported solid oxide fuel cell with an La0.6Sr0.4Co1.05O3_δ (LSC) infiltrated-Ce0.9Gd0.1O1.95 (CGO) cathode that shows a stable performance has been developed. The cathode was prepared by screen printing a porous CGO backbone on top of a laminated and co-fired anode supported half cell, consisting of a Nieyttia stabilized zirconia (YSZ) anode support, a Niescandia-doped yttria-stabilized zirconia (ScYSZ) anode, a ScYSZ electrolyte, and a CGO barrier layer. LSC was introduced into the CGO backbone by multiple infiltrations of an aqueous nitrate solution followed by firing. The cell was tested at 700 deg. C under a current density of 0.5 A cm⁻² for 1500 h using air as oxidant and humidified hydrogen as fuel. The electrochemical performance of the cell was analyzed by impedance spectroscopy and current voltage relationships. No measurable degradation in the cell voltage or increase in the resistance from the recorded impedance was observed during long term testing. The power density reached 0.79Wcm⁻² at a cell voltage of 0.6 V at 750 deg. C. Post test analysis of the LSC infiltrated-CGO cathode by scanning electron microscopy revealed no significant micro-structural difference to that of a nominally identical untested counterpart.

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Web of Science (2017): Impact factor 6.945
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 6.22 SJR 1.944 SNIP 1.5
Original language: English
Keywords: Cathode, Infiltration, Solid oxide fuel cells, Mixed ionic and electronic conductor, Electrochemical impedance spectroscopy, Scanning electron microscopy
DOIs: 10.1016/j.jpowsour.2012.05.040
Source: dtu
Investigation of the degradation of LSM-YSZ SOFC cathode by electrochemical impedance spectroscopy

The aim of this PhD study was to investigate degradation of the LSM-YSZ cathode of anode supported Ni-YSZ/YSZ/LSM-YSZ solid oxide fuel cells. The chosen cathode materials LSM25 and 8YSZ were investigated for their compatibility and stability, to confirm that expansion/contraction or decreasing conductivity would not be a problem during degradation experiments of the cells. The experiments carried out for this purpose include x-ray diffraction, conductivity and dilatometry.

LSM-YSZ/YSZ/LSM symmetrical cells were prepared and investigated by means of electrochemical impedance spectroscopy, at different operating conditions. An equivalent circuit was developed for the symmetrical cell, describing the processes taking place at the LSM-YSZ cathode. This equivalent circuit was applied in degradation studies, where the processes affected by degradation over time could be pinpointed. Furthermore, it was discovered that impurities in air cause significant degradation of the cathode. Humidity was found to increase the degradation rate, but other impurities might also be present and increasing degradation.

Then the anode supported Ni-YSZ/YSZ/LSM-YSZ single cells were prepared and tested. It was found that at the applied operating conditions the impedance data could not be deconvoluted as anode and cathode processes were overlapping. Nonetheless it appeared that at OCV the degradation of the cathode is similar for symmetrical and single cells. Under current degradation was significantly lower, so real performance and degradation data can only be obtained on single cells as symmetrical cells can only be tested at OCV. For single cells degradation caused by impurities from air was also observed.

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Contributors: Torres da Silva, I. M., Mogensen, M. B., Hjelm, J.
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Research output: Research - peer-review › Journal article – Annual report year: 2012

A high performance ceria based interdiffusion barrier layer prepared by spin-coating

A multiple spin-coating deposition procedure of Ce0.9Gd0.1O1.95 (CGO) for application in solid oxide fuel cells (SOFCs) was developed. The thin and dense CGO layer can be employed as a barrier layer between yttria stabilised zirconia (YSZ) electrolyte and a (La, Sr)(Co, Fe)O3 based cathode. The decomposition of the polymer precursor used in the spin-coating process was studied. The depositions were performed on anode supported half cells. By controlling the sintering temperature between each spin-coating process, dense and crack-free CGO films with a thickness of approximately 1 μm were obtained. The successive steps of dense layer production was investigated by scanning electron microscopy. X-ray diffraction was employed to monitor the crystal structure of the CGO layer sintered at different temperatures. The described spin coated barrier layer was evaluated using an anode supported cell with a composite cathode made of La0.58Sr0.4Co0.2Fe0.8O3 (LSCF) and Ce0.9Gd0.1O1.95. The developed CGO layer, was sufficient to restrain reactions between the perovskite based electrode and the electrolyte.

General information
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Organisations: Electroceramics, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electrochemical Evaluation, Microstructures and Interfaces
Contributors: Plonczak, P., Joost, M., Hjelm, J., Søgaard, M., Lundberg, M., Hendriksen, P. V.
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Publication date: 2011
Peer-reviewed: Yes
Development of Long-Term Stable and High-Performing Metal-Supported SOFCs

Metal-supported SOFCs are believed to have high potential for commercialization due to lower material costs and higher robustness in fabrication and operation. However, the development of the cell is challenged by the metal properties during fabrication, and the necessary lower operating temperatures, while retaining both the energy output and the stability. The metal-supported SOFC design developed at Risø DTU has been optimized to an ASR value of 0.62 cm² at 650 °C, and a steady degradation rate of 1.0% kh⁻¹ demonstrated for 3000 h on a 16 cm² active cell level. Additional improvement in the performance has been demonstrated possible with cobalt-based cathode materials in combination with a magnetron sputtered CGO cathode barrier layer. Initial ASR values down to 0.27 cm² at 650 °C and power densities up to 1.14 W cm⁻² was observed with this design on a 0.5 cm² active cell level. ©2011 COPYRIGHT ECS - The Electrochemical Society

General information
State: Published
Organisations: Ceramic processing, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electrochemical Evaluation, Danish Technological Institute
Contributors: Klemensø, T., Nielsen, J., Blennow Tullmar, P., Persson, Å. H., Stegk, T., Hjalmarsson, P., Christensen, B., Senderby, S., Hjelm, J., Ramousse, S.
In recent years extended focus has been placed on monitoring and understanding degradation mechanisms in both solid oxide fuel cells and solid oxide electrolysis cells. The time-consuming nature of degradation experiments and the disparate conclusions from experiment reproductions indicates that not all degradation mechanisms are fully understood.

Traditionally, cell degradation has been attributed to the materials, processing and cell operating conditions. More recently, focus has been placed on the effect of raw material and gas impurities and their long-term effect on cell degradation. Minor impurities have been found to play a significant role in degradation and in some cases can overshadow the cell operation condition related degradation phenomenon. In this review, several degradation diagnostic tools are discussed, a benchmark for a desirable degradation rate is proposed and degradation behaviour and mechanisms are discussed. For ease of navigation, the review is separated into the various cell components – fuel electrode, electrolyte and oxygen electrode. Finally, nano-particle impregnate stability is discussed.
High performance anode supported SOFC produced by multilayer tape casting

General information
State: Published
Organisations: Ceramic processing, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electrochemical Evaluation
Contributors: Brodersen, K., Hauch, A., Hjalmarsson, P., Hjelm, J.
Publication date: 2011
Peer-reviewed: No
Electronic versions:
High performance anode.pdf
Source: orbit
Source-ID: 314319
Research output: Research › Conference abstract for conference – Annual report year: 2011

Manufacturing and characterization of metal-supported solid oxide fuel cells
A metal-supported solid oxide fuel cell design offers competitive advantages, for example reduced material costs and improved robustness. This paper reports the performance and stability of a recently developed metal-supported cell design, based on a novel cermet anode, on a 25cm2 (1cm2/16cm2 active area) cell level. An electrochemical performance comparable to state-of-the-art anode-supported cells is demonstrated. Detailed electrochemical analysis allowed assignment of the overall polarization losses quantitatively to gas diffusion in the metal support, electrooxidation in the anode functional layer, oxygen reduction in the mixed ionic-electronic conducting cathode and an additional polarization process with a rather high relaxation frequency, which may be assigned to an insulating corrosion interlayer. The durability of the cells was investigated by means of galvanostatic operation for periods of up to 1000h as well as the dynamic behavior, such as redox-, load- and thermal cycling tests. The galvanostatic stability tests indicated a fair, but significant degradation rate (~5% decrease in cell voltage/1000h at 650°C and 0.25Acm−2). Furthermore, the metal-supported cells underwent an endurance test of 100 redox cycles at 800°C without severe degradation nor total failure.

General information
State: Published
Planar metal-supported SOFC with novel cermet anode

Metal-supported solid oxide fuel cells are expected to offer several potential advantages over conventional anode (Ni-YSZ) supported cells. For example, increased resistance against mechanical and thermal stresses and a reduction in material costs. When Ni-YSZ based anodes are used in metal supported SOFC, elements from the active anode layer may inter-diffuse with the metallic support during sintering. This work illustrates how the inter-diffusion problem can be circumvented by using an alternative anode design based on porous and electronically conducting layers, into which electrocatalytically active materials are infiltrated after sintering. The paper presents the electrochemical performance and durability of the novel planar metal-supported SOFC design. The electrode performance on symmetrical cells has also been evaluated. The novel cell and anode design shows a promising performance and durability at a broad range of temperatures and is especially suitable for intermediate temperature operation at around 650°C. © 2011 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

General information
State: Published
Organisations: Ceramic processing, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electrochemical Evaluation, Electrochemistry
Contributors: Blennow Tullmar, P., Hjelm, J., Klemensø, T., Persson, Å. H., Ramousse, S., Mogensen, M. B.
Pages: 661-668
Publication date: 2011
Peer-reviewed: Yes

Publication information
Journal: Fuel Cells
Volume: 11
Issue number: 5
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
Technological implementation of LSC40 as porous cathode in SOFCs

General information
State: Published
Contributors: Hjalmarsson, P., Mosbæk, R. R., Hjelm, J.
Towards Quantification of Relations Between Electrode Polarisation and Microstructure

The total cell resistances of three Riso DTU solid oxide fuel cells (SOFCs) were broken down into individual contributions, by the combined use of electrochemical impedance spectroscopy (EIS), and complex non-linear least squares fitting (CNLS). Information on the number of contributing processes, frequency regions, and thermal behaviour, was obtained from symmetric cell studies of individual electrodes. The frequency regions dominated by electrode specific processes in the full cells were assessed by systematic changes of temperature and/or partial pressure of reactant gases on both electrodes. Based on the combined information, an equivalent circuit consisting of a series resistance (R-s) and five (RQ) elements was found suitable to describe the polarisation resistance of all tested cells. The breakdown process also allowed the assessment of limitations, and possible simplifications. The proposed electrochemical analysis correlated well with the exhibited microstructural features of the cells, which were produced with different compositions and/or production parameters. (C) 2011 The Electrochemical Society. [DOI: 10.1149/1.3587113] All rights reserved.
Cathode-Electrolyte Interfaces with CGO Barrier Layers in SOFC

Electron microscopy characterization across the cathode–electrolyte interface of two different types of intermediate temperature solid oxide fuel cells (IT-SOFC) is performed to understand the origin of the cell performance disparity. One IT-SOFC cell had a sprayed-cosintered Ce0.90Gd0.01O1.95 (CGO10) barrier layer, the other had a barrier layer deposited by pulsed laser deposition (PLD) CGO10. Scanning electron microscopy, transmission electron microscopy (TEM), and electron backscatter diffraction (EBSD) investigations conclude that the major source of the cell performance difference is attributed to CGO–YSZ interdiffusion in the sprayed-cosintered barrier layer. From TEM and EBSD work, a dense CGO10 PLD layer is found to be deposited epitaxially on the 8YSZ electrolyte substrate—permitting a small amount of SrZrO3 formation and minimizing CGO–YSZ interdiffusion.
Comparison of the Degradation of the Polariation Resistance of Symmetrical LSM-YSZ cells, with Anode Supported Ni-YSZ/YSZ/LSM-YSZ SOFCs

Impedance spectra of a symmetrical cell with SOFC cathodes (LSM-YSZ/YSZ/LSM-YSZ) and an anode supported planar SOFC (Ni-YSZ/YSZ/LSM-YSZ) were collected at OCV at 650\(^\circ\)C in air (cathode) and humidified (4\%) hydrogen (anode), over 155 hours. The impedance was affected by degradation over time in the same frequency range for both cells (~10 Hz), possibly indicating that the same physical process was affected in both types of cell. However, deconvolution of the impedance data was not straightforward. When n-values of the constant phase elements in the otherwise identical equivalent circuit for the full cell were changed slightly, very different results were obtained, showing the high sensitivity of the fitting procedure. Therefore, the equivalent circuit applied should be refined further, requiring comparison of impedance data of the cells at higher temperature (750, 850\(^\circ\)C).

General information
State: Published
Contributors: Torres da Silva, I. M., Nielsen, J., Hjelm, J., Mogensen, M. B.
Pages: 489-498
Publication date: 2009
Peer-reviewed: Yes

Publication information
Journal: ECS Transactions
Volume: 25
Issue number: 2
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
Comparison of the Degradation of the Polarization Resistance of Symmetrical LSM-YSZ Cells, with Anode Supported Ni-YSZ/YSZ/LSM-YSZ SOFCs

General information
State: Published
Contributors: Torres da Silva, I. M., Nielsen, J., Hjelm, J., Mogensen, M. B.
Pages: Abstract 1508
Publication date: 2009
Development of Planar Metal Supported SOFC with Novel Cermet Anode

General information
State: Published
Organisations: Ceramic processing, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electrochemical Evaluation, Microstructures and Interfaces, Electroceramics, Electrochemistry
Pages: Abstract 1441
Publication date: 2009

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

Bibliographical note
Copyright The Electrochemical Society, Inc. [2009]. All rights reserved. Except as provided under U.S. copyright law, this work may not be reproduced, resold, distributed, or modified without the express permission of The Electrochemical Society (ECS).
Source: orbit
Source-ID: 264296
Research output: Research - peer-review » Conference abstract in proceedings – Annual report year: 2009

Development of Planar Metal Supported SOFC with Novel Cermet Anode

Metal-supported solid oxide fuel cells are expected to offer several potential advantages over conventional anode (Ni-YSZ) supported cells, such as increased resistance against mechanical and thermal stresses and a reduction in materials cost. When Ni-YSZ based anodes are used in metal supported SOFC, electrode material from the active anode layer may interdiffuse with the metallic support during sintering. The purpose of this work is to illustrate how the interdiffusion problem can be circumvented by using an alternative anode design based on porous and electronically conducting layers, into which electrocatalytically active materials are infiltrated after sintering. The paper presents the recent results on the electrochemical performance and durability of the novel planar metal-supported SOFC design. The results presented in the paper show that the novel cell and anode design has a promising performance and durability at a broad range of temperatures and is especially suitable for intermediate temperature operation.
Electrochemical impedance spectroscopy as diagnostic tool

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Contributors: Jensen, S. H., Hjelm, J., Hagen, A., Mogensen, M. B.
Number of pages: 13
Publication date: 2009

Host publication information
Title of host publication: Handbook of Fuel Cells – Fundamentals, Technology and Applications
Volume: 5-6
Publisher: Wiley
Editors: Vielstich, W., Gasteiger, H., Yokokawa, H.
ISBN (Print): 978-0-470-72311-1
Keywords: Solid Oxide Fuel Cells, Fuel Cells and hydrogen
Source: orbit
Source-ID: 236770
Research output: Research - peer-review › Book chapter – Annual report year: 2009

Electro- and photochemical switching of dithienylethene self-assembled monolayers on gold electrodes

The photochemical and electrochemical properties of self-assembled monolayers (SAM) of three structurally distinct hexahydro- and hexafluoro-dithienylcyclopentene-based photochromic switches on gold electrodes are reported. The photochemical and electrochemical switching between the open and closed forms of the dithienylethene SAMs is examined and found to be sensitive to the molecular structure of the switch. For the three dithienylethenes, the electrochemical behavior with respect to electrochemical ring opening/closing is retained in the SAMs. In contrast, a marked dependence on the nature of the anchoring group is observed upon immobilization in terms of the retention of the photochemical properties observed in solution. For the meta-thiophenol anchor both photochemical ring opening and closing are observed in the SAM, while for the thienyl-thiol-anchored switches the photochemically properties are changed
markedly compared with those observed in solution. The stability of the monolayers toward desorption following photochemical and electrochemical switching is examined through electrochemistry and X-ray photoelectron spectroscopy.

**General information**

**State:** Published

**Organisations:** Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy

**Contributors:** Browne, W., Kudernac, T., Katsonis, N., Areephong, J., Hjelm, J., Feringa, B.

**Pages:** 1183-1190

**Publication date:** 2008

**Peer-reviewed:** Yes

**Publication information**

**Journal:** Journal of Physical Chemistry C

**Volume:** 112

**Issue number:** 4

**ISSN (Print):** 1932-7447

**Ratings:**

BFI (2018): BFI-level 1

Web of Science (2018): Indexed yes

BFI (2017): BFI-level 1

Scopus rating (2017): CiteScore 4.58 SJR 2.135 SNIP 1.147

Web of Science (2017): Impact factor 4.484

Web of Science (2017): Indexed yes

BFI (2016): BFI-level 1

Scopus rating (2016): CiteScore 4.48 SJR 1.964 SNIP 1.195

Web of Science (2016): Impact factor 4.536

Web of Science (2016): Indexed yes

BFI (2015): BFI-level 1

Scopus rating (2015): CiteScore 4.68 SJR 1.886 SNIP 1.26

Web of Science (2015): Impact factor 4.509

Web of Science (2015): Indexed yes

BFI (2014): BFI-level 1

Scopus rating (2014): CiteScore 5.08 SJR 2.032 SNIP 1.447

Web of Science (2014): Impact factor 4.772

Web of Science (2014): Indexed yes

BFI (2013): BFI-level 1

Scopus rating (2013): CiteScore 5.14 SJR 2.143 SNIP 1.445

Web of Science (2013): Impact factor 4.835

ISI indexed (2013): ISI indexed yes

Web of Science (2013): Indexed yes

BFI (2012): BFI-level 1

Scopus rating (2012): CiteScore 4.98 SJR 2.529 SNIP 1.461

Web of Science (2012): Impact factor 4.814

ISI indexed (2012): ISI indexed yes

Web of Science (2012): Indexed yes

BFI (2011): BFI-level 1

Scopus rating (2011): CiteScore 4.92 SJR 2.339 SNIP 1.465

Web of Science (2011): Impact factor 4.805

ISI indexed (2011): ISI indexed yes

Web of Science (2011): Indexed yes

BFI (2010): BFI-level 1

Scopus rating (2010): SJR 2.462 SNIP 1.362

Web of Science (2010): Impact factor 4.524

Web of Science (2010): Indexed yes

BFI (2009): BFI-level 1
Electrochemical Characterization of Planar Anode Supported SOFC with Strontium-Doped Lanthanum Cobalt Oxide Cathodes

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electroceramics
Contributors: Hjelm, J., Søgaard, M., Knibbe, R., Hagen, A., Mogensen, M. B.
Pages: 285-299
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
ISI indexed (2013): ISI indexed no
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 0.29 SJR 0.241 SNIP 0.26
ISI indexed (2012): ISI indexed no
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 0.36 SJR 0.261 SNIP 0.28
ISI indexed (2011): ISI indexed no
On/Off Photoswitching of the Electropolymerizability of Terthiophenes

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, University of Groningen
Contributors: Areephong, J., Kudernac, T., de Jong, J. J., Carroll, G. T., Pantorott, D., Hjelm, J., Browne, W. R., Feringa, B. L.
Pages: 12850-12851
Publication date: 2008
Peer-reviewed: Yes

Publication information
Journal: Journal of the American Chemical Society
Volume: 130
Issue number: 39
ISSN (Print): 0002-7863
Ratings:
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 14.05 SJR 8.127 SNIP 2.641
Web of Science (2017): Impact factor 14.357
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 13.18 SJR 7.492 SNIP 2.596
Web of Science (2016): Impact factor 13.858
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 12.81 SJR 6.775 SNIP 2.63
Web of Science (2015): Impact factor 13.038
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): CiteScore 11.92 SJR 6.294 SNIP 2.587
Web of Science (2014): Impact factor 12.113
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): CiteScore 11.38 SJR 5.993 SNIP 2.466
Web of Science (2013): Impact factor 11.444
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): CiteScore 10.37 SJR 6.211 SNIP 2.38
Web of Science (2012): Impact factor 10.677
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): CiteScore 9.94 SJR 5.478 SNIP 2.321
Web of Science (2011): Impact factor 9.907
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 2
Scopus rating (2010): SJR 5.167 SNIP 2.138
Web of Science (2010): Impact factor 9.023
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 2
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 2
Scopus rating (2008): SJR 5.06 SNIP 2.16
Web of Science (2008): Indexed yes
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 4.662 SNIP 2.252
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 4.413 SNIP 2.223
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 3.841 SNIP 2.203
Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 3.421 SNIP 2.236
Web of Science (2003): Indexed yes
Scopus rating (2002): SJR 3.223 SNIP 2.345
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 3.506 SNIP 2.15
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 3.972 SNIP 2.163
Web of Science (2000): Indexed yes
Scopus rating (1999): SJR 3.438 SNIP 2.133
Original language: English
DOIs:
10.1021/ja803714p
Source: orbit
Source-ID: 227866
Research output: Research - peer-review › Journal article – Annual report year: 2008

ORGN 495 - From solutions to surfaces: Exploring the effect of surface confinement on molecular photochemistry and conformational behavior

General information
State: Published
Organisations: Electrochemical Evaluation, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, University of Groningen
Contributors: Browne, W. R., Areephong, J., Katsonis, N., Hjelm, J., Feringa, B. L.
Pages: 495-ORGN
Publication date: 2008

Host publication information
Title of host publication: Abstracts of Papers of the American Chemical Society
Performance and Electrochemical Characterisation of Thin Electrolyte SOFCs

The performance and electrochemical behavior of two anode-supported thin electrolyte cells, with different manufacturing parameters, is determined by polarization measurements and electrochemical impedance spectroscopy (EIS). In addition to characterization, a previously suggested equivalent circuit consisting of a series resistance (Rs) and five arcs to describe the polarization resistance of the cell is tested. The aim is to breakdown and assign the losses to each individual cell component around open circuit voltage at selected temperatures. As will be seen, the proposed in-depth electrochemical characterization can provide valuable guidelines for production/performance optimization.

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Performance and Electrochemical Characterisation of Thin Electrolyte SOFCs

General information
State: Published
Contributors: Ramos, T., Hjelm, J., Wandel, M., Hagen, A., Mogensen, M. B.
Pages: Abstract 377
Publication date: 2008

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

Bibliographical note
Copyright The Electrochemical Society, Inc. [2008]. All rights reserved. Except as provided under U.S. copyright law, this work may not be reproduced, resold, distributed, or modified without the express permission of The Electrochemical Society (ECS).

Photochromism and electrochemistry of a dithienylcyclopentene electroactive polymer

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Pages: 6334-6342
Publication date: 2008
Peer-reviewed: Yes

Publication information
Journal: Langmuir
Volume: 24
Issue number: 12
ISSN (Print): 0743-7463
Ratings:
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 4 SJR 1.479 SNIP 1.148
Web of Science (2017): Impact factor 3.789
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 3.99 SJR 1.559 SNIP 1.178
Web of Science (2016): Impact factor 3.833
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 4.33 SJR 1.65 SNIP 1.281
SOFC research and development at Risø DTU

General information
State: Published
Organisations: Ceramic processing, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electrochemistry
Contributors: Menon, M., Hjelm, J.
Publication date: 2008
Peer-reviewed: No
Source: orbit
Source-ID: 232445
Research output: Research › Paper – Annual report year: 2008

Electrochemical Impedance Studies of SOFC Cathodes
Mixed ion- and electron-conducting composite electrodes consisting of doped ceria and perovskite have been studied by electrochemical impedance spectroscopy (EIS) at different temperatures and oxygen partial pressures. This paper aims to describe the different contributions to the polarisation impedance of the cathode at intermediate operating temperatures. The perovskite is of the La-Sr-Co-Fe type. The EIS response of symmetrical cells with a thick (similar to 200 µm) gadolinia doped ceria electrolyte was compared with the impedance contribution of the cathode of a full anode supported cell. The full cells had a Ni-YSZ anode and anode support, a thin YSZ electrolyte, and a CGO barrier layer. The symmetric and full cell cathode responses were compared at open-circuit voltage. Humidified hydrogen was used as the fuel in the full cell measurements. Differential analysis of the impedance data was used to identify frequency ranges where changes occur upon degradation and oxygen partial pressure variations.

General information
State: Published
Contributors: Hjelm, J., Søgaard, M., Wandel, M., Mogensen, M. B., Menon, M., Hagen, A.
Pages: 1261-1270
Publication date: 2007
Peer-reviewed: Yes

Publication information
Journal: E C S Transactions
Volume: 7
Issue number: 1
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
ISI indexed (2013): ISI indexed no
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 0.29 SJR 0.241 SNIP 0.26
ISI indexed (2012): ISI indexed no
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 0.36 SJR 0.261 SNIP 0.28
Synthesis and characterization of dinuclear ruthenium(II) complexes based on 4,4'-bipyridyl type gridding ligands

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Contributors: Guckian, A., Browne, W., Hjelm, J., Vos, J.
Pages: 1151-1161
Publication date: 2007
Peer-reviewed: Yes

Publication information
Journal: Journal of the Chinese Chemical Society
Volume: 54
ISSN (Print): 0009-4536
Ratings:
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 0.87 SJR 0.261 SNIP 0.323
Web of Science (2017): Impact factor 0.862
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 0.74 SJR 0.256 SNIP 0.266
Web of Science (2016): Impact factor 0.935
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 0.79 SJR 0.29 SNIP 0.398
Web of Science (2015): Impact factor 0.879
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 0.72 SJR 0.26 SNIP 0.409
Web of Science (2014): Impact factor 0.648
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 0.9 SJR 0.265 SNIP 0.455
Web of Science (2013): Impact factor 0.856
ISI indexed (2013): ISI indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 0.87 SJR 0.303 SNIP 0.544
Web of Science (2012): Impact factor 0.879
ISI indexed (2012): ISI indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 0.73 SJR 0.255 SNIP 0.374
Web of Science (2011): Impact factor 0.678
ISI indexed (2011): ISI indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 0.268 SNIP 0.414
Web of Science (2010): Impact factor 0.718
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 0.272 SNIP 0.348
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 0.312 SNIP 0.483
Scopus rating (2007): SJR 0.297 SNIP 0.55
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 0.249 SNIP 0.428
Scopus rating (2005): SJR 0.315 SNIP 0.445
Scopus rating (2004): SJR 0.266 SNIP 0.472
Scopus rating (2003): SJR 0.291 SNIP 0.429
Scopus rating (2002): SJR 0.321 SNIP 0.399
Scopus rating (2001): SJR 0.31 SNIP 0.39
Scopus rating (2000): SJR 0.186 SNIP 0.35
Scopus rating (1999): SJR 0.222 SNIP 0.287

Original language: English
Source: orbit
Source-ID: 223168
Research output: Research - peer-review › Journal article – Annual report year: 2007

Projects:

**Organic Redox Flow Batteries: Active Materials and Cell Stability**
Hoffmeyer, D., PhD Student, Department of Energy Conversion and Storage
Hjelm, J., Main Supervisor, Department of Energy Conversion and Storage
Vegge, T., Supervisor, Department of Energy Conversion and Storage
Samfinansieret - Andet
01/01/2018 → 31/12/2020
Award relations: Organic Redox Flow Batteries: Active Materials and Cell Stability
Project: PhD

**Electrochemical Characterization for Improvement of PEM Eletrolysers for Flexible Energy Storage**
Eisæ, K., PhD Student, Department of Energy Conversion and Storage
Mogensen, M. B., Main Supervisor, Department of Energy Conversion and Storage
Hjelm, J., Supervisor, Department of Energy Conversion and Storage
Hauch, A., Examiner, Department of Energy Conversion and Storage
Hauch, A., Examiner, Department of Energy Conversion and Storage
Sunde, S., Examiner
Sunde, S., Examiner
Forskningsrådsfinansiering
15/06/2015 → 14/06/2018
Award relations: Electrochemical Characterization for Improvement of PEM Eletrolysers for Flexible Energy Storage
Project: PhD
**In-operando spatially resolved probing of solid oxide electrolysis/fuel cells**  
Pitscheider, S., PhD Student, Department of Energy Conversion and Storage  
Chatzichristodoulou, C., Main Supervisor, Department of Energy Conversion and Storage  
Chueh, W. C., Supervisor  
Hansen, K. V., Supervisor, Department of Energy Conversion and Storage  
Hjelm, J., Supervisor, Department of Energy Conversion and Storage  
Hendriksen, P. V., Examiner, Department of Energy Conversion and Storage  
Fleig, J., Examiner  
Knudsen, J., Examiner  
Forskningsrådsfinansiering  
01/05/2015 → 16/08/2018  
Award relations: In-operando spatially resolved probing of solid oxide electrolysis/fuel cells  
Project: PhD

**Acid-base chemistry and HT-polymer electrolyte membranes**  
Becker, H., PhD Student, Department of Energy Conversion and Storage  
Li, Q., Main Supervisor, Department of Energy Conversion and Storage  
Jensen, J. O., Supervisor, Department of Energy Conversion and Storage  
Hjelm, J., Examiner, Department of Energy Conversion and Storage  
Büchi, F. N., Examiner  
Kallitsis, J., Examiner  
Forskningsrådsfinansiering  
15/12/2014 → 18/04/2018  
Award relations: Acid-base chemistry and HT-polymer electrolyte membranes  
Project: PhD

**In-operando localizied probing of solid oxide electrolysis/fuel cells by controlled atmosphere high temperature scanning probe microscopy**  
Kreka, K., PhD Student, Department of Energy Conversion and Storage  
Hansen, K. V., Main Supervisor, Department of Energy Conversion and Storage  
Chatzichristodoulou, C., Supervisor, Department of Energy Conversion and Storage  
Mogensen, M. B., Supervisor, Department of Energy Conversion and Storage  
Norman, K., Supervisor, Department of Energy Conversion and Storage  
Hjelm, J., Examiner, Department of Energy Conversion and Storage  
Bieberle-Hütter, A., Examiner  
Küngas, R., Examiner  
Forskningsrådsfinansiering  
15/03/2015 → 14/06/2018  
Award relations: In-operando localizied probing of solid oxide electrolysis/fuel cells by controlled atmosphere high temperature scanning probe microscopy  
Project: PhD

**Electron microscopy of electrochemical processes**  
Canepa, S., PhD Student, Department of Micro- and Nanotechnology  
Mølhave, K., Main Supervisor, Department of Micro- and Nanotechnology  
Sun, H., Supervisor, Department of Micro- and Nanotechnology  
Wagner, J. B., Supervisor  
Hjelm, J., Examiner  
Alloyeau, D., Examiner  
Dahl, S., Examiner  
Forskningsrådsfinansiering  
15/04/2014 → 23/08/2017  
Award relations: Electron microscopy of electrochemical processes  
Project: PhD

**In situ characterisation of structure and transport in battery electrolytes and electrodes**  
Sveinbjörnsson, D. P., PhD Student, Department of Energy Conversion and Storage  
Vegge, T., Main Supervisor, Department of Energy Conversion and Storage  
Mogensen, M. B., Supervisor, Department of Energy Conversion and Storage  
Norby, P., Supervisor, Department of Energy Conversion and Storage  
Hjelm, J., Examiner, Department of Energy Conversion and Storage  
David, W. I. F., Examiner  
Sørby, M. H., Examiner
Globaliseringsmidler
01/01/2011 → 26/02/2014
Award relations: In situ characterisation of structure and transport in battery electrolytes and electrodes
Project: PhD

Elektrode Kinetics and Gas Conversion in Solid Oxide Cells
Njodzefon, J., PhD Student, Department of Energy Conversion and Storage
Hjelm, J., Main Supervisor, Department of Energy Conversion and Storage
Graves, C. R., Supervisor, Department of Energy Conversion and Storage
Weber, A., Supervisor
Hauch, A., Examiner, Department of Energy Conversion and Storage
Krügel, A., Examiner
Scheinfeld, J., Examiner
Eksternt finansieret virksomhed
01/01/2014 → 20/04/2016
Award relations: Elektrode Kinetics and Gas Conversion in Solid Oxide Cells
Project: PhD

Investigating the Stability of Electrode-electrolyte-interfaces
Torres da Silva, I. M., PhD Student
Hjelm, J., Main Supervisor, Department of Energy Conversion and Storage
Mogensen, M. B., Supervisor, Department of Energy Conversion and Storage
Skaarup, S., Examiner
Skou, E. M., Examiner
Weber, A., Examiner
Forskningsrådsfinansiering
01/02/2008 → 08/02/2012
Award relations: Investigating the Stability of Electrode-electrolyte-interfaces
Project: PhD

Low Temperature NOx decomposition using an electrochemical Reactor
Shao, J., PhD Student, Department of Energy Conversion and Storage
Kammer Hansen, K., Main Supervisor, Department of Energy Conversion and Storage
Hjelm, J., Examiner, Department of Energy Conversion and Storage
Kustov, A., Examiner
Vernoux, P., Examiner
Forskningsrådsfinansiering
01/03/2010 → 19/08/2013
Award relations: Low Temperature NOx decomposition using an electrochemical Reactor
Project: PhD

Computional Investigations of transport mechanisms across battery interfaces
Loftager, S., PhD Student, Department of Energy Conversion and Storage
Veggge, T., Main Supervisor, Department of Energy Conversion and Storage
Garcia-Lastra, J. M., Supervisor
Hjelm, J., Examiner, Department of Energy Conversion and Storage
Khalifah, P., Examiner
Rossmeisl, J., Examiner
1/3 FUU, 1/3 inst 1/3 Andet
01/07/2013 → 16/11/2016
Award relations: Computional Investigations of transport mechanisms across battery interfaces
Project: PhD

High Temperature Alkaline Electrolyser Cell
Allebrod, F., PhD Student, Department of Energy Conversion and Storage
Mogensen, M. B., Main Supervisor, Department of Energy Conversion and Storage
Ebbesen, S. D., Supervisor, Department of Energy Conversion and Storage
Hjelm, J., Supervisor, Department of Energy Conversion and Storage
Holtappels, P., Examiner, Department of Energy Conversion and Storage
Skou, E. M., Examiner
Sunde, S., Examiner
Offentlig finansiering
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
Hagen, A., Examiner, Department of Energy Conversion and Storage
Adelhelm, P., Examiner
Edström, E. K., Examiner
Forskningsrådsfinansiering
01/12/2012 → 22/02/2016
Award relations: Investigation of performance and lifetime limiting effects in Li-air battery cells
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
Lindbergh, G., 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

Degradation mechanism of the LSCF Cathode
Zhang, W., PhD Student, Department of Energy Conversion and Storage
Chen, M., Main Supervisor, Department of Energy Conversion and Storage
Barfod, R. G., Supervisor
Hendriksen, P. V., Supervisor, Department of Energy Conversion and Storage
Hjelm, J., Examiner, Department of Energy Conversion and Storage
Halistedt, B., Examiner
Markus, T., Examiner
DTU, Samfinansiering
15/04/2010 → 19/03/2013
Award relations: Degradation mechanism of the LSCF Cathode
Project: PhD

SOFC Stack Diagnostics
Mosbæk, R. R., PhD Student, Department of Energy Conversion and Storage
Hjelm, J., Main Supervisor, Department of Energy Conversion and Storage
Barfod, R. G., Supervisor
Hendriksen, P. V., Supervisor, Department of Energy Conversion and Storage
Nielsen, J., Examiner, Department of Energy Conversion and Storage
Haart, L. G. J. D., Examiner
Weber, A., Examiner
Institut, samfinansiering
01/04/2011 → 04/02/2015
Award relations: SOFC Stack Diagnostics
Project: PhD

Lifetime limiting effects in pre-commercial solid cell devices
Skaffe, T. L., PhD Student, Department of Energy Conversion and Storage
Hjelm, J., Main Supervisor, Department of Energy Conversion and Storage
Blennow Tullmar, P., Supervisor, Department of Energy Conversion and Storage
Graves, C. R., Supervisor, Department of Energy Conversion and Storage
Hauch, A., Examiner, Department of Energy Conversion and Storage
Lanzini, A., Examiner
Weber, A., Examiner
Industrial PhD
01/03/2014 → 21/06/2017
Award relations: Lifetime limiting effects in pre-commercial solid cell devices
Project: PhD

Towards Smart Grid Ready SOFC
Hjelm, J., Project Coordinator, Department of Energy Conversion and Storage, Applied Electrochemistry
External Project ID: ForskEL 2012-1-10747
01/08/2012 → 31/07/2014
Keywords: solid oxide fuel cells, smart grid, reformate gas, electrochemistry, durability, load cycles, stacks
Collaborators: IRD Fuel Cells A/S, Haldor Topsoe AS
Project: Research

Durable and Robust SOFC
This 2-year project had as one of its' overarching goals to improve durability and robustness of the Danish solid oxide fuel cells. The project focus was on cells and cell components suitable for SOFC operation in the temperature range 600 – 750 °C. The cells developed and/or studied in this project are intended for use within the CHP (Combined Heat and Power) market segment with stationary power plants in the range 1 – 250 kW e in mind. A significant part of this project was concerned with improved understanding of degradation and failure mechanisms.
Hjelm, J., Project Coordinator, Department of Energy Conversion and Storage, Applied Electrochemistry
External Project ID: ForskEL 2010-1-10441
ForskEL (Energinet.dk): DKK11,000,000.00
01/07/2010 → 31/07/2012
Keywords: solid oxide fuel cells, electrochemistry, degradation, durability, lifetime limiting factors, porous electrode, functional ceramics, high temperature materials, hydrogen
Collaborators: IRD Fuel Cells A/S
Award relations: Durable and Robust SOFC
Project: Research