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 LiFePO_4/Graphite cylindrical cell. The ECM is based on measurements and modeling of impedance spectra recorded separately on cathode (LiFePO_4) and anode (Graphite) samples, harvested from the commercial cell. Modeling of the single-electrode impedance spectra provided information about the electronic and ionic resistance in the porous composite electrodes, as well as the solid state diffusion. Focused Ion Beam (FIB)/Scanning Electron Microscopy (SEM) of anode and cathode samples was used to make 3-D maps of the electrode microstructures and to obtain microstructural data for the ECM. The complementary analysis was crucial for the resolution of the single electrode impedance parameters and the proposal and validation of a new equivalent circuit used to model the full commercial
battery impedance.

**General information**

State: Published
Organisations: Department of Energy Conversion and Storage, Applied Electrochemistry, Imaging and Structural Analysis
Authors: Scipioni, R. (Intern), Jørgensen, P. S. (Intern), Graves, C. R. (Intern), Hjelm, J. (Intern), Jensen, S. H. (Intern)
Pages: A2017-A2030
Publication date: 2017
Main Research Area: Technical/natural sciences

**Publication information**

Journal: Electrochemical Society. Journal
Volume: 164
Issue number: 9
ISSN (Print): 0013-4651
Ratings:

BFI (2017): BFI-level 1
Web of Science (2017): Indexed Yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 2.97 SJR 1.134 SNIP 0.867
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 1.037 SNIP 1 CiteScore 3.17
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 1.147 SNIP 1.206 CiteScore 3.36
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 1.151 SNIP 1.299 CiteScore 2.92
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): SJR 1.329 SNIP 1.296 CiteScore 2.61
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): SJR 1.33 SNIP 1.345 CiteScore 2.74
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 1.417 SNIP 1.312
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 1.45 SNIP 1.267
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 1.608 SNIP 1.416
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 1.58 SNIP 1.325
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 1.611 SNIP 1.54
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 1.519 SNIP 1.484
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 1.719 SNIP 1.706
Web of Science (2004): Indexed yes
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.

General information

State: Published
Organisations: Mixed Conductors, Department of Energy Conversion and Storage, Brno University of Technology, Academy of Sciences of the Czech Republic, University of Modena and Reggio Emilia
Authors: Boccaccini, D. N. (Intern), Sevecek, O. (Ekstern), Frandsen, H. L. (Intern), Dlouhy, I. (Ekstern), Molin, S. (Intern), Charlas, B. (Intern), Hjelm, J. (Intern), Cannio, M. (Ekstern), Hendriksen, P. V. (Intern)
Pages: 3565-3578
Publication date: 2017
Main Research Area: Technical/natural sciences

Publication information
Journal: Journal of the European Ceramic Society
Volume: 37
Issue number: 11
ISSN (Print): 0955-2219
Ratings:
BFI (2017): BFI-level 1
Web of Science (2017): Indexed Yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 3.25 SJR 1.135 SNIP 1.776
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 1.15 SNIP 1.841 CiteScore 3.03
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 1.187 SNIP 2.099 CiteScore 3.16
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 1.122 SNIP 1.794 CiteScore 2.57
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 thermomechanical 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 cm−2, 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.

General information

State: Published
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) that 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 Li₂O₂ forms predominantly through either the surface- or solution-based mechanism.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Atomic scale modelling and materials, Applied Electrochemistry, University of California
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Number of pages: 7
Pages: A2065-A2071
Publication date: 2016
Main Research Area: Technical/natural sciences

Publication information
Journal: Journal of the Electrochemical Society
Volume: 163
Issue number: 9
ISSN (Print): 0013-4651
Ratings:
BFI (2017): BFI-level 1
Web of Science (2017): Indexed Yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 2.97 SJR 1.134 SNIP 0.867
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 1.037 SNIP 1 CiteScore 3.17
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 1.147 SNIP 1.206 CiteScore 3.36
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 1.151 SNIP 1.299 CiteScore 2.92
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): SJR 1.329 SNIP 1.296 CiteScore 2.61
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): SJR 1.33 SNIP 1.345 CiteScore 2.74
Electrochemical impedance spectroscopy, pressure change measurements, and scanning electron microscopy were used to investigate the nonequilibrium 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.
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.

General information
State: Published
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Publication date: 2016
Conference: PRiME 2016/230th ECS Meeting, Honolulu, United States, 02/10/2016 - 02/10/2016
Main Research Area: Technical/natural sciences

Publication information
Journal: Electrochemical Society. Meeting Abstracts (Online)
Volume: MA2016-02
Issue number: 38
Article number: 2408
ISSN (Print): 2151-2043
Original language: English
Links:
http://ma.ecsdl.org/content/MA2016-02/38/2408.abstract?sid=f6e5ab15-e4a2-45ba-82f8-177f51791c37
Source: FindIt
Source-ID: 2344805999
Publication: Research - peer-review › Conference abstract in journal – Annual report year: 2016

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 2015-1-12276 “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.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Mixed Conductors, Applied Electrochemistry
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Publication date: 2016
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.
Degradation mechanism, Focused ion beam scanning electron microscopy, Loss in electron percolation, Low accelerating voltage, Three-dimensional analysis of LiFePO4/Carbon electrode

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

General information
State: Published
Organisations: Mixed Conductors, Department of Energy Conversion and Storage, Applied Electrochemistry, Brno University of Technology, Academy of Sciences of the Czech Republic
Authors: Boccaccini, D. N. (Intern), Sevecisk, O. (Ekstern), Frandsen, H. L. (Intern), Dlouhy, I. (Ekstern), Molin, S. (Intern), Cannio, M. (Ekstern), Hjelm, J. (Intern), Hendriksen, P. V. (Intern)
Pages: 250–253
Publication date: 2016
Main Research Area: Technical/natural sciences

Publication information
Journal: Materials Letters
Volume: 162
ISSN (Print): 0167-577X
Ratings:
BFI (2017): BFI-level 1
Web of Science (2017): Indexed Yes
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.

General information
State: Published
Organisations: Applied Electrochemistry, Department of Energy Conversion and Storage, Fundamental Electrochemistry, Karlsruher Institut für Technologie
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Number of pages: 12
Pages: F1451-F1462
Publication date: 2016
Main Research Area: Technical/natural sciences
Performance and Lifetime Limiting Effects in Li-ion Batteries

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

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Applied Electrochemistry, Atomic scale modelling and materials
Authors: Scipioni, R. (Intern), Jensen, S. H. (Intern), Hjelm, J. (Intern), Norby, P. (Intern)
Number of pages: 203
Publication date: 2016

Publication information
Place of publication: Risø, Denmark
Publisher: Department of Energy Conversion and Storage, Technical University of Denmark
Original language: English
Main Research Area: Technical/natural sciences
Electronic versions:
Performance_and_Lifetime_Limiting_Effects_in_Li_Ion_Batteries.pdf
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.

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. LiPF$_6$ and LiClO$_4$ 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 (SXPES) 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 LiPF$_6$ 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

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 Li₂O₂, the Main Discharge Product in the Li-O₂ 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 battery, it is crucial to understand the fundamental mechanisms that govern and limit the system during electrochemical operation. Here we present a redox probing study of the charge transfer across the deposition product lithium peroxide, Li₂O₂, 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 Li₂O₂ 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 Li₂O₂ 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. McCloskey, 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 Li₂O₂ in Li-O₂ Batteries," J. Phys. Chem. Lett., vol. 4, pp. 3494-3499, 2013.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Fundamental Electrochemistry, Applied Electrochemistry, Technical University of Denmark
Authors: Knudsen, K. B. (Intern), Jensen, S. H. (Intern), Luntz, A. C. (Ekstern), Hjelm, J. (Intern)
Number of pages: 1
Pages: 20367-367
Publication date: 2015
Carbon Deposition during CO2 Electrolysis in Ni-Based Solid-Oxide-Cell Electrodes

Local gradients within an operating solid oxide electrolyzer cell were studied using current-potential measurements and electrochemical impedance spectroscopy. The cells and operating conditions were closely related to commercial applications. The cells with fuel electrodes of nickel and yttria-stabilized zirconia (Ni-YSZ) cermets were supplied by Haldor Topsoe A/S and the operating conditions were chosen so as to take stack relevant considerations into account.

Formation of carbon nanotubes in Ni containing fuel electrodes has previously been observed in co-electrolysis of H2O and CO2 [1]. Hence, 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 pCO/pCO2 ratios than that corresponding to the thermodynamic threshold for carbon formation. Electrochemical impedance spectroscopy in both H2/H2O and CO/CO2 revealed an increase in resistance of the fuel electrode after each CO2 electrolysis current-voltage curve, indicating possible carbon deposition. The difference in partial oxygen pressure between inlet and outlet was analyzed to verify carbon deposition. The increase in voltage is likely due to either blocking of the reaction sites by the deposited carbon, or due to the microstructural damage caused by the carbon, or a combination of these.

Moreover, the current step-size and step-length was varied to investigate the time-dependence of the detection of carbon deposition. Initial results indicate that for longer current steps, carbon formation is observed at lower pCO/pCO2 ratios. This is related to the rate of carbon deposition and the averaged nature of the cell voltage measurements. Possible 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.
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.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Applied Electrochemistry, Haldor Topsoe AS
Authors: Skafte, T. L. (Intern), Graves, C. R. (Intern), Blennow, P. (Ekstern), Hjelm, J. (Intern)
Number of pages: 9
Pages: 3429-3437
Publication date: 2015
Main Research Area: Technical/natural sciences

Publication Information
Journal: E C S Transactions
Volume: 68
Issue number: 1
ISSN (Print): 1938-5862
Ratings:
BFI (2017): BFI-level 1
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 0.4 SJR 0.231 SNIP 0.246
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 0.214 SNIP 0.257 CiteScore 0.36
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 0.214 SNIP 0.246 CiteScore 0.36
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 0.192 SNIP 0.237 CiteScore 0.27
ISI indexed (2013): ISI indexed no
BFI (2012): BFI-level 1
Scopus rating (2012): SJR 0.24 SNIP 0.263 CiteScore 0.29
ISI indexed (2012): ISI indexed no
BFI (2011): BFI-level 1
Scopus rating (2011): SJR 0.262 SNIP 0.284 CiteScore 0.36
ISI indexed (2011): ISI indexed no
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 0.247 SNIP 0.245
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 0.241 SNIP 0.266
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 0.253 SNIP 0.25
Scopus rating (2007): SJR 0.213 SNIP 0.206
Scopus rating (2006): SJR 0.135 SNIP 0.062
Original language: English
DOIs:
10.1149/06801.3429ecst
Source: FindIt
Source-ID: 2279821838
Publication: Research - peer-review › Conference article – Annual report year: 2015
Degradation Studies on LiFePO$_4$ cathode

In this paper we examine a laboratory LiFePO$_4$ (LFP) cathode and propose a simple model that predicts the electrode capacity as function of C-rate, number of cycles and calendar time. Microcracks were found in Li$_{1-x}$FePO$_4$ particles in a degraded LFP electrode and low-acceleration voltage (1 kV) FIB/SEM analysis allowed us to obtain phase contrast between FePO$_4$ and LiFePO$_4$. The evolution of micro-cracks is expected to increase the concentration of LFP particles which are not electronically accessible and thus cause a loss in capacity.
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/O2 fuel mixture, the Ni/8 mol % yttria-stabilized zirconia (Ni/YSZ) fuel electrode showed slower reaction kinetics operating under cathodic polarization than anodic—the same finding had been reported in literature from investigations on full cells whereby 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/Ce0.9Gd0.101.95 (LSC/CGO) 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 (LSCF) 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-, electrolysise-, 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 LSC/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.

Enhancement of the chemical stability in confined δ-Bi₂O₃
Bismuth-oxide-based materials are the building blocks for modern ferroelectrics1, multiferroics2, gas sensors3, light photocatalysts4, and fuel cells5,6. Although the cubic fluorite δ-phase of bismuth oxide (δ-Bi2O3) exhibits the highest conductivity of known solid-state oxygen ion conductors5, its instability prevents use at low temperature7–10. 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 gastight 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.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Ceramic Engineering & Science, Applied Electrochemistry, Fundamental Electrochemistry, Mixed Conductors
Pages: 89-95
Publication date: 2015
Main Research Area: Technical/natural sciences

Publication information
Journal: Journal of Power Sources
Volume: 273
ISSN (Print): 0378-7753
Ratings:
BFI (2017): BFI-level 1
Web of Science (2017): Indexed Yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 6.22 SJR 1.945 SNIP 1.483
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 1.945 SNIP 1.686 CiteScore 6.34
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 1.983 SNIP 2.071 CiteScore 6.3
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 1.985 SNIP 2.138 CiteScore 5.63
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): SJR 2.293 SNIP 2.016 CiteScore 5.04
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): SJR 2.247 SNIP 2.181 CiteScore 5.13
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 2.297 SNIP 1.981
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 2.117 SNIP 1.793
Web of Science (2009):Indexed yes
BFI (2008): BFI-level 2
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.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Ceramic Engineering & Science, Applied Electrochemistry, Mixed Conductors, Topsoe Fuel Cell
Number of pages: 10
Pages: 1495–1504
Publication date: 2015
Main Research Area: Technical/natural sciences

Publication information
Journal: European Ceramic Society. Journal
Volume: 35
Issue number: 5
ISSN (Print): 0955-2219
Ratings:
BFI (2017): BFI-level 1
Web of Science (2017): Indexed Yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 3.25 SJR 1.135 SNIP 1.776
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 1.15 SNIP 1.841 CiteScore 3.03
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 1.187 SNIP 2.099 CiteScore 3.16
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 sometimes 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\textsuperscript{1}. 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/cm\textsuperscript{2} in cathodic polarization, an overpotential of ca. 150 mV is required, compared with 100 mV in anodic polarization. This reflects asymmetry\textsuperscript{2}-\textsuperscript{6}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\textsuperscript{7}. 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. Skafte, 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. Holtappels, L. G. J. de Haart, and U. Stimming, J. Electrochem. Soc., 146, 1620–1625 (1999). 6. J.-C. Njodzefon, D. Klotz, A. Kromp, 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 Ni-YSZ Composite Electrodes

AC and DC techniques were applied to investigate the electrochemical reaction kinetics of porous composite Ni/8-mol% yttria-stabilized zirconia (Ni/YSZ) solid oxide cell (SOC) electrodes using a novel pseudo-3-electrode cell geometry. From OCV impedance spectra an activation energy Ea of 1.13 eV, prefactor yan of 3.7·10\textsuperscript{5}·T, hydrogen and steam partial pressure dependencies a and b respectively of -0.07 and 0.22 were determined. DC current density vs. overpotential curves compared with those predicted using the determined kinetic parameters. Apparent Butler-Volmer charge transfer coefficients α were determined from the current density vs. overpotential curves. Values ranging from 0.57 at 650°C to 0.64 at 850°C were determined from the anodic branch and 0.85 to 0.81 from the cathodic branch in the same range, with higher fitting accuracy in the anodic branch. The lower fitting accuracy of the cathodic branch and the need for different α values for each branch suggests that a simple BV model of the measured electrode kinetics is insufficient and/or different reaction mechanisms might be occurring in anodic vs cathodic polarization.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Applied Electrochemistry, Ceramic Engineering & Science
Authors: Njodzefon, J. (Intern), Hjelm, J. (Intern), Graves, C. R. (Intern), Sudireddy, B. R. (Intern)
Publication date: 2015
Main Research Area: Technical/natural sciences

Publication information
Journal: ECS Transactions
Volume: 68
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.

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.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Applied Electrochemistry, Fundamental Electrochemistry, Karlsruhe Institute of Technology
Authors: Njodzefon, J. (Intern), Graves, C. R. (Intern), Mogensen, M. B. (Intern), Weber, A. (Ekstern), Hjelm, J. (Intern)
Pages: 51-65
Publication date: 2015
Conference: 226th Meeting of the Electrochemical Society (ECS) and 7th Meeting of the Mexico Section of the Electrochemical Society ECS and SMEQ Joint International Meeting, Cancun, Mexico, 05/10/2014 - 05/10/2014
Main Research Area: Technical/natural sciences

Publication information
Journal: ECS Transactions
Volume: 64
Issue number: 18
ISSN (Print): 1938-5862
Low - voltage FIB/SEM Tomography for 3D Microstructure Evolution of LiFePO4/C Electrode

Li-ion batteries find widespread use in many electricity storage applications, from portable devices to electric vehicles [1-3], and LiFePO4 (LFP) is one of the most common cathodes because of its long durability and high safety [4, 5] but, since its low ionic and electronic conduction, it is always mixed with carbon black (CB) additives to increase electronic percolation in the electrode. Focused Ion Beam (FIB)/Scanning Electron Microscopy (SEM) Tomography is one of the most used techniques for the study of the three-dimensional microstructure of porous electrodes [6-8]. Imaging at low-kV has been shown to be an excellent technique for studying electron percolation in Ni-network in solid oxide fuel cells [9]. In this work we study the degradation process that occurs in a LFP/C electrode by Low-kV FIB/SEM Tomography, using the low-voltage percolation technique to identify compositional changes in the CB network in three-dimensions. FIB/SEM images of a fresh and degraded cathode are compared and LFP grains are seen to crack with cycling, resulting in the formation of secondary disconnected particles with increased ionic resistivity; CB particles are instead observed to agglomerate, reducing the electrochemically active surface area. Using low voltage imaging (1 kV) a significant fraction of the large carbon agglomerates found in the aged electrode show a higher secondary electron yield compared to the fresh CB particles at low accelerating voltage. This suggests that degradation occurs both due to morphological changes and due to amorphous-crystalline phase transitions in the carbon network, resulting in non-percolating CB agglomerates. Figure 1 shows lateral Everhart – Thornley (E-T) and in-lens detector images of fresh (a, b) and degraded cathode (c, d) collected after FIB slicing. White grains are LFP, black particles are CB additive and grey regions are pores infiltrated with silicon resin to improve phase contrast [7]. The CB network appears entirely dark in the in-lens detector image of the fresh electrode (Fig. 1b). It is possible to notice some charging effects from the insulating silicon resin. The in-lens detector image of the degraded electrode (Fig. 1d) is instead characterized by the presence of big carbon agglomerates (red rings) which are brighter because they charge as the electron beam hit them. This indicates a lower electric conductivity. 3D reconstruction of the entire network (Fig. 2) revealed that the carbon phase with lower electric conductivity accounted for approximately 25% of the volume of the total carbon in the sample. This of course affects electrode capacity since a reduced electron percolation in the CB network impedes (de)lithiation process of LFP particles. References - M. Armand, J. M. Tarascon, Nature, 451, 652-657 (2008). - B. Scrosati, J. Hassoun, and Y.K. Sun, Energy Environ. Sci., 4, 3287-3295 (2011). - J.M. Tarascon, M. Armand, Nature, 414, 359-367 (2001). - A.K. Padhi, K.S. Nanjundaswamy, and J. B. Goodenough, J. Electrochem. Soc., 144(4), 1188-1194 (1997). - Y. Wang, P. He, and H. Zhou, Energy Environ. Sci., 4, 805-817 (2010). - T. Hutzenlaub et al., Electrochemical and Solid-State Letters, 15 (3), A33-A36 (2012). - M. Ender et al, J. Electrochem. Soc., 159(7), A972-A980 (2012). - Z. Liu et al., J. Power Sources, 227, 267-274 (2013). - K. Thydén, Y.L. Liu, and J.B. Bilde-Sørensen, Solid State Ionics, 178, 1984-1989 (2008).
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.
In the field of energy storage devices the pursuit for cheap, high energy density, reliable secondary batteries is at the top of the agenda. The Li−O2 battery is one of the possible technologies that, in theory, should be able to close the gap, which exists between the present state-of-the-art Liion technologies and the demand placed on batteries by technologies such as electrical vehicles. Here we present a redox probing study of the charge transfer across the main deposition product lithium peroxide, Li2O2, in the Li−O2 battery using outer-sphere redox shuttles. The change in heterogeneous electron transfer exchange rate as a function of the potential and the Li2O2 layer thickness (∼depth-of-discharge) was determined using electrochemical impedance spectroscopy. The attenuation of the electron transfer exchange rate with film thickness is dependent on the probing potential, providing evidence that hole transport is the dominant process for charge transfer through Li2O2 and showing that the origin of the sudden death observed upon discharge is due to charge transport limitations.
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.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Applied Electrochemistry
Authors: Graves, C. R. (Intern), Hjelm, J. (Intern)
Number of pages: 16
Publication date: 2014
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 (1̅100) 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.

General information
State: Published
Organisations: Center for Atomic-scale Materials Design, Department of Energy Conversion and Storage, Atomic scale modelling and materials, Department of Physics, Fundamental Electrochemistry, Applied Electrochemistry
Authors: Mekonnen, Y. S. (Intern), Knudsen, K. B. (Intern), Mýrdal, J. S. G. (Intern), Younesi, R. (Intern), Højberg, J. (Intern), Hjelm, J. (Intern), Norby, P. (Intern), Vegge, T. (Intern)
Number of pages: 5
Publication date: 2014
Main Research Area: Technical/natural sciences

Publication information
Journal: Journal of Chemical Physics
Volume: 140
Article number: 121101
ISSN (Print): 0021-9606
Ratings:
BFI (2017): BFI-level 2
Web of Science (2017): Indexed Yes
BFI (2016): BFI-level 2
Scopus rating (2016): SJR 1.073 SNIP 0.755 CiteScore 2.13
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): SJR 0.953 SNIP 0.767 CiteScore 1.98
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): SJR 1.386 SNIP 0.989 CiteScore 2.54
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): SJR 1.532 SNIP 1.17 CiteScore 2.95
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): SJR 1.787 SNIP 1.118 CiteScore 2.86
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
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.
Fuel flow distribution in SOFC stacks revealed by impedance spectroscopy

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.

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.

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.
Impedance perspectives on Li-air battery overpotentials

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)1 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. (f > 103Hz in Figure 1) Figure 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 though 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

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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 $\phi_h$. 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, $\phi_h$ was found to be around (0.34 ± 0.04). The maximum volume fraction (\(\phi_m\)) was also estimated and found to be (0.55 ± 0.01).
Ceramic dispersion, Cerium-gadolinium oxide, Rheological properties, Flow behaviour

DOIs:
10.1016/j.jeurceramsoc.2013.09.022
Solid Oxide Fuel Cell Stack Diagnostics
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. 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 that 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 was 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.

Structural instability and electrical properties in epitaxial Er$_2$O$_3$-stabilized Bi$_2$O$_3$ 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 (Er$_{0.4}$Bi$_{1.6}$O$_3$ − δ) as thin films. These are deposited by pulsed laser deposition onto several single crystal substrates (MgO, Al$_2$O$_3$ and SrTiO$_3$). 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.
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 (CH4/H2O/H2:30/60/10), with 2 ppm H2S exposure to the anode for 500 hours at 850°C, at different current densities. This work focus 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 NiYSZ anode from this tests showed increased porosity and lack of percolating Ni in the few microns of the anode closest to the anode/electrolyte interface.
The performance and durability of solid oxide fuel cell (SOFC) cathodes consisting of a porous Ce$_{0.9}$Gd$_{0.1}$O$_{1.95}$ (CGO) infiltrated with nitrates corresponding to the nominal compositions La$_{0.6}$Sr$_{0.4}$Co$_{1.05}$O$_{3-δ}$ (LSC), LaCoO$_{3-δ}$ (LC), and Co$_3$O$_4$ are discussed. At 600°C, the polarization resistance, $R_p$, varied as: LSC ($0.062\Omega\text{cm}^2$)<LC ($0.079\Omega\text{cm}^2$)<Co$_3$O$_4$ ($0.27\Omega\text{cm}^2$). 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 $11\text{nΩcm}^2\text{h}^{-1}$. 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.
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, 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 °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.

General information

State:Published
Organisations:Department of Energy Conversion and Storage, Applied Electrochemistry, Mixed Conductors, Topsoe Fuel Cell
Authors: Mosbæk, R. R. (Intern), Hjelm, J. (Intern), Barfod, R. (Ekstern), Høgh, J. V. T. (Intern), Hendriksen, P. V. (Intern)
Pages:605-611
Publication date:2013
Conference:10th European SOFC Forum, Luzern, Switzerland, 26/06/2012 - 26/06/2012
Main Research Area:Technical/natural sciences

Publication information

Journal:Fuel Cells
Volume:13
Issue number:4
ISSN (Print):1615-6846
Ratings:
BFI (2017): BFI-level 1
Web of Science (2017): Indexed Yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 1.79 SJR 0.498 SNIP 0.62
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 0.699 SNIP 0.787 CiteScore 2.02
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 0.629 SNIP 0.816 CiteScore 2.05
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 0.841 SNIP 0.848 CiteScore 1.99
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): SJR 1.25 SNIP 1.008 CiteScore 2.76
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): SJR 1.656 SNIP 1.238 CiteScore 3.31
Degradation, Electrochemical Impedance Spectroscopy, Solid Oxide Fuel Cell (SOFC), Stack

ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 1.632 SNIP 1.243
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 1.368 SNIP 1.12
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 1.523 SNIP 1.226
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 1.349 SNIP 1.086
Scopus rating (2006): SJR 1.194 SNIP 1.228
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 0.45 SNIP 0.501
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 0.232 SNIP 0.215
Web of Science (2004): Indexed yes
Web of Science (2001): Indexed yes
Original language: English

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 reliable 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 automatized 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 electrocatalysts 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 to 0.34 Ω cm² after infiltration with STF10.

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.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Ceramic Engineering & Science, Applied Electrochemistry
Authors: Marani, D. (Intern), Hjelm, J. (Intern), Wandel, M. (Intern)
Publication date: 2013
Event: Abstract from 8th Annual European Rheology Conference, Leuven, Belgium.
Main Research Area: Technical/natural sciences
Source: PublicationPreSubmission
Source-ID: 106753956
Publication: Research - peer-review › Conference abstract for conference – Annual report year: 2013

Sulfur Poisoning of Ni/stabilized-zirconia Anodes – Effect on Long-Term Durability
Sulfur impurities in carbon containing fuels for solid oxide fuel cells (SOFC), e.g. natural gas and biogas, can lead to significant losses in performance due to the sulfur sensitivity of Ni/YSZ SOFC anodes. Full cells having Ni/YSZ and Ni/ScYSZ anodes have been characterized during long-term galvanostatic operation in internal reforming gas mixture (CH4/H2O/H2:30/60/10), with 2 ppm H2S exposure to the anode, at different current densities. The aim was not only to investigate the well-known initial performance drop associated with adsorbed sulfur in the Ni/stabilized-zirconia anodes, but also to focus on the long-term effect over a few hundred of hours. This work describes and correlates the observed evolution of anode performance, over hundreds of hours, with sulfur poisoning with the different operating conditions.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Applied Electrochemistry, Fundamental Electrochemistry
Authors: Hauch, A. (Intern), Hagen, A. (Intern), Hjelm, J. (Intern), Ramos, T. (Intern)
Pages: 615-625
Publication date: 2013
Conference: 13th International Symposium on Solid Oxide Fuel Cells (SOFC-XIII), Okinawa, Japan, 06/10/2013 - 06/10/2013
Main Research Area: Technical/natural sciences

Publication information
Journal: E C S Transactions
Volume: 57
Issue number: 1
ISSN (Print): 1938-5862
Ratings:
BFI (2017): BFI-level 1
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 0.4 SJR 0.231 SNIP 0.246
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 0.214 SNIP 0.257 CiteScore 0.36
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 0.214 SNIP 0.246 CiteScore 0.36
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 0.192 SNIP 0.237 CiteScore 0.27
ISI indexed (2013): ISI indexed no
BFI (2012): BFI-level 1
Scopus rating (2012): SJR 0.24 SNIP 0.263 CiteScore 0.29
ISI indexed (2012): ISI indexed no
BFI (2011): BFI-level 1
Scopus rating (2011): SJR 0.262 SNIP 0.284 CiteScore 0.36
ISI indexed (2011): ISI indexed no
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 0.247 SNIP 0.245
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 0.241 SNIP 0.266
The Influence of CO2 Poisoning on Overvoltages and Discharge Capacity in Non-aqueous Li-Air Batteries

General information
State: Published
Organisations: Center for Atomic-scale Materials Design, Department of Energy Conversion and Storage, Atomic scale modelling and materials, Department of Physics, Fundamental Electrochemistry, Applied Electrochemistry
Authors: Mekonnen, Y. S. (Intern), Knudsen, K. B. (Intern), Mýrdal, J. S. G. (Intern), Younesi, R. (Intern), Højberg, J. (Intern), Hjelm, J. (Intern), Norby, P. (Intern), Vegge, T. (Intern)
Number of pages: 1
Publication date: 2013
Main Research Area: Technical/natural sciences
Electronic versions:
The_Influence_of_CO2_Poisoning.pdf
Source: dtu
Source-ID: u::9953
Publication: Research › Poster – Annual report year: 2013

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

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Ceramic Engineering & Science, Imaging and Structural Analysis, Applied Electrochemistry, Proton conductors, Fundamental Electrochemistry, Technical University of Denmark
Authors: Zhang, W. (. (Intern), Zhang, W. (Intern), Simonsen, S. (Ekstern), Esposito, V. (Intern), Hjelm, J. (Intern), Ramousse, S. (Intern), Jensen, J. O. (Intern), Shao, J. (Intern)
Number of pages: 1
Publication date: 2013
Event: Abstract from Annual meeting of the Danish Electrochemical Society, Ringsted, Denmark.
Main Research Area: Technical/natural sciences
Electronic versions:
Three_dimensional_Nanofiber_Cathode.pdf
Source: dtu
Source-ID: u::10495
Publication: Research › Conference abstract for conference – Annual report year: 2014

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.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Ceramic Engineering & Science, Applied Electrochemistry, Secretariat, IT
Detailed Electrochemical Characterisation of Large SOFC Stacks

As solid oxide fuel cell (SOFC) technology is moving closer to a commercial breakthrough, 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.

**General information**

State: Published
Number of pages: 53
Publication date: 2012

**Publication information**

Publisher: energinet.dk
Original language: English
Main Research Area: Technical/natural sciences
Electronic versions:
Durable_and_Robust_Solid_Oxide_Fuel_Cells.pdf

**Bibliographical note**

Energinet.dk project no. 2010-1-10441
Publication: Research › Report – Annual report year: 2013

**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.

**General information**

State: Published
Organisations: Department of Energy Conversion and Storage, Applied Electrochemistry, University of Groningen
Authors: Logtenberg, H. (Ekstern), van der Velde, J. H. M. (Ekstern), de Mendoza, P. (Ekstern), Areephong, J. (Ekstern), Esposito, V. (Intern), Brodersen, K. (Intern), Hagen, A. (Intern), Nielsen, J. (Intern), Feringa, B. L. (Ekstern), Browne, W. R. (Ekstern)
Pages: 24136−24142
Publication date: 2012
Main Research Area: Technical/natural sciences

**Publication information**
Highly durable anode supported solid oxide fuel cell with an infiltrated cathode

An anode supported solid oxide fuel cell with an La$_{0.6}$Sr$_{0.4}$Co$_{1.05}$O$_{3-\delta}$ (LSC) infiltrated-Ce$_{0.9}$Gd$_{0.1}$O$_{1.95}$ (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 Nieytria stabilized zirconia (YSZ) anode support, a Niercandia-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 evoltage 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.

**General information**

State: Published
Organisations: Department of Energy Conversion and Storage, Mixed Conductors, Applied Electrochemistry
Authors: Samson, A. J. (Intern), Hjalmarsson, P. (Intern), Søgaard, M. (Intern), Hjelm, J. (Intern), Bonanos, N. (Intern)
Pages: 124-130
Publication date: 2012
Main Research Area: Technical/natural sciences

**Publication information**

Journal: Journal of Power Sources
Volume: 216
ISSN (Print): 0378-7753
Ratings:
BFI (2017): BFI-level 1
Web of Science (2017): Indexed Yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 6.22 SJR 1.945 SNIP 1.483
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 1.945 SNIP 1.686 CiteScore 6.34
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 1.983 SNIP 2.071 CiteScore 6.3
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 1.985 SNIP 2.138 CiteScore 5.63
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): SJR 2.293 SNIP 2.016 CiteScore 5.04
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): SJR 2.247 SNIP 2.181 CiteScore 5.13
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 2.297 SNIP 1.981
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 2.117 SNIP 1.793
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 2
Scopus rating (2008): SJR 1.968 SNIP 1.726
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 1.597 SNIP 1.489
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 1.8 SNIP 2.224
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-YSZ 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.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Fundamental Electrochemistry, Applied Electrochemistry
Authors: Torres da Silva, I. M. (Intern), Mogensen, M. B. (Intern), Hjelm, J. (Intern)
Number of pages: 106
Publication date: 2012

PhD_thesis_Iris_Maura_Torres_da_Silva..PDF
Publication: Research › Ph.D. thesis – 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.
Determination of overpotential characteristics of reversible solid oxide cells via impedance spectroscopy and correlation with cell degradation

General information
State: Published
Authors: Graves, C. R. (Intern), Ebbesen, S. D. (Intern), Hjelm, J. (Intern)
Publication date: 2011

Host publication information
Title of host publication: Proceedings
Main Research Area: Technical/natural sciences
Conference: 220th ECS Meeting, Boston, MA, United States, 09/10/2011 - 09/10/2011
Electronic versions:
Determination of overpotential.pdf
Source: orbit
Source-ID: 316445
Publication: Research - peer-review › Conference abstract in proceedings – Annual report year: 2011

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 Wcm⁻² was observed with this design on a 0.5 cm² active cell level. ©2011 COPYRIGHT ECS - The Electrochemical Society
Durability of Solid Oxide Cells

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.

**General information**

State: Published


Authors: Knibbe, R. (Intern), Hauch, A. (Intern), Hjelm, J. (Intern), Ebbesen, S. D. (Intern), Mogensen, M. B. (Intern)

Pages: 141-169

Publication date: 2011

Main Research Area: Technical/natural sciences

**Publication information**

Journal: Green

Volume: 16

Issue number: 2

ISSN (Print): 1869-876X

Ratings:

- Scopus rating (2016): SJR 0.442 SNIP 0.677 CiteScore 1.14
- Scopus rating (2015): SJR 1.016 SNIP 1.276 CiteScore 2.46
- Scopus rating (2014): SJR 0.595 SNIP 0.999 CiteScore 1.74
- Scopus rating (2013): SJR 0.705 SNIP 1.317 CiteScore 2.9
- ISI indexed (2013): ISI indexed no
- Scopus rating (2012): SJR 0.647 SNIP 0.349
- ISI indexed (2012): ISI indexed no
- ISI indexed (2011): ISI indexed no

Original language: English

Solid Oxide Fuel Cells

DOIs:

10.1515/GREEN.2011.015

Source: orbit

Source-ID: 281466

Publication: Research - peer-review › Journal article – Annual report year: 2011

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**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

Authors: Brodersen, K. (Intern), Hauch, A. (Intern), Hjalmarsson, P. (Intern), Hjelm, J. (Intern)

Publication date: 2011


Main Research Area: Technical/natural sciences

Electronic versions:

- High performance anode.pdf

Source: orbit

Source-ID: 314319

Publication: Research › Conference abstract for conference – Annual report year: 2011

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**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.
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
Authors: Blennow Tullmar, P. (Intern), Hjelm, J. (Intern), Klemensø, T. (Intern), Persson, Å. H. (Intern), Ramousse, S. (Intern), Mogensen, M. B. (Intern)
Pages: 661-668
Publication date: 2011
Main Research Area: Technical/natural sciences
Technological implementation of LSC40 as porous cathode in SOFCs

General information
State: Published
Authors: Hjalmarsson, P. (Intern), Mosbæk, R. R. (Intern), Hjelm, J. (Intern)
Publication date: 2011
Event: Poster session presented at European Ceramic Society Conference, Stockholm (SE), .
Main Research Area: Technical/natural sciences

Electronic versions:
Poster_Stockholm.pdf

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 backscattered 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 Polarisation 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(°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(°C)).

General information
State: Published
Authors: Torres da Silva, I. M. (Intern), Nielsen, J. (Intern), Hjelm, J. (Intern), Mogensen, M. B. (Intern)
Pages: 489-498
Publication date: 2009
Conference: 216th ECS Meeting, Vienna, Austria, 04/10/2009 - 04/10/2009

DOI: 10.1111/j.1551-2916.2010.03763.x
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
Authors: Torres da Silva, I. M. (Intern), Nielsen, J. (Intern), Hjelm, J. (Intern), Mogensen, M. B. (Intern)
Pages: Abstract 1508
Publication date: 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
Authors: Jensen, S. H. (Intern), Hjelm, J. (Intern), Hagen, A. (Intern), Mogensen, M. B. (Intern)
Number of pages: 13
Publication date: 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 photochemical 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.
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
Authors: Hjelm, J. (Intern), Søgaard, M. (Intern), Knibbe, R. (Intern), Hagen, A. (Intern), Mogensen, M. B. (Intern)
Pages: 285-299
Publication date: 2008
Conference: 213th ECS Meeting, Phoenix, United States, 18/05/2008 - 18/05/2008
Main Research Area: Technical/natural sciences

Publication information
Journal: ECS Transactions
Volume: 13
Issue number: 26
ISSN (Print): 1938-5862
Ratings:
BFI (2017): BFI-level 1
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 0.4 SJR 0.231 SNIP 0.246
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 0.214 SNIP 0.257 CiteScore 0.36
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 0.214 SNIP 0.246 CiteScore 0.36
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 0.192 SNIP 0.237 CiteScore 0.27
ISI indexed (2013): 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
Authors: Areephong, J. (Ekstern), Kudernac, T. (Ekstern), de Jong, J. J. (Ekstern), Carroll, G. T. (Ekstern), Pantorott, D. (Ekstern), Hjelm, J. (Intern), Browne, W. R. (Ekstern), Feringa, B. L. (Ekstern)
Pages: 12850-12851
Publication date: 2008
Main Research Area: Technical/natural sciences

Publication information
Journal: Journal of the American Chemical Society
Volume: 130
Issue number: 39
ISSN (Print): 0002-7863
Ratings:
BFI (2017): BFI-level 2
Web of Science (2017): Indexed Yes
BFI (2016): BFI-level 2
Scopus rating (2016): SJR 7.368 SNIP 2.584 CiteScore 13.18
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): SJR 6.826 SNIP 2.632 CiteScore 12.81
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): SJR 6.273 SNIP 2.578 CiteScore 11.92
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): SJR 5.953 SNIP 2.455 CiteScore 11.38
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): SJR 6.141 SNIP 2.379 CiteScore 10.37
ISI indexed (2012): ISI indexed yes
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
Authors: Browne, W. R. (Ekstern), Areephong, J. (Ekstern), Katsonis, N. (Ekstern), Hjelm, J. (Intern), Feringa, B. L. (Ekstern)
Pages: 495-ORGN
Publication date: 2008

Host publication information
Title of host publication: Abstracts of Papers of the American Chemical Society
Volume: 235
Publisher: American Chemical Society
Main Research Area: Technical/natural sciences
Conference: 235th National Meeting of the American Chemical Society, New Orleans, LA, United States, 06/04/2008 - 06/04/2008
Source: orbit
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
Authors: Ramos, T. (Intern), Hjelm, J. (Intern), Wandel, M. (Intern), Hagen, A. (Intern), Mogensen, M. B. (Intern)
Pages: Abstract 377
Publication date: 2008

Host publication information
Title of host publication: Meeting Abstracts - Electrochemical Society
Publisher: Electrochemical Society, Incorporated
Main Research Area: Technical/natural sciences
Conference: 213th ECS Meeting, Phoenix, AZ, United States, 18/05/2008 - 18/05/2008
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).
Source: orbit
Source-ID: 264288
Publication: Research - peer-review › Conference abstract in proceedings – Annual report year: 2008

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
Authors: Wesenhagen, P. (Ekstern), Areephong, J. (Ekstern), Landaluce, T. (Ekstern), Heureux, N. (Ekstern), Katsonis, N. (Ekstern), Hjelm, J. (Intern), Rudolf, P. (Ekstern), Browne, W. (Ekstern), Feringa, B. (Ekstern)
Pages: 6334-6342
Publication date: 2008
Main Research Area: Technical/natural sciences

Publication information
Journal: Langmuir
Volume: 24
Issue number: 12
ISSN (Print): 0743-7463
Ratings:
BFI (2017): BFI-level 2
Web of Science (2017): Indexed Yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 3.99 SJR 1.55 SNIP 1.188
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): SJR 1.686 SNIP 1.308 CiteScore 4.33
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): SJR 1.816 SNIP 1.391 CiteScore 4.59
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): SJR 1.895 SNIP 1.356 CiteScore 4.55
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): SJR 2.177 SNIP 1.382 CiteScore 4.37
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): SJR 2.051 SNIP 1.357 CiteScore 4.42
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 2
Scopus rating (2010): SJR 2.148 SNIP 1.4
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 2
Scopus rating (2009): SJR 2.156 SNIP 1.351
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 2.383 SNIP 1.34
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 2.449 SNIP 1.434
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 2.375 SNIP 1.428
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 2.157 SNIP 1.463
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 1.963 SNIP 1.458
Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 1.953 SNIP 1.4
Web of Science (2003): Indexed yes
Scopus rating (2002): SJR 2.011 SNIP 1.489
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 2.01 SNIP 1.382
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 2.039 SNIP 1.479
Web of Science (2000): Indexed yes
Scopus rating (1999): SJR 1.719 SNIP 1.496
Original language: English
DOIs:
10.1021/la703999x
Source: orbit
Source-ID: 222877
Publication: Research - peer-review › Journal article – Annual report year: 2008

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
Authors: Menon, M. (Intern), Hjelm, J. (Intern)
Publication date: 2008
Main Research Area: Technical/natural sciences
Source: orbit
Source-ID: 232445
Publication: 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.
Synthesis and characterization of dinuclear ruthenium(II) complexes based on 4,4’-bipyridyl type bridging ligands

General information
State: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Authors: Guckian, A. (Ekstern), Browne, W. (Ekstern), Hjelm, J. (Intern), Vos, J. (Ekstern)
Pages: 1151-1161
Publication date: 2007
Main Research Area: Technical/natural sciences

Publication Information
Journal: Journal of the Chinese Chemical Society
Volume: 54
ISSN (Print): 0009-4536
Ratings:
BFI (2017): BFI-level 1
Web of Science (2017): Indexed Yes
BFI (2016): BFI-level 1
Scopus rating (2016): SJR 0.256 SNIP 0.247 CiteScore 0.74
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 0.291 SNIP 0.401 CiteScore 0.79
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 0.263 SNIP 0.407 CiteScore 0.72
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 0.263 SNIP 0.454 CiteScore 0.9
ISI indexed (2013): ISI indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): SJR 0.302 SNIP 0.556 CiteScore 0.87
ISI indexed (2012): ISI indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): SJR 0.257 SNIP 0.413 CiteScore 0.73
ISI indexed (2011): ISI indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 0.265 SNIP 0.396
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 0.27 SNIP 0.353
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 0.306 SNIP 0.516
Scopus rating (2007): SJR 0.294 SNIP 0.553
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 0.249 SNIP 0.425
Scopus rating (2005): SJR 0.313 SNIP 0.439
Scopus rating (2004): SJR 0.268 SNIP 0.476
Scopus rating (2003): SJR 0.287 SNIP 0.435
Scopus rating (2002): SJR 0.301 SNIP 0.403
Scopus rating (2001): SJR 0.315 SNIP 0.402
Scopus rating (2000): SJR 0.266 SNIP 0.392
Scopus rating (1999): SJR 0.226 SNIP 0.295
Projects:

Electrochemical Characterization for Improvement of PEM Electrolysers for Flexible Energy Storage
Department of Energy Conversion and Storage
Period: 15/06/2015 → 14/06/2018
Number of participants: 4
PhD Student:
Elsøe, Katrine (Intern)
Supervisor:
Hjelm, Johan (Intern)
Ramos, Tania (Intern)
Main Supervisor:
Mogensen, Mogens Bjerg (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Forskningsrådsfinansiering
Project: PhD

In-operando spatially resolved probing of solid oxide electrolysis/fuel cells
Department of Energy Conversion and Storage
Period: 01/05/2015 → 30/04/2018
Number of participants: 5
PhD Student:
Pitscheider, Simon (Intern)
Supervisor:
Chueh, William C. (Ekstern)
Hansen, Karin Vels (Intern)
Hjelm, Johan (Intern)
Main Supervisor:
Chatzichristodoulou, Christodoulos (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: Forskningsrådsfinansiering
Project: PhD

Electron microscopy of electrochemical processes
Department of Micro- and Nanotechnology
Period: 15/04/2014 → 14/04/2017
Number of participants: 7
PhD Student:
Canepa, Silvia (Intern)
Supervisor:
Sun, Hongyu (Intern)
Wagner, Jakob Birkedal (Intern)
Main Supervisor:
Mølhave, Kristian (Intern)
Examiner:
Hjelm, Johan (Intern)
Alloyeau, Damien (Ekstern)
Dahl, Søren (Intern)
**Financing sources**
Source: Internal funding (public)
Name of research programme: Forskningsrådsfinansiering
Project: PhD

**Lifetime limiting effects in pre-commercial solid cell devices**
Department of Energy Conversion and Storage
Period: 01/03/2014 → 21/06/2017
Number of participants: 7
Phd Student:
Skafte, Theis Løye (Intern)
Supervisor:
Blennow Tullmar, Peter (Intern)
Graves, Christopher R. (Intern)
Main Supervisor:
Hjelm, Johan (Intern)
Examiner:
Hauch, Anne (Intern)
Lanzini, Andrea (Ekstern)
Weber, André (Ekstern)

**Financing sources**
Source: Internal funding (public)
Name of research programme: Industrial PhD
Project: PhD

**Elektrode Kinetics and Gas Conversion in Solid Oxide Cells**
Department of Energy Conversion and Storage
Period: 01/01/2014 → 20/04/2016
Number of participants: 7
Phd Student:
Njodzefon, Jean-Claude (Intern)
Supervisor:
Graves, Christopher R. (Intern)
Weber, André (Ekstern)
Main Supervisor:
Hjelm, Johan (Intern)
Examiner:
Hauch, Anne (Intern)
Krügel, Albert (Ekstern)
Schefold, Josef (Ekstern)

**Financing sources**
Source: Internal funding (public)
Name of research programme: Eksternt finansieret virksomhed

**Relations**
Publications:
Electrode Kinetics and Gas Conversion in Solid Oxide Cells
Project: PhD

**Computational Investigations of transport mechanisms across battery interfaces**
Department of Energy Conversion and Storage
Period: 01/07/2013 → 16/11/2016
Number of participants: 6
Phd Student:
Loftager, Simon (Intern)
Supervisor:
Garcia-Lastra, Juan Maria (Ekstern)
Main Supervisor:
Vegge, Tejs (Intern)
Examiner:
Hjelm, Johan (Intern)
Khalifah, Peter (Ekstern)
Rossmeisl, Jan (Intern)

Financing sources
Source: Internal funding (public)
Name of research programme: 1/3 FUU, 1/3 inst 1/3 Andet
Project: PhD

Performance and lifetime limiting effects in Li-ion batteries
Department of Energy Conversion and Storage
Period: 01/06/2013 → 20/09/2016
Number of participants: 8
Phd Student:
Scipioni, Roberto (Intern)
Supervisor:
Hjelm, Johan (Intern)
Norby, Poul (Intern)
Main Supervisor:
Jensen, Søren Højgaard (Intern)
Examiner:
Bowen, Jacob R. (Intern)
Bowen, Jacob R. (Intern)
Lindbergh, Göran (Ekstern)
Lindbergh, Göran (Ekstern)

Financing sources
Source: Internal funding (public)
Name of research programme: Offentlig finansiering

Relations
Publications:
Performance and Lifetime Limiting Effects in Li-ion Batteries
Project: PhD

Investigation of performance and lifetime limiting effects in Li-air battery cells
Department of Energy Conversion and Storage
Period: 01/12/2012 → 22/02/2016
Number of participants: 6
Phd Student:
Knudsen, Kristian Bastholm (Intern)
Supervisor:
Jensen, Søren Højgaard (Intern)
Main Supervisor:
Hjelm, Johan (Intern)
Examiner:
Hagen, Anke (Intern)
Adelhelm, Philipp (Ekstern)
Edström, Ester Kristina (Ekstern)

Financing sources
Source: Internal funding (public)
Name of research programme: Forskningsrådsfinansiering
Project: PhD
Towards Smart Grid Ready SOFC
Department of Energy Conversion and Storage
Applied Electrochemistry
IRD Fuel Cells A/S
Topsoe Fuel Cell
Period: 01/08/2012 → 31/07/2014
Number of participants: 1
solid oxide fuel cells, smart grid, reformate gas, electrochemistry, durability, load cycles, stacks
Project Coordinator:
Hjelm, Johan (Intern)

SOFC Stack Diagnostics
Department of Energy Conversion and Storage
Period: 01/04/2011 → 04/02/2015
Number of participants: 7
Phd Student:
Mosbæk, Rasmus Rode (Intern)
Supervisor:
Barfod, Rasmus Gottrup (Intern)
Hendriksen, Peter Vang (Intern)
Main Supervisor:
Hjelm, Johan (Intern)
Examiner:
Nielsen, Jimmi (Intern)
Haart, L. G. J. de (Ekstern)
Weber, André (Ekstern)

Financing sources
Source: Internal funding (public)
Name of research programme: Institut, samfinansiering
Project: PhD

In situ characterisation of structure and transport in battery electrolytes and electrodes
Department of Energy Conversion and Storage
Period: 01/01/2011 → 26/02/2014
Number of participants: 7
Phd Student:
Sveinbjörnsson, Dadi Þorsteinn (Intern)
Supervisor:
Mogensen, Mogens Bjerg (Intern)
Norby, Poul (Intern)
Main Supervisor:
Vegge, Tejs (Intern)
Examiner:
Hjelm, Johan (Intern)
David, William Ian Fraser (Ekstern)
Serby, Magnus H. (Ekstern)

Financing sources
Source: Internal funding (public)
Name of research programme: Globaliseringsmidler
Project: PhD
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.

Department of Energy Conversion and Storage
Applied Electrochemistry
IRD Fuel Cells A/S
Period: 01/07/2010 → 30/06/2012
Number of participants: 1
solid oxide fuel cells, electrochemistry, degradation, durability, lifetime limiting factors, porous electrode, functional ceramics, high temperature materials, hydrogen
Project Coordinator:
Hjelm, Johan (Intern)

Financing sources
Source: Public research programme (public)
Name of research programme: ForskEL (Energinet.dk)
Web address: http://www.forskel.dk
Amount: 11,000,000.00 Danish Kroner
Year of approval: 2010

Degradation mechanism of the LSCF Cathode
Department of Energy Conversion and Storage
Period: 15/04/2010 → 19/03/2013
Number of participants: 7
Phd Student:
Zhang, Weiwei (Intern)
Supervisor:
Barfod, Rasmus Gottrup (Intern)
Hendriksen, Peter Vang (Intern)
Main Supervisor:
Chen, Ming (Intern)
Examiner:
Hjelm, Johan (Intern)
Hallstedt, Bengt (Ekstern)
Markus, Torsten (Ekstern)

Financing sources
Source: Internal funding (public)
Name of research programme: DTU, Samfinansiering
Project: PhD

Low Temperature NOx decomposition using an electrochemical Reactor
Department of Energy Conversion and Storage
Period: 01/03/2010 → 19/08/2013
Number of participants: 5
Phd Student:
Shao, Jing (Intern)
Main Supervisor:
Kammer Hansen, Kent (Intern)
Examiner:
Hjelm, Johan (Intern)
Kustov, Arkadii (Intern)
Vernoux, Philippe (Ekstern)
Financing sources
Source: Internal funding (public)
Name of research programme: Forskningsrådsfinansiering
Project: PhD

High Temperature Alkaline Electrolyser Cell
Department of Energy Conversion and Storage
Period: 15/05/2009 → 01/03/2013
Number of participants: 7
Phd Student:
Allebrod, Frank (Intern)
Supervisor:
Ebbesen, Sune Dalgaard (Intern)
Hjelm, Johan (Intern)
Main Supervisor:
Mogensen, Mogens Bjerg (Intern)
Examiner:
Holtappels, Peter (Intern)
Skou, Eivind Morten (Ekstern)
Sunde, Svein (Ekstern)

Investigating the Stability of Electrode-electrolyte-interfaces
Department of Energy Conversion and Storage
Period: 01/02/2008 → 08/02/2012
Number of participants: 6
Phd Student:
Torres da Silva, Iris Maura (Intern)
Supervisor:
Mogensen, Mogens Bjerg (Intern)
Main Supervisor:
Hjelm, Johan (Intern)
Examiner:
Skaarup, Steen (Intern)
Skou, Eivind Morten (Ekstern)
Weber, André (Ekstern)

Financing sources
Source: Internal funding (public)
Name of research programme: Forskningsrådsfinansiering
Project: PhD