NOx conversion in La$_{0.85}$Sr$_{0.15}$Co$_{0.03}$Mn$_{0.97}$O$_{3+d}$-Ce$_{0.9}$Gd$_{0.1}$O$_{1.95}$ porous cell stacks infiltrated with Pt

Porous cell stacks with composite electrodes of La$_{0.85}$Sr$_{0.15}$Co$_{0.03}$Mn$_{0.97}$O$_{3+d}$ and Ce$_{0.9}$Gd$_{0.1}$O$_{1.95}$ were characterized for the electrochemical reduction of NO in net oxidizing atmosphere in absence or presence of propene in the feed gas. No NOx was converted when the porous cell stacks were at OCV or when polarized. However, when the cells were infiltrated with Pt, an effect of this was observed. It was shown that Pt had a very positive effect on the NOx removal properties of the porous cell stacks, and that NOx could be removed both at OCV or when the porous cells stacks was polarized, both in the absence or presence of propene under net oxidizing conditions. The porous cell stacks was also investigated using electrochemical impedance spectroscopy. It was shown that the impedance data could be de-convoluted into four arcs. Each arc could be fitted with a constant phase elements in series with a resistor. The arcs could be attributed to physical processes.
Effect of cobalt on the activity of dual phase \((\text{Gd}_{0.6}\text{Sr}_{0.4})_{0.99}\text{Fe}_{1-x}\text{CoxO}_{3-\delta}\) SOFC cathodes

The effect of the amount of cobalt in \((\text{Gd}_{0.6}\text{Sr}_{0.4})_{0.99}\text{Fe}_{1-x}\text{CoxO}_{3-\delta}\) solid oxide fuel cell (SOFC) cathodes \((x = 0.00, 0.05, 0.10, 0.15, 0.20\text{ and } 0.25)\) was studied by powder X-ray diffraction (XRD), scanning electron microscopy (SEM), dilatometry, four-point DC conductivity measurements, cone-shaped electrodes, and electrochemical impedance spectroscopy (EIS). XRD and scanning electron microscopy revealed two phases: a cubic phase and an orthorhombic phase. One phase has a small particle size (cubic phase), and the other phase (orthorhombic) has a larger particle size. Dilatometry indicated that the thermal expansion coefficient increased with increasing Co content. The electrical conductivity maximum for the Co-rich compound was found to have a value of 82 S cm\(^{-1}\) at approximately 300 °C. It was shown that the activity of the cathodes was strongly dependent on the amount of cobalt. The highest activity was found for the cobalt-rich compound \((\text{Gd}_{0.6}\text{Sr}_{0.4})_{0.99}\text{Fe}_{0.75}\text{CoxO}_{3-\delta}\) with an area-specific resistance of 0.9 Ωcm\(^{2}\) at 600 °C, measured on a cone-shaped electrode in air.

Highly porous Ce-W-TiO\(_2\) free-standing electrospun catalytic membranes for efficient de-NO\(_x\) via ammonia selective catalytic reduction

Highly porous Ce-W-TiO\(_2\) free-standing nanofibrous membranes (FSM) are fabricated via electrospinning techniques to serve as NO\(_x\)-SCR catalysts. The precursor of the ceramic nanofibers (sol-gel solution) is co-electrospun with a poly(vinyl alcohol) (PVA) water solution. PVA integration into FSM is proven to avoid excessive bending of the nanofibers, and to prevent mechanical failure of the final ceramic nanofibrous structure. This is demonstrated to be associated with PVA higher thermal stability compared with the other organic additives. 3D tomography reconstruction indicates a resulting ceramic membrane with a great open and interconnected porosity of ca. 96%. The catalytic characterization, performed at the best working conditions (in absence of H\(_2\)O and SO\(_2\)), indicates the amorphous FSM as the best performing catalytic membrane. Superior catalytic performances for the developed FSM, over the nanofibers and the nanoparticles catalysts are proven, as a result of superior surface, morphological, and structural features. Long-term stability (120 h) and reproducibility (over 5 cycles) of the FSM are also demonstrated.
Highly porous Ce–W–TiO₂ free-standing electrospun catalytic membranes for efficient de-NOₓ via ammonia selective catalytic reduction

Highly porous Ce–W–TiO₂ free-standing nanofibrous membranes (FSM) are fabricated via electrospinning techniques to serve as NOₓ-SCR catalysts. The precursor of the ceramic nanofibers (sol–gel solution) is co-electrospun with poly(vinyl alcohol) (PVA) water solution. PVA integration into FSM has been proven to avoid excessive bending of the nanofibers and to prevent mechanical failure of the final ceramic nanofibrous structure. This is demonstrated to be associated with higher thermal stability of PVA compared with that of other organic additives. 3D tomography reconstruction indicates a resulting ceramic membrane with remarkable open and interconnected porosity of ca. 96%. Catalytic characterization, performed at the best working conditions (in absence of H₂O and SO₂), indicates that amorphous FSM is the best performing catalytic membrane. Superior catalytic performances of the developed FSM over those of other nanofibers and nanoparticle catalysts are proven because of superior surface, morphological, and structural features. Long-term stability (120 h) and reproducibility (over 5 cycles) of FSM are also demonstrated.

Silver Modified Cathodes for Solid Oxide Fuel Cells

La₁₋ₓ₋₅Sr₅AgₓMnO₃₋δ was synthesized via modified Pechini method. Silver (Ag) was incorporated into La₁₋ₓ₋₅Sr₅AgₓMnO₃₋δ perovskite single phase formed at 800°C, and the solubility of Ag in the crystal lattice of the perovskite was up to 5 mol%. The single-phase materials revealed thermal stability in different oxidizing atmospheres up to sintering temperature of the cathode at 1050°C. The exsolution of the metallic Ag nanoparticles was performed at 350–600°C in reducing atmosphere. Scanning electron and scanning transmission electron microscopy revealed a good mechanical contact of the Ag nanoparticles to the surface of the perovskite after reducing conditions. The electrochemical tests of the materials showed a good electrocatalytic effect of nanosized Ag toward oxygen reduction reaction. The electrochemical
performance of the cathodes revealed the dependence on electrolyte material and exsolution time.

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**Activation/Deactivation Phenomena in the Electrochemical Reduction of Nitric oxide and Oxygen on LSM perovskites**
The effect of anodic pretreatment on LaMnO3+delta, La0.85Sr0.15MnO3+delta and La0.5Sr0.5MnO3 point electrodes were investigated using cyclic voltammetry in the temperature range 200 to 400 degrees C in either 10% oxygen in argon or 1% nitric oxide in argon. The electrodes, in most cases, were deactivated in oxygen-containing gas and activated in nitric oxide-containing gas after anodic polarization. This was thought to be due to potential-induced SrO segregation to the surface of the electrodes and annihilation of oxygen vacancies. This work shows that the effect in some cases is significant and must be considered when evaluating the electrodes. In addition, the activity of the electrodes increases with increasing strontium content with respect to the reduction of both oxygen and nitric oxide. The electrodes were all more active with respect to the reduction of nitric oxide than with respect to the reduction of oxygen in the investigated temperature range.

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**Amorphous saturated Cerium-Tungsten-Titanium oxide nanofibers catalysts for NOx selective catalytic reaction**
Herein for the first time, Ce0.184W0.07Ti0.748O2-δ nanofibers are prepared by electrospinning to serve as catalyst in the selective catalytic reduction (SCR) process. The addition of cerium is proven to inhibit crystallization of TiO2, yielding an
amorphous TiOx-based solid solution stable up to 500 °C in air, with supersaturated substitutional Ce. However, at higher temperatures, anatase phase (titanium oxide) is then observed along with fluorite (cerium oxide). Tungsten is instead demonstrated to promote the reduction of the Ce$^{4+}$ to Ce$^{3+}$ with formation of oxygen vacancies ($\delta$). Catalytic experiments at the best working conditions (dry and in absence of SO$_2$) are performed to characterize the intrinsic catalytic behavior of the new catalysts. At temperature lower than 300 °C, superior NOx conversion properties of the amorphous TiOx nanofibers over the crystallized TiO$_2$(anatase) nanofibers are observed and attributed to higher specific surface area (SSA), larger amount of oxygen vacancies, and higher amount of Ce$^{3+}$ over the Ce$^{4+}$. Comparison with literature data for ceria-tungsten-based nanoparticles also points out higher catalytic performances for the developed nanofibers at the lowest temperatures (< 300°C). This is mainly attributed to the unique nanofibrous morphology and to the doping approach. Stability of the amorphous Ce-W-TiOx nanofibers over time (120 h) and over a number of cycles (5) is demonstrated. Yet, superior catalytic performances of the developed catalysts in a wide range of temperatures (200-500 °C) over state-of-the-art material V-W-titania nanoparticles and nanofibers are also proven.

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Organisations: Department of Energy Conversion and Storage, Ceramic Engineering & Science, Imaging and Structural Analysis, Mixed Conductors, Silpakorn University, Technical University of Denmark
Contributors: Dankeaw, A., Gualandris, F., Silva, R. H., Norrman, K., Gudik-Sørensen, M., Kammer Hansen, K., Ksapabutr, B., Esposito, V., Marani, D.
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Source-ID: 147135777
Research output: Contribution to journal → Journal article – Annual report year: 2018 → Research → peer-review

**Communication—Perovskite Electrochemical System for Highly Selective NO$_x$ Reduction of Diesel Engine Exhaust**
A perovskite electrochemical system was developed for selective NO$_x$ reduction of diesel engine exhaust. The system was composed of a La$_{0.9}$Sr$_{0.1}$CoO$_{3+\delta}$ oxidation catalyst and a (La$_{0.85}$Sr$_{0.15}$)$_{0.95}$MnO$_{3+\delta}$/Ce$_{0.9}$Gd$_{0.1}$O$_{1.95}$ electrochemical cell with BaO nanoparticles. A selectivity of 25–35% was achieved with a NO$_x$ conversion of 65–75% in 1000 ppm NO with 8% O$_2$ at 375°C. The superior performance of the system was suggested to be ascribed to the promotion in NO$_2$ formation substantially activating the NO$_x$ trapping and reduction processes on the electrode.

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Organisations: Department of Energy Conversion and Storage, Proton conductors, Electrochemical Materials and Interfaces, Shenzhen University, Southern University of Science and Technology
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Cr- and Ti-Based Spinels as Materials for Anodic Catalyst Support in PEM Electrolysis Cells: Assessing Corrosion Stability and Support Role in Catalyst Activity of Corrosion Stable Ceramics

This work aims to determine the stability of Cr- and Ti-based spinels as catalyst supports for oxygen evolution reaction (OER) catalyst in PEM electrolyzers (PEMECs). Different compositions of MCr2O4 (M=Ni, NiFe, Zn, Mg) and MTi2O4 (Li, Mg, Mn) have been synthesized by solid state synthesis. Pure and doped Cr-based spinels exhibit low conductivities at the operating temperatures of PEMECs (1.5 V vs SHE. LiT2O4 is completely oxidized upon cycling up to 2.0 V vs SHE. Mixtures of IrO2/oxide support deposited on glassy carbon were tested toward OER, which showed a 10% higher absolute current at 2.0 V vs SHE in the case of IrO2/Cu-MnCr2O4 compared with pure IrO2.

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Organisations: Department of Energy Conversion and Storage, Electrochemical Materials and Interfaces, University of St Andrews
Contributors: Fenini, F., Hansen, K. K., Savaniu, C., Irvine, J. T. S., Mogensen, M. B.
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Research output: Contribution to journal > Journal article – Annual report year: 2018 > Research > peer-review
Electrochemical removal of NOx using oxide-based electrodes - A review

Solid-state electrochemical reduction of NOx using oxide-based electrodes is reviewed. Different types of electrode and cell geometries have been used in the literature. Using simple ceramic point electrodes prepared using different materials; it has been shown that nitric oxide is reduced at different rates on different materials. For perovskites, it has been shown that the amount of trivalent transition metal and amount of oxide ion vacancies are important for the reduction of nitric oxide. The same applies to Cu and Ni-based K2NiF4 structures. For spinels, the pattern is less clear, but they are all able to reduce nitric oxide. Current densities are much higher when reducing nitrogen dioxide compared with nitric oxide on both perovskites and spinels. In gas mixtures containing nitric oxide and oxygen, the addition of BaO leads to fairly high conversion of nitric oxide into nitrogen. Finally, it has been shown that oxidizing nitric oxide to nitrogen dioxide before reducing to nitrogen is very beneficial, leading to current efficiencies of up to 65%.

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Novel Processing of Cathodes for Solid Oxide Fuel Cells

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Studies of A-site Deficient (Gd0.6Sr0.4)1−xFexCo0.2O3−δ Cathodes in SOFCs
A series of A-site deficient Gd-containing Fe-Co-based cathodes were synthesized using the glycine-nitrate process. All the compounds consisted of two phases, i.e., a cubic and orthorhombic phase, as determined by powder X-ray diffraction (XRD). The thermal expansion coefficient decreased with increasing A-site deficiency, which could be because cobalt was expelled from the main phases. Likewise, the electrochemical activities of the Gd-containing Fe-Co-based solid oxide fuel
cell cathodes toward the reduction of oxygen were improved by a factor of approximately 3 by making them A-site deficient. The composition (Gd0.6Sr0.4)0.85Fe0.8Co0.2O3–δ showed the highest activity toward the reduction of oxygen among the five compounds. Electrochemical impedance spectroscopy estimated the area specific resistance as 5.77 Ω cm⁻² at 600 °C. The electrochemical impedance spectroscopy measurements revealed three arcs for most of the compounds at the three temperatures where the measurements were performed.

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**The Role of Pore-Formers on Grain Interior and Grain Boundary Conductivity in Tape-Cast Porous Sheets for Electrochemical Flue Gas Purification**

Ce₀.₉Gd₀.₁O₁.₉₅ (CGO) electrolytes for electrochemical flue gas purification were fabricated by means of tape casting with different types, shapes and sizes of pore-formers. The sintered bodies were characterized with electrochemical impedance spectroscopy, to investigate the role of the different pore-formers on the electrochemical properties of the cast tapes. A strong effect of the different pore-formers on the conductivity (both grain interior and grain boundary conductivities) was observed. In addition, the conductivity data were also correlated with previously obtained gas permeability data. The conductivity data correlated with the permeability data in the sense that a higher permeability lead to a lower conductivity. The porosity of the samples also influenced the conductivities. The higher the porosity of the sintered bodies, the lower the conductivity was, as expected.

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**Cathode-supported hybrid direct carbon fuel cells**
The direct conversion of coal to heat and electricity by a hybrid direct carbon fuel cell (HDCFC) is a highly efficient and cleaner technology than the conventional combustion power plants. HDCFC is defined as a combination of solid oxide fuel cell and molten carbonate fuel cell. This work investigates cathode-supported cells as an alternative configuration for
HDCFC, with better catalytic activity and performance. This study aims to define the best processing route to manufacture highly efficient cathode-supported cells based on La0.75Sr0.25MnO3/yttria-stabilized zirconia infiltrated backbones. The challenges on the development of high-performance backbones are discussed. In this study, cathode-supported configuration was confirmed to be more efficient for the oxidation of carbon than anode supported configuration. The maximum power density of the cathode-supported cell increased almost by a factor of two when compared with the anode-supported cell.

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**Cone-Shaped Gd_{1-x}Sr_{x}Fe_{0.8}Co_{0.2}O_{3-δ} Electrodes for SOFC Cathodes**
Five Gd_{1-x}Sr_{x}Fe_{0.8}Co_{0.2}O_{3-δ} perovskites were synthesized using the glycine-nitrate process. The compounds were evaluated as solid oxide fuel cell cathodes using cone-shaped electrodes and electrochemical impedance spectroscopy. It was shown that the electrochemical activity depended on the amount of strontium in the perovskite; the main difference seemed to be whether the perovskite is a single or a two-phase compound. However, high-strontium-substituted Fe-Co-based perovskites have slightly higher performances than the low-strontium-substituted Fe-Co-based perovskites, as determined by electrochemical impedance spectroscopy on cone-shaped electrodes.

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Determination of the Resistance of Cone-Shaped Solid Electrodes

A cone-shaped electrode pressed into an electrolyte can with advantage be utilized to characterize the electro-catalytic properties of the electrode, because it is less dependent on the electrode microstructure than e.g. thin porous composite electrodes, and reactions with the electrolyte occurring during processing can be avoided. Newman's formula for current constriction in the electrolyte is then used to deduce the active contact area based on the ohmic resistance of the cell, and from this the surface specific electro-catalytic activity. However, for electrode materials with low electrical conductivity (like Ce$_{1-x}$Pr$_x$O$_{2-δ}$), the resistance of the cell is significantly influenced by the ohmic resistance of the cone electrode, wherefore it must be included. In this work the ohmic resistance of a cone is modelled analytically based on simplified geometries. The two analytical models only differ by a model specific pre-factor, which is consequently determined by a finite element model. The model was applied to measurements on cones of Ce$_{1-x}$Pr$_x$O$_{2-δ}$ characterized on a YSZ electrolyte. Conclusively, the finite element model was used to obtain a formula for the resistance for different cone angles with a small contact area. This reproduces Newman's formula for a cone angle equal to 90°, i.e. a semi-infinite body.

Direct Coal Oxidation in Modified Solid Oxide Fuel Cells

Hybrid direct carbon fuel cells employ a classical solid oxide fuel cell together with carbon dispersed in a carbonate melt on the anode side. In a European project, the utilization of various coals has been investigated with and without addition of an oxidation catalyst to the carbon-carbonate slurry or anode layer. The nature of the coal affects both open circuit voltage and power output. Highest OCV and power densities were observed for bituminous coal and by adding manganese oxide or praseodymium-doped ceria to the carbon/carbonate mixture. Comparing the carbon black fueled performance of an anode supported (315 μm anodes) and cathode supported cell (15 μm anode) indicates a superior performance of the latter. Using un-catalyzed biomass (charcoal) as fuel results in an OCV of 941 mV and a maximum power density of 78 mW/cm$^2$ at 755°C similar to the power output of manganese oxide catalyzed bituminous coal (73 mW/cm$^2$).
Effect of CeO2 Addition on Hybrid Direct Carbon Fuel Cell Performance

The effect of CeO2 infiltration into the anode or CeO2 mixed with the carbon-fuel on the performance of a Hybrid Direct Carbon Fuel Cell (HDCFC) was studied through the use of polarization curves and electrochemical impedance spectroscopy. The use CeO2 in both ways helped to increase the cell performance. In particular, mixing CeO2 with carbon represents the best strategy to increase the cell power output, probably due to increased formation of CO.

Materials for Catalysis, Synthetic Fuels and Chemical Energy Conversion

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NOx Selective Catalytic Reduction (SCR) on Self-Supported V-W-doped TiO2 Nanofibers

Electrospun V–W–TiO2 catalysts, resulting in a solid solution of V and W in the anatase phase, are prepared as nonwoven nanofibers for NOx selective catalytic reduction (SCR). Preliminary catalytic characterization indicates their superior NOx conversion efficiency to the-state-of-the-art material. A novel concept of a self-supported, ultra-compact, and lightweight nanofibrous SCR-reactor is defined.

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Permeability, strength and electrochemical studies on ceramic multilayers for solid-state electrochemical cells

An electrochemical reactor can be used to purify flue gasses. Such a reactor can be a multilayer structure consisting of alternating layers of porous electrodes and electrolytes (a porous cell stack). In this work optimization of such a unit has been done by changing the pore former composition and the electrode powder pre-treatment. The effect on permeability, mechanical strength and electrochemical behavior was studied in this work. The effects were evaluated by measuring the pressure difference over the samples in relation to the flow through the sample, by the ball on ring method and by electrochemical impedance spectroscopy in air at temperatures between 300 and 450 °C. The resulting structures were also evaluated with scanning electron microscopy. The work showed a dependence on the pore former composition and electrode powder pre-treatment resulting in variations in porosity, strength and flow resistance. A higher porosity gives a lower backpressure. The electrochemical performance shows that both thickness and amount of pore former in the electrolyte is important, but almost no dependence of electrode composition on the polarization resistances within the tested compositions.

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Effect of pore formers on properties of tape cast porous sheets for electrochemical flue gas purification

Ce0.9Gd0.1O1.95 (CGO) electrolytes for electrochemical flue gas purification multilayers were fabricated by tape casting and sintering using different types, shapes and sizes of pore formers. The resulting tapes (with thickness of about 400μm) were characterized by scanning electron microscopy, gas permeability measurements, mercury porosimetry and pore orientation measurements, to investigate the role of the different pore formers on the properties after sintering at a temperature of 1250°C. Those tapes prepared from different non-spherical pore formers with comparable porosity of about 43%, showed significant differences in gas permeability which could be correlated to an increase in mean pore size and pore connectivity. The degree of pore orientation in the tape casting direction was determined by best-fit ellipse method and a modified linear intercept method and the obtained data were correlated with the corresponding gas permeability.

Electrochemical Reduction of NO₂
Electrochemical removal of NOx using solid oxide cells

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Fabrication of doped Titania (TiO2) nanofibers to serve as catalysts in NH3-Selective Catalytic Reduction (SCR)

In a context of significant interest for energy and environment, nanostructured-based ceramic materials are considered ideal candidates for the development of cost and energy efficient innovative systems. Such an attention is essentially due to the unique properties originating from the confinement of either one or more dimensions into the nanoscale level. Among others the large surface-to-volume ratio is a feature that greatly increases the reactivity of the nanomaterials towards gaseous species when compared with the non-nanodimensional materials. With this regards, catalysis is one of those applications that unquestionable benefits from this novel feature. In addition, when nanofibers (1D nanostructure) are used as catalysts, the further advantage of a self-supported wide open and well-interconnected porous structure is achieved. Herein we demonstrate nanofibers as catalysts for the removal of the NOx in exhausts via the NH3 Selective Catalytic Reduction (SCR) method. By combining electrospinning and sol-gel chemistry, materials are processed as nanofibers with the catalytic components (e.g. V2O5-WO3) incorporated as dopants into the supporting anatase phase (e.g TiO2). Remarkable high NOx conversion efficiencies are obtained and associated with the unique features deriving from the synergism among the doping approach, the nanoscale confinement, and the nano-fibrous texture. A novel concept of self-supported, lightweight and ultra-compact design SCR reactor is defined.

General information
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Highly selective NOx reduction for diesel engine exhaust via an electrochemical system

It is challenging to reduce the nitrogen oxides (NOx) in diesel engine exhaust due to the inhibiting effect of excess oxygen. In this study, a novel electrochemical deNOx system was developed, which eliminated the need for additional reducing materials or a sophisticated controlling system as used in current diesel after-treatment techniques. The electrochemical system consisted of an electrochemical cell modified with NOx adsorbents and a diesel oxidation catalyst placed upstream of the cell. The system offers highly selective NOx reduction and a strong resistance to oxygen interference with almost
zero emission of secondary pollutants.

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Organisations: Department of Energy Conversion and Storage, West Virginia University
Contributors: Shao, J., Tao, Y., Kammer Hansen, K.
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Research output: Contribution to journal › Journal article – Annual report year: 2016 › Research › peer-review

**Influence of pore former on porosity and mechanical properties of Ce0.9Gd0.1O1.95 electrolytes for flue gas purification**
Single layered porous Ce0.9Gd0.1O1.95 electrolytes were fabricated by tape casting using different types, shapes and sizes of pore formers and their respective strength and stiffness were compared. The sintered bodies were characterized by scanning electron microscopy, mercury porosimetry, impulse excitation technique (Young modulus) and flexural strength measurements, to investigate the role of the different pore formers on the properties of the compounds. The compared techniques used to evaluate porosity give consistent results. The ratio between open and total porosities, evaluated from mercury porosimetry, varies depending on the used pore formers. The stiffness and strength of the compounds show an exponential dependency to the total porosity. By considering the open porosity instead (functional porosity), we observe that samples with platelets shaped pore formers have higher in-plane strength than spherical pore formers. An optimum can be found in term of Weibull strength and strain of samples obtained with the various pore formers by considering the dependency on the functional open porosity instead of the total porosity.
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Keywords: Tape casting, Pore former, Mechanical properties, Strength, CGO
DOI:
New Hypothesis for SOFC Ceramic Oxygen Electrode Mechanisms

A new hypothesis for the electrochemical reaction mechanism in solid oxide cell ceramic oxygen electrodes is proposed based on literature including our own results. The hypothesis postulates that the observed thin layers of SrO-La2O3 on top of ceramic perovskite and other Ruddlesden-Popper structured electrode materials are sufficiently electron and oxygen ion conducting to provide reaction sites despite that the bulk phase of such an oxide layer is insulating. We claim that a few nanometer thin layer of mixed SrO-La2O3 that contains some dissolved transition metal and some impurities plus two space charge layers – one towards the gas phase and the other towards the perovskite – will be sufficiently oxide ion (vacancy) and electron conducting to support the electrode process. We also present some considerations about a possible mechanism of improved electrodes.

Catalytic Enhancement of Carbon Black and Coal-Fueled Hybrid Direct Carbon Fuel Cells

Hybrid direct carbon fuel cells (HDCFCs) consisting of a solid carbon (carbon black)-molten carbonate ((62–38 wt% Li-K)2CO3) mixtures in the anode chamber of an anode-supported solid oxide fuel cell type full-cell are tested for their electrochemical performance between 700 and 800°C. Performance was investigated using current-voltage-power density curves. In the anode chamber, catalysts are mixed with the carbon-carbonate mixture. These catalysts include various manganese oxides (MnO2, Mn2O3, Mn3O4, MnO), metal carbonates (Ag2CO3, MnCO3, Ce2(CO3)3), metals (Ag, Ce, Ni), doped-ceria (CeO2, Ce1-xGdxO2-x/2, Ce1-xREExO2-δ (REE = Pr, Sm)) and metal oxides (LiMn2O4, Ag2O). Materials showing the highest activity in carbon black (Mn2O3, CeO2, Ce0.6Pr0.4O2-δ, Ag2O) were subsequently tested for catalytic activity toward bituminous coal, as revealed by both I-V-P curves and electrochemical impedance spectroscopy (EIS). Catalytic activity was evaluated as a function of various physical characteristics of doped ceria and manganese-based materials.
Direct Coal Oxidation in Modified Solid Oxide Fuel Cells

Hybrid direct carbon fuel cells employ a classical solid oxide fuel cell together with carbon dispersed in a carbonate melt on the anode side. In a European project, the utilization of various coals has been investigated with and without addition of an oxidation catalyst to the carbon-carbonate slurry or anode layer. The nature of the coal affects both open circuit voltage and power output. Highest OCV and power densities were observed for bituminous coal and by adding manganese oxide or praseodymium-doped ceria to the carbon/carbonate mixture. Comparing the carbon black fueled performance of an anode supported (315 µm anodes) and cathode supported cell (15 µm anode) indicates a superior performance of the latter. Using un-catalyzed biomass (charcoal) as fuel results in an OCV of 941 mV and a maximum power density of 78 mW/cm² at 755ºC similar to the power output of manganese oxide catalyzed bituminous coal (73 mW/cm²).

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Effect of Co₃O₄ and Co₃O₄/CeO₂ infiltration on the catalytic and electro-catalytic activity of LSM₁₅/CGO₁₀ porous cells stacks for oxidation of propane

The objective of this work was to study the effect of CoO₄ and CoO₄/CeO₂ infiltration on the propene oxidation catalytic activity of a La₀.₈₅Sr₀.₁₅MnO₃/Co₀.₉Gd₀.₁O₁₅ electrochemical porous cell stack (11 layers, 5 single cells in series). The effect of the infiltration of Co₃O₄ and Co₃O₄/CeO₂ on the electrochemical properties of the porous cell stack was also investigated by electrochemical impedance spectroscopy (EIS). Co₃O₄ and Co₃O₄/CeO₂ exhibited high catalytic activity for propene oxidation. The increase of propene oxidation rate with +4 V (0.8 V/cell) polarization reached 10% for the Co₃O₄ infiltrated reactor and 48% of efficiency at 300 °C. The Co₃O₄/CeO₂ co-infiltration decreased the reactor polarization resistance, while Co₃O₄ infiltration had negligible effect on reactor electrochemical performance. The beneficial effect of CeO₂ on the electrode activity was attributed to the increased concentration of stable oxygen species on the electrode surface.

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Organisations: Department of Energy Conversion and Storage, Ceramic Engineering & Science, Fundamental Electrochemistry, Centro Nacional del Hidrogeno
Contributors: Deleebeeck, L., Gil, V., Ippolito, D., Campana, R., Kammer Hansen, K., Holtappels, P.
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Electrochemical reduction of NO with propene in the presence of oxygen on LSCoM/CGO porous cell stacks impregnated with BaO

The electrochemical reduction of NO with propene in the presence of 10 % O2 was studied on a La0.85Sr0.15Co0.03Mn0.97O3-Ce0.9Gd0.1O1.95 11-layer electrochemical reactor. BaO was impregnated into the porous reactor, and electrochemical impedance spectroscopy was used for characterisation in the temperature range of 300–400 °C. They were subjected to different magnitudes of polarisations, and the BaO impregnation was shown to have increased the NO reduction with increasing polarisation in the presence of excess oxygen. At 350 °C, more than 60 % was removed and a current efficiency for the cell stack of up to 30 % was achieved. In the presence of propene, the electrochemical promotion on the NOx conversion decreased and only 13 % was removed. A propene oxidation of 35 % was achieved as the highest at 400 °C. The propene acted as a reducing agent on the barium nitrates and caused a greater reduction of the nitrates and hereby releasing more NO than in the gas without propene. This caused an increase in NO in the outlet gas since not all of it was reduced further to nitrogen. The results indicate that the electrochemical reduction of the nitrates when no propene is present is a surface reaction, whereas the propene enables further reduction of the bulk nitrate.

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Contributors: Friedberg, A. Z., Kammer Hansen, K.
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Enhancing hybrid direct carbon fuel cell anode performance using Ag$_2$O

A hybrid-direct carbon fuel cell (HDCFC), consisting of a molten slurry of solid carbon black and (Li-K)2CO3 added to the anode chamber of a solid oxide fuel cell, was characterized using current-potential-power density curves, electrochemical impedance spectroscopy, and cyclic voltammetry. Two types of experimental setups were employed in this study, an anode-supported full cell configuration (two electrodes, two atmospheres setup) and a 3-electrode electrolyte-supported half-cell setup (single atmosphere). Anode processes with and without catalysts were investigated as a function of temperature (700-800 °C) and anode sweep gas (N2, 4-100% CO2 in N2-CO2). It was shown that the addition of silver based catalysts (Ag, Ag2O, Ag2CO3) into the carbon-carbonate slurry enhanced the performance of the HDCFC.
Fabrication of doped Titania (TiO2) nano-catalysts in the shape of nanofibers

Nanostructured materials have attracted incredible interest during the recent years for a large variety of applications. In heterogeneous catalysis the use of nano-sized catalytic materials is expected to significantly impact the performances of materials as consequence of their large surface-to-volume ratios [1]. The “nanomaterial” approach enables to achieve structures with incredible large exposed surface area. When nanofibers are used as nano-catalysts, the further advantage of a quite open porous structure is further achieved. In this work, nanomaterial approach was adopted to fabricate nano-catalysts for the removal of the NOx in exhausts via the NH3 Selective Catalytic Reduction method (SCR). The state-of-art system WO3-V2O5 was incorporated into TiO2 ceramic nanofibers through combination of electrospinning and sol-gel process. Catalytic functionalized nanofibers were characterized using SEM, TEM, EDX, BET, XRD, to analyze fiber diameter, morphology, composition, specific surface area, crystallization phases of TiO2 functionalized, atomic percentage of elements in fiber respectively. Catalytic activity was also measured.

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Source-ID: 106753942
Research output: Contribution to conference › Conference abstract for conference – Annual report year: 2015 › Research › peer-review

Hybrid direct carbon fuel cell anode processes investigated using a 3-electrode half-cell setup

A 3-electrode half-cell setup consisting of a yttria-stabilized zirconia (YSZ) electrolyte support was employed to investigate the chemical and electrochemical processes occurring in the vicinity of a model hybrid direct carbon fuel cell (HDCFC) anode (Ni-YSZ) in contact with a molten carbon-alkali carbonate slurry. Electrochemical testing, cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS), with and without the Ni-YSZ layer highlighted the promotional effect of the Ni-YSZ anode layer, and revealed the contributions of Ni/NiO, and potentially K/K2O, redox couple(s). Treated anthracite and bituminous coals, as well as carbon black, were tested, revealing similar open circuit potential and activation energies in mixed 96-4vol% N2-CO2 and 50-50vol% CO-CO2 environments between 700 and 800°C. Bituminous coal showed the highest activity, likely associated to a high O/C ratio and hydrogen content. Based on acquired data, a reaction scheme was proposed for processes at the working electrode, including the role of bubble formation in the vicinity of the electrochemically active solid/molten medium interface.

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Organisations: Department of Energy Conversion and Storage, Fundamental Electrochemistry, Instituto Nacional del Carbón
Contributors: Deleebeeck, L., Arenillas, A., Menendez, J., Kammer Hansen, K.
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Scopus rating (2015): CiteScore 3.46 SJR 1.33 SNIP 1.264
Hybrid Direct Carbon Fuel Cell Performance with Anode Current Collector Material

The influence of the current collector on the performance of a hybrid direct carbon fuel cell (HDCFC), consisting of solid oxide fuel cell (SOFC) with a molten carbonate-carbon slurry in contact with the anode, has been investigated using current-voltage curves. Four different anode current collectors were studied: Au, Ni, Ag, and Pt. It was shown that the performance of the direct carbon fuel cell (DCFC) is dependent on the current collector materials, Ni and Pt giving the best performance, due to their catalytic activity. Gold is suggested to be the best material as an inert current collector, due to its low catalytic activity.

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Contributors: Deleebeeck, L., Kammer Hansen, K.
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Research output: Contribution to journal → Journal article – Annual report year: 2016 → Research → peer-review

In Situ Studies of Fe4+ Stability in β-Li3Fe2(PO4)3 Cathodes for Li Ion Batteries

In commercial Fe-based batteries the Fe2+/Fe3+ oxidation states are used, however by also utilizing the Fe4+ oxidation state, intercalation of up to two Li ions per Fe ion could be possible. In this study, we investigate whether Fe4+ can be formed and stabilized in β-Li3Fe2(PO4)3. The work includes in situ synchrotron X-ray powder diffraction studies (XRPD) during charging of β-Li3Fe2(PO4)3 up to 5.0 V vs. Li/Li+. A novel capillary-based micro battery cell for in situ XRPD has been designed for this. During charge, a plateau at 4.5 V was found and a small contraction in volume was observed, indicating some Li ion extraction. The volume change of the rhombohedral unit cell is anisotropic, with a decrease in the a parameter and an increase in the c parameter during the Li ion extraction. Unfortunately, no increased discharge capacity was observed and Mössbauer spectroscopy showed no evidence of Fe4+ formation. Oxidation of the organic electrolyte is inevitable at 4.5 V but this alone cannot explain the volume change. Instead, a reversible oxygen redox process (O2→ O−) could possibly explain and charge compensate for the reversible extraction of lithium ions from β-Li3Fe2(PO4)3.

General information
Publication status: Published
Organisations: Department of Energy Conversion and Storage, Atomic Scale Materials Modelling, Department of Physics, Neutrons and X-rays for Materials Physics, Applied Electrochemistry, Fundamental Electrochemistry
Method and system for the purification of exhaust gas with an electrochemical cell
The present invention relates to a method for electrochemical reduction of nitrogen oxides and concomitant oxidation of soot, as well as systems useful therefor. Such methods and systems in particular are useful in the context of exhaust gas purification, in particular for diesel engines.

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Organisations: Department of Energy Conversion and Storage, Fundamental Electrochemistry
Contributors: Kammer Hansen, K., Mogensen, M. B., Shao, J.
Publication date: 2015

Publication information
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Nano scaled electro catalysts, a versatile concept for novel solid state fuel cells and electro-catalytic reactors

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Organisations: Department of Energy Conversion and Storage, Fundamental Electrochemistry, Ceramic Engineering & Science
Contributors: Kammer Hansen, K., Holtappels, P., Ramos, T., Sudireddy, B. R., Traulsen, M. L.
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Place of publication: Lyngby
**NOx Conversion of Porous LSF15-CGO10 Cell Stacks**

A porous electrochemical reactor, made of La0.85Sr0.15FeO3 as electrode and Ce0.8Pr0.2O2-d as electrolyte, was studied for the electrochemical reduction of NO with Propene. In order to enhance the effect of polarization, the reactor was impregnated with Ce0.8Gd0.2O1.95, CeO2 or Ce0.8Pr0.2O2-d nanoparticles. The HC-SCR on the cells was increased on the impregnated cells, but no electrochemical enhancement of this was observed. The applied overpotential on the impregnated cells changed the oxidation reaction of NO into NO2 which is considered an intermediate in the NO reduction to nitrogen.

**Catalytic Enhancement of Solid Carbon Oxidation in HDCFCs**

Hybrid direct carbon fuel cells consisting of a solid carbon (carbon black)-molten carbonate ((62-38 wt% Li-K)(2)CO3) mixtures in the anode chamber of an anode-supported solid oxide fuel cell type full-cell are tested for their electrochemical performance between 700 and 800 degrees C. Performance was investigated using current-potential-power density curves. In the anode chamber, catalysts are mixed with the carbon-carbonate mixture. These catalysts include various manganese oxides (MnO2, Mn2O3, and Mn3O4) and dopedceria (CeO2, Ce1-xGdxO2-x/2, Ce1-xRExO2-delta (RE = Pr, Sm)), the effectiveness of these families of catalysts are discussed with respect to electrochemical, chemical and post-mortem analysis.
Catalytic Enhancement of Solid Carbon Oxidation in HDCFCs

Hybrid direct carbon fuel cells (HDCFCs) consisting of a solid carbon (carbon black)-molten carbonate ((62-38 wt% Li-K)2CO3) mixtures in the anode chamber of an anode-supported solid oxide fuel cell (SOFC)-type full-cell (NiO-yttria-stablized zirconia (YSZ)|YSZ|lanthanum strontium manganite (LSM)-YSZ/LSM) are tested for their electrochemical performance between 700 and 800°C. Performance was investigated using electrochemical impedance spectroscopy (EIS) and current-potential-power density curves. EIS data is interpreted using a model circuit (R-RQ-RQ-RQ), and the dominant processes revealed by the impedance data as a function of temperature, anode and cathode atmospheres, and their flow rates are discussed. In the anode chamber, catalysts are mixed with the carbon-carbonate mixture. These catalysts include various manganese oxides (MnO2, Mn2O3, and Mn3O4, Fig. 1) and doped-ceria (CeO2, Ce1-xGdxO2, Ce1-xRExO2 (RE = Pr, Gd, Sm, etc.)), the effectiveness of these families of catalysts are discussed with respect to electrochemical, chemical and post-mortem analysis. Fig. 1. Current-potential-power density curves acquired for a blank (SiC) and manganese oxide (MnO2, Mn2O3, Mn3O4) catalysts suspended in the carbon-carbonate mixture in the anode chamber of an HDCFC. 96-4 vol% N2-CO2 (anode), air (cathode), 755°C, 0-600 mA, 50 mA/step. Power density corrected to cathode geometric surface area. [Formula]

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Contributors: Deleebeeck, L., Ippolito, D., Kammer Hansen, K.
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Research output: Contribution to journal › Conference abstract in journal – Annual report year: 2014 › Research › peer-review

Effect of CeO2 Infiltration on Hybrid Direct Carbon Fuel Cell Performance

The effect of CeO2 infiltration into the anode or CeO2 mixed with the carbon-fuel on the performance of a Hybrid Direct Carbon Fuel Cell (HDCFC) was studied through the use of polarization curves and electrochemical impedance spectroscopy. The use CeO2 in both ways helped to increase the cell performance. In particular, mixing CeO2 with carbon represents the best strategy to increase the cell power output.

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Contributors: Ippolito, D., Deleebeeck, L., Kammer Hansen, K.
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Publication information
Effect of Co3O4 and CeO2 Infiltration on the Activity of a LSM15/GDC10 Highly Porous Electrochemical Reactor

The reduction of air pollution has become an international concern over the last ten years because of increases in emissions from mobile and stationary sources. Among these sources, volatile organic compounds (VOC) represent a serious environmental problem, together with NOx, SOx and particulate matter, which lead to the formation of ozone in urban and regional areas [1]. The electrocatalytic activity of a porous electrochemical reactor, made of La0.85Sr0.15MnO3±δ (LSM) as electrode and Ce0.9Gd0.1O1.95 (GDC) as electrolyte, was studied for the electrochemical oxidation of propene (C3H6), a major VOC component of Diesel engine exhausts, over a wide range of temperatures. The porous reactor was thought as a highly porous catalytic filter for a possible application in a Diesel exhausts purification system. The porous reactor was used as a backbone for the infiltration of Co3O4 and Co3O4/CeO2. The effect of the infiltration on the electrochemical properties and catalytic activity of the reactor was investigated by electrochemical impedance spectroscopy (EIS) and gas analysis. Figure 1 shows the SEM cross-section micrograph of the electrochemical reactor made of 11 alternating layers of electrode (LSM) and electrolyte (GDC). Figure 2 shows the Nyquist plot of the impedance spectra of Co3O4 infiltrated backbone recorded at OCP with 10% O2 and 10% O2 + 1000 ppm C3H6, 2 L/h, 400 °C. [1] R. Atkinson, Atmospheric Chemistry of VOCs and NOx, Atmos Environ. 34 (2000) 2063.

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Electrochemical Reduction of Oxygen and Nitric Oxide at Low Temperature on La$_{1-x}$Sr$_x$FeO$_{3-\delta}$ Cathodes

A series of six strontium-substituted lanthanum ferrites (La$_{1-x}$Sr$_x$FeO$_{3-\delta}$, $x = 0.00, 0.05, 0.15, 0.25, 0.35, \text{ and } 0.50$) were synthesized using the glycine-nitrate process and evaluated as cathodes for the electrochemical reduction of oxygen and nitric oxide in the temperature range 200 to 400 degrees C, using cone-shaped electrodes and cyclic voltammetry. It was shown that the ferrites had a higher activity towards the electrochemical reduction of nitric oxide than towards the electrochemical reduction of oxygen, in the investigated temperature range. The highest activity towards the electrochemical reduction of nitric oxide was found for La$_{0.95}$Sr$_{0.05}$FeO$_{3-\delta}$ at 400 degrees C. This compound also showed the highest activity towards the electrochemical reduction of oxygen at 400 degrees C. The highest apparent selectivity was found for the compound LaFeO$_3$ at 200 degrees C. The materials showed ability to oxidize nitric oxide to nitrogen dioxide.

Electrochemical Reduction of Oxygen and Nitric Oxide at Low Temperature on La$_{1-x}$Sr$_x$FeO$_{3-\delta}$ Cathodes

A series of six strontium-substituted lanthanum ferrites (La$_{1-x}$Sr$_x$FeO$_{3-\delta}$, $x = 0.00, 0.05, 0.15, 0.25, 0.35, \text{ and } 0.50$) were synthesized using the glycine-nitrate process and evaluated as cathodes for the electrochemical reduction of oxygen and nitric oxide in the temperature range 200 to 400 degrees C, using cone-shaped electrodes and cyclic voltammetry. It was shown that the ferrites had a higher activity towards the electrochemical reduction of nitric oxide than towards the electrochemical reduction of oxygen, in the investigated temperature range. The highest activity towards the electrochemical reduction of nitric oxide was found for La$_{0.95}$Sr$_{0.05}$FeO$_{3-\delta}$ at 400 degrees C. This compound also showed the highest activity towards the electrochemical reduction of oxygen at 400 degrees C. The highest apparent selectivity was found for the compound LaFeO$_3$ at 200 degrees C. The materials showed ability to oxidize nitric oxide to nitrogen dioxide.
HDCFC Performance as a Function of Anode Atmosphere (N2-CO2)
The mechanism for the oxidation of solid carbon in a fuel cell made as a hybrid between a molten carbonate fuel cell and a solid oxide fuel cell, known as a hybrid direct carbon fuel cell (HDCFC), was investigated. Fuel cell performance was measured by electrochemical impedance spectroscopy (EIS) and using current-potential-power density curves (I-V-P) between 700 to 800 degrees C. The impacts of the gas species introduced at the cathode (air vs. pure O2) and at the anode (pure N2, pure CO2, and mixed N2-CO2) were investigated, as well as the influences of temperature and anode gas flow rate. The majority of the impedance data could be modeled using an equivalent circuit consisting of a resistor (Rs) in series with three resistor-constant phase element units (RQ, in parallel), depending on anode gas atmosphere. An explanation was proposed for each impedance element, and the literature relating to impedance data acquired for carbon-carbonate mixture in a DCFC anode were discussed. By varying of the anode gas mixtures between pure N2 and pure CO2, together with variations in their flow rates, it was suggested that CO2 is a chemically active species which is not electrochemically active, a chemical intermediate in the oxidation of solid carbon in such a HDCFC. (C) 2013 The Electrochemical Society.

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High Performance Infiltrated Backbones for Cathode-Supported SOFC's
A four-step infiltration method has been developed to infiltrate La0.75Sr0.25MnO3+δ (LSM25) nanoparticles into porous structures (YSZ or LSM-YSZ backbones). The pore size distribution in the backbones is obtained either by using PMMA and/or graphites as pore formers or by leaching treatment of samples with Ni remained in the YSZ structure at high temperatures. All impregnated backbones, presented Rs comparable to a standard screen printed cathode, which proves that LSM nanoparticles forms a pathway for electron conduction.

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Publication information
High Performance Infiltrated Backbones for Cathode-Supported SOFC's

The concept of using highly ionic conducting backbones with subsequent infiltration of electronically conducting particles has widely been used to develop alternative anode-supported SOFC's. In this work, the idea was to develop infiltrated backbones as an alternative design based on cathode-supported SOFC. The cathodes are obtained by infiltrating LSM into a sintered either thick (300 μm) ytria stabilized zirconia (YSZ) backbone or a thin YSZ backbone (10-15 μm) integrated onto a thick (300 μm) porous strontium substituted lanthanum manganite (LSM) and YSZ composite.

Fabrication challenges, microstructural characterization and electrochemical testing are discussed. Data on polarization resistance, Rp, are obtained from impedance spectra recorded on quasi-symmetrical cells (YSZ backbones/YSZ/LSM-YSZ (screen printed)). The backbones are infiltrated with LSM and compared to a standard LSM-YSZ screen printed symmetrical cells. Samples with LSM/YSZ composite and YSZ backbones made with graphite+PMMA as pore formers exhibited comparable Rp values to the screen printed LSM/YSZ cathode. This route was chosen as the best to fabricate the cathode supported cells. SEM micrograph of a cathode supported cell with infiltrated LSM nanoparticles is shown in Fig. 1. Figure 1. Cross section of LSM infiltrated cathode supported cell. [Formula]
system design (continuous supply of solid fuel), performance (power density, efficiency), environmental burden (fresh water consumed, solid waste produced, CO2 emitted, ease of combination with CCS) and economics (levelized cost of electricity).

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**Impedance Spectroscopy and Catalytic Activity Characterization of a La_{0.85}Sr_{0.15}MnO_3/Ce_{0.9}Gd_{0.1}O_1.95 Porous Electrochemical Reactor for the Oxidation of Propene**

This study aims to characterize the catalytic and electrochemical behavior of a La_{0.85}Sr_{0.15}MnO_3/Ce_{0.9}Gd_{0.1}O_1.95 porous reactor for the oxidation of propene in the presence of oxygen. The application of anodic polarization strongly increased the propene oxidation rate up to 71 %, although the current efficiency remained low. The effect of prolonged polarization on the reactor catalytic activity was evaluated. Prolonged polarization enhanced both the reactor intrinsic catalytic activity and the electrode performance due to the formation of oxygen vacancies on the electrode surface. Electrochemical impedance spectroscopy was used to investigate the effect of propene introduction on the reactor impedance response. The introduction of propene into reactive system caused a strong increase of electrode resistance, mainly located in the low-frequency region of the impedance spectrum. This effect was caused by the strong adsorption of propene on electrode surfaces inhibiting the adsorption and dissociation of oxygen.

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**Removal of NO\textsubscript{x} with Porous Cell Stacks with La\textsubscript{0.85}Sr\textsubscript{0.15}Co\textsubscript{x}Mn\textsubscript{1-x}O\textsubscript{3-\delta}\textsuperscript{+\textdelta}Ce\textsubscript{0.9}Gd\textsubscript{0.1}O\textsubscript{1.95} Electrodes Infiltrated with BaO**

Porous cell stacks with composite electrodes of La\textsubscript{0.85}Sr\textsubscript{0.15}Co\textsubscript{x}Mn\textsubscript{1-x}O\textsubscript{3-\delta}\textsuperscript{+\textdelta}Ce\textsubscript{0.9}Gd\textsubscript{0.1}O\textsubscript{1.95} were tested for activity toward selective electrochemical reduction of NO\textsubscript{x} to N\textsubscript{2} in the presence of 10% O\textsubscript{2}. The cell stacks were produced by tape casting, laminating and sintering the backbone structure followed by infiltration with BaO or La\textsubscript{0.85}Sr\textsubscript{0.15}MnO\textsubscript{3-\delta}. The cell stacks were tested in an atmosphere of 1000 ppm NO or NO\textsubscript{2} + 10% O\textsubscript{2} in Ar with 10% O\textsubscript{2} in Ar as reference, and in the temperature range of 250 to 500 °C. The cell stacks were investigated electrochemically with cyclic voltammetry and polarization, and the outlet gas composition was monitored. Doping with Co increases the electrodes activity for reducing NO\textsubscript{x}, and up to 93% conversion in the presence of 10% O\textsubscript{2} at 400 °C was measured. Infiltration with BaO was necessary to achieve any formation of N\textsubscript{2}. Square wave polarization increased the activity of the electrodes achieving a current efficiency as high as 20% at 350 °C, and this indicates a problem with mass transfer. An atmosphere containing NO\textsubscript{2} also increased the activity of the electrodes, which points to a mechanism with a NO\textsubscript{2} related intermediate. © 2014 The Electrochemical Society. All rights reserved.

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**A combined SEM, CV and EIS study of multi-layered porous ceramic reactors for flue gas purification**

The effect of sintering temperature of 12-layered porous ceramic reactors (comprising 5 cells) was studied using scanning electron microscopy (SEM), cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS). The difference in microstructures of the reactors was evaluated by SEM. Additional information on the influence of sintering temperature on the properties of the reactors could be gained by the use of EIS. The present work has provided the first set of fundamental electrochemical data and their interpretation in terms of fabrication conditions, for the multi-layered porous ceramic reactors.

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Characterization of LSM/CGO Symmetric Cells Modified by NOx Adsorbents for Electrochemical NOx Removal with Impedance Spectroscopy

This study uses electrochemical impedance spectroscopy (EIS) to characterize an LSM/CGO symmetric cell modified by NOx adsorbents for the application of electrochemical NOx reduction. Three cells were prepared and tested: a blank cell, a cell impregnated with BaO, and a cell coated with a BaO-Pt-Al2O3 layer. The impedance analysis revealed that modification with the NOx adsorbents, either by impregnating the BaO into the electrode or by adding a BaO-Pt-Al2O3 layer on top of the electrode significantly enhanced the electrode activity. This activity enhancement was mainly due to the decrease in the resistance of the low-frequency processes, which were ascribed to adsorption, diffusion, and transfer of O2 species and NOx species at or near the triple phase boundary (TPB) region and the formation of the reaction intermediate NO2. The BaO impregnation improved the adsorption of NOx on the LSM/CGO electrode by selectively trapping NO2 in the form of nitrate over the BaO sites and provided availability for a direct reduction of the stored nitrate. The BaO-Pt-Al2O3 layer enhanced the NOx adsorption and promoted the formation of NO2 due to the NO oxidation ability of the Pt catalyst, but hindered the gas diffusion to the reaction sites.

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Contributors: Shao, J., Kammer Hansen, K.
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Effect of Infiltration Material on a LSM\textsubscript{15}/CGO\textsubscript{10} Electrochemical Reactor in the Electrochemical Oxidation of Propene

The effect of infiltrating on a La\textsubscript{0.85}Sr\textsubscript{0.15}MnO\textsubscript{3}/Ce\textsubscript{0.9}Gd\textsubscript{0.1}O\textsubscript{1.95} 11-layer electrochemical reactor with CeO\textsubscript{2} and Ce\textsubscript{0.8}Pr\textsubscript{0.2}O\textsubscript{2−δ} was studied in propene oxidation at open-circuit voltage and under polarization as a function of reaction temperature. This work outlined the importance of catalytic and electrochemical properties of infiltrated material on the ability to increase propene conversion under polarization with good faradaic efficiency. Electrochemical impedance spectroscopy was used to study the effect of infiltration material on electrode properties. The infiltration of a mixed ionic and electronic conductor, like Ce\textsubscript{0.8}Pr\textsubscript{0.2}O\textsubscript{2−δ}, increased the electrode performance at low temperature but decreased the lifetime of the oxygen ion promoters on the catalyst/electrode surface, reducing the faradaic efficiency of the reaction. The infiltration of CeO\textsubscript{2} provided high propene conversion at open circuit and high effect of polarization associated with good faradaic efficiency, especially at low temperature.

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Electrochemical NO\textsubscript{x} reduction on an LSM/CGO symmetric cell modified by NO\textsubscript{x} adsorbents
This study investigated the effect of modifying a (La\textsubscript{0.85}Sr\textsubscript{0.15})\textsubscript{0.99}MnO\textsubscript{3} (LSM)/Ce\textsubscript{0.9}Gd\textsubscript{0.1}O\textsubscript{1.95} (CGO) symmetric cell by NO\textsubscript{x} adsorbents on the electrochemical reduction of NO\textsubscript{x} under O\textsubscript{2}-rich conditions. The modification was based on a full ceramic cell structure without any noble metals. Three cells were prepared and tested: a blank cell, a cell impregnated with BaO, and a cell coated with a BaO/Pt/Al\textsubscript{2}O\textsubscript{3} layer. The electrochemical reduction of NO\textsubscript{x} on the three cells was studied by conversion measurement, degradation testing, and microstructure characterization. The modification, either by impregnating the BaO into the electrode or by coating the Ba/Pt/Al\textsubscript{2}O\textsubscript{3} layer on the surface of the electrode, significantly increased the activity and selectivity of the NO\textsubscript{x} reduction on the LSM/CGO symmetric cell by enhancing the adsorption and storage of the NO\textsubscript{x} species or providing reaction sites for direct nitrate reduction. The cell with the BaO/Pt/Al\textsubscript{2}O\textsubscript{3} layer exhibited a preferable performance at low temperatures (350 and 400 °C) and low voltages (1.5 to 2 V) due to the NO oxidation ability of the Pt catalyst, although its performance was relatively poor at elevated temperatures and voltages due to the impedance of the diffusion of NO\textsubscript{x} to the reaction sites by the adsorption layer. For lowering the operation temperature and minimizing the power consumption, adding an adsorption layer was shown to be the optimum approach for modifying the electrochemical cell by NO\textsubscript{x} adsorbents. The square wave (SV) polarization can balance the trapping and reduction rates of NO\textsubscript{x} species on the electrochemical cells to further improve the NO\textsubscript{x} reduction relative to the direct current (DC) polarization.

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Contributors: Shao, J., Kammer Hansen, K.
Pages: 7137-7146
Electrochemical reduction of oxygen and nitric oxide at low temperature on Ce$_{1-x}$Pr$_x$O$_{2-\delta}$ cathodes

The ability of praseodymium doped cerium oxide materials to electrochemically reduce NO and O$_2$ was studied using cone-shaped electrodes in conjunction with cyclic voltammetry, in the temperature range 200–400 °C. Four samples were studied; Ce$_{1-x}$Pr$_x$O$_{2-\delta}$ (x = 0.1, 0.2, 0.3 and 0.4). It was shown that the current densities in both NO and O$_2$ gases increased with increasing praseodymium doping. Ce$_{1-x}$Pr$_x$O$_{2-\delta}$ had higher activity in an atmosphere containing NO than in an atmosphere containing O$_2$ in most cases and this trend was stronger at the lower temperatures. Ce$_{0.8}$Pr$_{0.2}$O$_{2-\delta}$ and Ce$_{0.9}$Pr$_{0.1}$O$_{2-\delta}$ seemed to have the highest ratio of maximum cathodic current density (i$_{NO}$/i$_{O2}$), which is used as an indication of a higher activity toward reduction of NO compared to reduction of O$_2$. The apparent selectivity generally decreased with increasing temperature for all the compositions.

General information

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Organisations: Department of Energy Conversion and Storage, Fundamental Electrochemistry
Contributors: Werchmeister, R. M. L., Kammer Hansen, K.
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Electrochemical Reduction of Oxygen and Nitric oxide at low Temperature on La$_{1-x}$Sr$_x$Cr$_{0.97}$V$_{0.03}$O$_{3-\delta}$ Cathodes

Five La$_{1-x}$Sr$_x$Cr$_{0.97}$V$_{0.03}$O$_{3-\delta}$ (x = 0, 0.05, 0.15, 0.25, 0.35) perovskites were synthesized and characterized by powder XRD and cyclic voltammetry on cone-shaped electrodes in 10% oxygen in argon or 1% nitric oxide in argon at 200, 300 and 400°C. It was shown that the activation energy for the reduction of oxygen is higher than the activation energy for the reduction nitric oxide. The activity for the reduction of both oxygen and nitric oxide was shown to be highest for the end member La$_0.65$Sr$_{0.35}$Cr$_{0.97}$V$_{0.03}$O$_{3-\delta}$. The highest ratio between the current densities in the nitric oxide and oxygen containing atmospheres was found for the end member La$_0.65$Sr$_{0.35}$Cr$_{0.97}$V$_{0.03}$O$_{3-\delta}$. The chromites also showed activity as anodes for either oxygen evolution or oxidation of nitric oxide to nitrogen dioxide.
Electrochemical reduction of oxygen and nitric oxide at low temperature on La$_{1-x}$Sr$_x$MnO$_{3+δ}$ cathodes
Six La$_{1-x}$Sr$_x$MnO$_{3+δ}$ ($x=0, 0.05, 0.15, 0.25, 0.35, 0.50$) perovskites were synthesised and characterised by powder XRD and cyclic voltammetry on cone-shaped electrodes in 10% oxygen in argon or 1% nitric oxide in argon at 200, 300 and 400°C. The activity of the manganite based perovskites were highest in the nitric oxide containing atmosphere compared to the activity in the oxygen containing atmosphere at 200°C. At 300 and 400°C the activity in the nitric oxide and oxygen containing atmospheres were similar. The highest ratio between the cathodic current densities in the nitric oxide and oxygen containing atmospheres was found for the La$_{0.95}$Sr$_{0.05}$MnO$_{3+δ}$ perovskite at 200°C.
Enhancement of NO\textsubscript{x} removal performance for \((\text{La}_{0.85}\text{Sr}_{0.15})_{0.99}\text{MnO}_{3} / \text{Ce}_{0.9}\text{Gd}_{0.1}\text{O}_{1.95})\) electrochemical cells by NO\textsubscript{x} storage/reduction adsorption layers

This study investigated the effect of adding a NO\textsubscript{x} adsorption layer to the cathode of an electrochemical cell on the removal of NO\textsubscript{x} from gaseous mixtures. The cathode was a composite of \((\text{La}_{0.85}\text{Sr}_{0.15})_{0.99}\text{MnO}_{3}\) (LSM15) and \text{Ce}_{0.9}\text{Gd}_{0.1}\text{O}_{1.95} (CGO10). Two different kinds of adsorption layers, K–Pt–Al\textsubscript{2}O\textsubscript{3} layer and Ba–Pt–Al\textsubscript{2}O\textsubscript{3} layer (known as NO\textsubscript{x} storage/reduction (NSR) catalyst), were studied. The effects of the NSR adsorption layers on the electrode processes were characterized by electrochemical impedance spectroscopy (EIS). Both adsorption layers increased the reduction of NO\textsubscript{x} to N\textsubscript{2} in an atmosphere that contained only NO. When O\textsubscript{2} was present with NO in the atmosphere, the K–Pt–Al\textsubscript{2}O\textsubscript{3} adsorption layer significantly enhanced the conversion of NO\textsubscript{x} to N\textsubscript{2}, but the Ba–Pt–Al\textsubscript{2}O\textsubscript{3} adsorption layer had no effect. The selective removal of NO\textsubscript{x} under O\textsubscript{2}-rich conditions was achieved by modifying the LSM15/CGO10 cell with a suitable NSR adsorption layer. The improvement for NO\textsubscript{x} reduction by the adsorption layers was mainly contributed by the promotion of the adsorption and surface diffusion of NO\textsubscript{x} species at/near the triple phase boundary (TPB) regions of the electrode and probably the formation of a short and effective reaction path for NO\textsubscript{x} reduction. A stronger capability for oxidizing NO and/or trapping NO\textsubscript{x} under the test conditions may have contributed to the superior performance of the K–Pt–Al\textsubscript{2}O\textsubscript{3} adsorption layer relative to the Ba–Pt–Al\textsubscript{2}O\textsubscript{3} layer. © 2012 Elsevier Ltd. All rights reserved.

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Fabrication and Characterization of multi-layer ceramics for electrochemical flue gas purification

Multi-layered ceramics for electrochemical flue gas purification were fabricated by tape casting and lamination or by multi-layer tape casting. The sintered bodies were studied by scanning electron microscopy and electrochemical impedance spectroscopy. It was shown that the samples made by multi-layer tape casting had a lower resistance than the laminated samples. This was mainly due to a decrease of the resistance of the middle frequency arc, attributed to charge transfer close to the triple phase boundary. The cells showed ability to oxidize propene both under open circuit voltage and when polarized.

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Publication information
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Volume: 160
Fabrication of highly porous LSM/CGO cell stacks for electrochemical flue gas purification

In this study porous cell stacks for electrochemical flue gas purification were fabricated using tape casting and lamination followed by sintering. Two different mixtures of pore formers were used; either a mixture of two types of graphite or a mixture of graphite with polymethyl methacrylate micro-particles. It was shown that the porous cell stacks fabricated with polymethyl methacrylate had a higher porosity but a similar back pressure compared to the porous cell stacks fabricated with only graphite as a pore former. This was due to a high back pressure of the electrolyte layer. The porous cell stacks fabricated with polymethyl methacrylate as a pore former seem to be well suited for i.e. caption of soot particles. Furthermore, the back pressure of the electrode layer was significantly reduced when using polymethyl methacrylate pore formers. However, a better interconnectivity of the pores formed by the polymethyl methacrylate pore former, especially in the electrolyte layer, is needed, in order to lower the back pressure of the porous cell stack.

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

General information
Publication status: Published
Organisations: Department of Energy Conversion and Storage, Atomic Scale Materials Modelling, Applied Electrochemistry, Department of Physics, Experimental Surface and Nanomaterials Physics, Fundamental Electrochemistry
Contributors: Christiansen, A. S., Johnsen, R., Norby, P., Jensen, S. H., Frandsen, C., Mørup, S., Kammer Hansen, K., Holtappels, P.
NOx reduction on Ag electrochemical cells with a K-Pt-Al2O3 adsorption layer

Ag electrochemical cells with and without a K-Pt-Al2O3 adsorption layer were tested for NOx reduction under oxygen-rich conditions. The effect of the addition of the adsorption layer on the electrochemical reduction of NOx was investigated by a conversion measurement, an impedance analysis and a microstructure characterization. The blank Ag cell was incapable of converting NOx to N2 under any of the investigated conditions. In contrast, the Ag cell with an adsorption layer showed good NOx reduction activity. An 82% NOx conversion with 100% N2 selectivity and 7.7% current efficiency was achieved at -1.25 V and 500°C. An impedance analysis revealed that the adsorption layer promoted the adsorption and the surface diffusion of the NOx species at or near the triple phase boundaries (TPBs) and the formation of NO2. A severe degradation was also observed on the cell with the adsorption layer, which was caused by the corrosion of the Ag cathode and the subsequent migration of the Ag into the adsorption layer during the operation. © 2013 The Electrochemical Society.

Production of a half cell with a LSM/CGO support for electrochemical flue gas purification

Described herein is the production of a half cell with a strontium-substituted lanthanum manganite/cerium gadolinium oxide support and dense cerium gadolinium oxide electrolyte for electrochemical flue gas purification. The half cells were constructed through tape casting a strontium-substituted lanthanum manganite/cerium gadolinium oxide support and cerium gadolinium oxide electrolyte. The half cells were produced by laminating the support and electrolyte layers followed by sintering. Perfectly flat half cells were constructed with a porous strontium-substituted lanthanum manganite/cerium gadolinium oxide support layer and dense cerium gadolinium oxide electrolyte by adjusting sintering shrinkage at the electrolyte layer and altering the sintering aid.
A combined SEM and CV Study of Solid Oxide Fuel Cell Interconnect Steels

Scanning electron microscopy and cyclic voltammetry were used to investigate the high temperature oxidation behavior of two solid oxide fuel cell interconnect steels. One alloy had a low content of manganese; the other alloy had a high content of manganese. Four reduction and four oxidation peaks were observed in the voltammograms of the low manganese alloy at a temperature of 800 °C. The voltammograms of the alloy with a high content of manganese were different. At 600 °C three reduction peaks and two oxidation peaks were observed. At 800 °C additional peaks were observed in the voltammogram for this alloy.

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Organisations: Risø National Laboratory for Sustainable Energy, Fuel Cells and Solid State Chemistry Division, Electrochemistry, Electroceramics
Contributors: Kammer Hansen, K., Ofoegbu, S., Mikkelsen, L.
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**Diffuse Reflectance Infrared Fourier Transform Study of NOx Adsorption on CGO10 Impregnated with K2O or BaO**

In the present work, Diffuse Reflectance Infrared Fourier Transform (DRIFT) spectroscopy is applied to study the adsorption of NOx at 300-500 °C in different atmospheres on gadolinium doped ceria (CGO), an important material in electrodes investigated for electrochemical NOx removal. Furthermore, the effect on the NOx adsorption when adding K2O or BaO to the CGO is investigated. The DRIFT study shows mainly the presence of nitrate species at 500 °C, while at lower temperature a diversity of adsorbed NOx species exists on the CGO. Presence of O2 is shown to have a strong effect on the adsorption of NO, but no effect on the adsorption of NO2. Addition of K2O and BaO dramatically affects the NOx adsorption and the results also show that the adsorbed NOx species are mobile and capable of changing adsorption state in the investigated temperature range.

**General information**

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- Web of Science (2012): Indexed yes
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Keywords: NOx removal, NOx reduction, Electrochemical deNOx, FTIR
DOIs: 10.1021/jp300608d

**Electrochemical oxidation of propene by use of LSM15/CGO10 electrochemical reactor**

The propene catalytic oxidation was studied over an 11-layers porous electrochemical reactor made by La0.85Sr0.15MnO3 and Ce0.9Gd0.1O1.95 with the objective to simulate the abatement of exhaust gases emitted from Diesel engines. This work shows the possibility to enhance the catalytic activity through infiltration of Ce0.9Gd0.1O1.95 using the porous electrochemical reactor as a catalyst support.

The infiltration of an oxygen ion conductor as Ce0.9Gd0.1O1.95 showed an increased activity either at open circuit voltage (OCV) or under polarization with respect to the non infiltrated cell. The use of infiltration permitted to maintain high the effect of polarization also at low temperatures. The electrode was found to be strongly affected by morphology of infiltrated material and long term polarization brought about to an increase of catalytic activity. Moreover the phenomenon of electrochemical promotion of catalysis (EPOC) was found at low temperature.

**General information**

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**Publication information**

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Volume: 159
Electrochemical testing of composite electrodes of (La1−x Sr x )sMnO3 and doped ceria in NO-containing atmosphere

The possibility of using electrochemical cells for removal of NOx from an exhaust gas with excess O2 has been examined. (La1−x Sr x )s MnO3 (LSM) and ceria doped with Pr or Gd were selected as electrode materials and investigated in three-electrode cells. The electrodes were characterised electrochemically with electrochemical impedance spectroscopy (EIS) and cyclic voltammetry (CV), and the gas composition monitored while the electrodes were polarised. The electrodes of (La0.5Sr0.5)0.99MnO3 (LSM50) and Ce0.8Pr0.2O2−δ exhibit higher current densities in 0.1% NO in Ar than in air at 300 to 400 °C during CV. This indicates some apparent selectivity towards NO compared to O2. The electrodes can remove NO, when polarised to at least −0.6 V vs. Pt/Air at 600 °C, and EIS measurements under polarisation indicate that the kinetics of the electrodes change, when the electrode potential gets below −0.6 V vs. Pt/Air.

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NOx conversion on LSM15-CGO10 cell stacks with BaO impregnation

The electrochemical conversion of NOx on non-impregnated and BaO-impregnated LSM15-CGO10 (La0.85Sr0.15MnO3-Ce0.9Gd0.101.95) porous cell stacks has been investigated, and extensive impedance analysis have been performed to identify the effect of the BaO on the electrode processes. The investigation was conducted in the temperature range 300-500 degrees C, a polarisation range from 3 V to 9 V and in atmospheres containing 1000 ppm NO, 1000 ppm NO + 10%...
O-2 and 10% O-2. On the non-impregnated cell stacks no NOx conversion was observed under any of the investigated conditions. However, BaO impregnation greatly enhanced the NOx conversion and at 400 degrees C and 9 V polarisation a BaO-impregnated cell stack showed 60% NOx conversion into N-2 with 8% current efficiency in 1000 ppm NO + 10% O-2. This demonstrates high NOx conversion can be achieved on an entirely ceramic cell without expensive noble metals. Furthermore the NOx conversion and current efficiency was shown to be strongly dependent on temperature and polarisation. The impedance analysis revealed that the BaO-impregnation increased the overall activity of the cell stacks, but also changed the adsorption state of NOx on the electrodes; whether the increased activity or the changed adsorption state is mainly responsible for the improved NOx conversion remains unknown.

General information
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NOx-conversion on Porous LSF15-CGO10 Cell Stacks with KNO3 or K2O Impregnation
In the present work, it was investigated how addition of KNO3 or K2O affected the NOx conversion on LSF15–CGO10 (La0.85Sr15FeO3–Ce0.9Gd0.1O1.95) composite electrodes during polarization. The LSF15–CGO10 electrodes were part of a porous 11-layer cell stack with alternating layers of LSF15–CGO10 electrodes and CGO10 electrolyte. The KNO3 was added to the electrodes by impregnation and kept either as KNO3 in the electrode or thermally decomposed into K2O before testing. The cell stacks were tested in the temperature range 300–500 °C in 1,000 ppm NO, 10% O2, and 1,000 ppm NO+10% O2. During testing, the cells were characterized by electrochemical impedance spectroscopy, and the NO conversion was measured during polarization at −3 V for 2 h. The concentration of NO and NO2 was monitored by a chemiluminescence detector, while the concentration of O2, N2, and N2O was detected on a mass spectrometer. A significant effect of impregnation with KNO3 or K2O on the NOx conversion was observed. In 1,000 ppm NO, both impregnations caused an increased conversion of NO into N2 in the temperature range of 300–400 °C with a current efficiency up to 73%. In 1,000 ppm NO+10% O2, no formation of N2 was observed during polarization, but the impregnations altered the conversion between NO and NO2 on the electrodes. Both impregnations caused increased degradation of the cell stack, but the exact cause of the degradation has not been identified yet.

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Optimization of an electrochemical cell with an adsorption layer for NOx removal

The structure of a multilayer electrochemical cell with an adsorption layer was optimized by removing an yttria-stabilized zirconia cover layer. It was found that the NOx removal properties of the electrochemical cell were dramatically enhanced through the optimization, especially under conditions of low voltage, intermediate temperature, and high O2 concentration. The pronounced increase in activity and selectivity for NOx decomposition after removing the yttria-stabilized zirconia cover layer was attributed to the extensive release of selective reaction sites for NOx species and a strong promotion for NOx reduction from the interaction of the directly connected adsorption layer with both the Pt and catalytic layers. The optimized electrochemical cell may provide a promising solution for NOx emission control.

Pore former induced porosity in LSM/CGO cathodes for electrochemical cells for flue gas purification

In this study the effect of the characteristics of polymethyl methacrylate (PMMA) pore formers on the porosity, pore size distribution and the air flow through the prepared lanthanum strontium manganate/gadolinium-doped cerium oxide (LSM/CGO) cathodes was investigated. Porous cathodes were obtained and the highest porosity measured was 46.4% with an average pore diameter of 0.98 μm. The air flow through this cathode was measured to 5.8 ml/(min mm2). Also the effect of exposure time to the solvent was tested for the most promising PMMA pore former and it was found that the average pore diameter decreases as a result of elongated exposure. Also prolong milling of the LSM powder was found to decrease the porosity of the final cathode and milling time should be highly controlled in order to obtain as porous cathodes as possible.
Improvement of LSM15-CGO10 electrodes for electrochemical removal of NOx by KNO3 and MnOx impregnation

LSM15-CGO10 (La0.85Sr0.15MnO3-Ce0.90Gd0.1O1.95) electrodes were impregnated with either KNO3 or MnOx, and the effect of the impregnations on the activity in NO containing atmospheres was investigated by electrochemical impedance spectroscopy and cyclic voltammetry. The electrodes were tested in 1000 ppm NO, 10% O2 and 1000 ppm NO + 10% O2 in the temperature range 300-500 °C and the electrodes were investigated by scanning electron microscopy before and after testing. At 400-450 °C a NOx-storage process was observed on the KNO3-impregnated electrodes, this process appeared to be dependent on preceding catalytically formation of NO2. Despite a marked difference in the microstructure of the impregnated KNO3 and MnOx, both impregnations caused a significant reduction in the polarization resistance of the electrodes, due to a general decrease in resistance of all the identified electrode processes. The effect of the impregnation was strongest at low temperatures, likely because the microstructure of the impregnated compounds changed at higher temperatures. Scanning electron microscopy images revealed a significant change in the microstructure of the impregnated samples after the test.
Low temperature reduction of NO and O2 on A-site deficient (Pr0.6Sr0.4)1−sFe0.8Co0.2O3−δ perovskites

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Optimizing the Performance of Porous Electrochemical Cells for Flue Gas Purification using the DOE method

The DOE model was used to improve the performance of cells for electrochemical gas purification. Three factors were chosen: the amount of graphite, the Lanthanum Strontium Manganate/Gadolinium-doped Cerium oxide weight % ratio, and the Lanthanum Strontium Manganate pre-calcination temperature (with or without Lanthanum Strontium Manganate calcinated at 1000 °C). The effects of the following physical properties were measured: porosity, pore size, shrinkage, and conductivity. The sintered tapes were also characterized with scanning electron microscopy. Graphite was added as a pore former. The work shows, that a change in a factor not only changes the performance property that one would expect, but also influence other properties.

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Ceria and strontium titanate based electrodes
A ceramic anode structure obtainable by a process comprising the steps of: (a) providing a slurry by dispersing a powder of an electronically conductive phase and by adding a binder to the dispersion, in which said powder is selected from the group consisting of niobium-doped strontium titanate, vanadium-doped strontium titanate, tantalum-doped strontium titanate, and mixtures thereof, (b) sintering the slurry of step (a), (c) providing a precursor solution of ceria, said solution containing a solvent and a surfactant, (d) impregnating the resulting sintered structure of step (b) with the precursor solution of step (c), (e) subjecting the resulting structure of step (d) to calcination, and (f) conducting steps (d)-(e) at least once.

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Characterization of (La$_{1-x}$Sr$_x$)$_3$MnO$_3$ and Doped Ceria Composite Electrodes in NO$_x$-Containing Atmosphere with Impedance Spectroscopy

This study used electrochemical impedance spectroscopy (EIS) to characterize composite metal oxide electrodes in atmospheres containing NO, NO$_2$, and O$_2$. Symmetrical cells with electrodes of (La$_{1-x}$Sr$_x$)$_3$MnO$_3$ [(x, s)=(0.15, 0.9) and (0.5, 0.99)] and doped ceria [Ce$0.9$Gd$0.1$O$_2$ and Ce$_{1-x}$GdxO$_2$ (x=0.1, 0.2)] were subjected to EIS while varying the temperature (from 300 to 600°C), the composition of the atmosphere, and the gas flow. The impedance spectra were fitted to equivalent circuits, and common arcs were identified and sought related to physical and chemical processes. The electrodes had a much lower polarization resistance ($R_p$) when NO or NO$_2$ was present in the atmosphere at low temperatures (300–400°C) than in air. The impedance spectra for electrodes in 1% NO in Ar were dominated by a low frequency arc at high temperatures (500–600°C). This arc seemed to be a type of conversion arc, which is related to a gaseous intermediate (possibly NO$_2$), formed from NO, through which the electrode reaction occurs. Indications were found that the electrodes are not electrochemically active toward NO around open-circuit voltages. ©2010 The Electrochemical Society
Effect of impregnation of La$_{0.85}$Sr$_{0.15}$MnO$_3$/Yttria Stabilized Zirconia Solid Oxide Fuel Cell cathodes with La$_{0.85}$Sr$_{0.15}$MnO$_3$ or Al$_2$O$_3$ nano-particles

Strontium substituted lanthanum manganite and yttria stabilized zirconia solid oxide fuel cell composite electrodes were impregnated with nano-particles of strontium substituted lanthanum manganite or alumina. A clear positive effect was observed on low performing electrodes and on good performing electrodes if the temperature was kept low after the impregnation with strontium substituted lanthanum manganite. On good performing electrodes the effect disappeared on heating. Alumina nano-particles had a detrimental effect on the activity of the strontium substituted lanthanum manganite based electrodes.

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EIS measurements on La$_{1-x}$Sr$_x$Co$_{1-y}$Fe$_y$O$_{3.5}$ based composite electrodes in NO$_x$ containing atmosphere

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Electrochemical reduction of nitrous oxide on La$_{1-x}$Sr$_x$FeO$_3$ perovskites
The electrochemical reduction of nitrous oxide and oxygen has been studied on cone-shaped electrodes of La$_{1-x}$Sr$_x$FeO$_3$-delta perovskites in an all solid state cell, using cyclic voltammetry. It was shown that the activity of the La$_{1-x}$Sr$_x$FeO$_3$-delta perovskites for the electrochemical reduction of nitrous oxide mainly depends on the amount of Fe(III) and oxide ion vacancies. The activity of the La$_{1-x}$Sr$_x$FeO$_3$-delta perovskites towards the electrochemical reduction of nitrous oxide is much lower than the activity of the La$_{1-x}$Sr$_x$FeO$_3$-delta perovskites towards the electrochemical reduction of oxygen, making the possibility of electrochemically reducing nitrous oxide selectively in an exhaust gas containing excess oxygen on this type of materials very doubtful.

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Electrochemical Reduction of NOx Gases on Spinel-Type Electrode Materials

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Electrochemical Reduction of Oxygen and Nitric Oxide at Low Temperature on La\textsubscript{1-x}Sr\textsubscript{x}CoO\textsubscript{3-delta} Cathodes
Six La\textsubscript{1-x}Sr\textsubscript{x}CoO\textsubscript{3} (x= 0, 0.05, 0.15, 0.25, 0.35, 0.50) perovskites were synthesised and characterised by powder XRD and cyclic voltammetry on cone-shaped electrodes in either air or nitric oxide in argon at 200, 300 and 400°C. At 200°C the current densities in air was highest for the strontium free cobaltite, whereas the current densities was highest for La\textsubscript{0.95}Sr\textsubscript{0.05}CoO\textsubscript{3-} in the nitric oxide containing atmosphere. This was also the compound with the highest INO/IO\textsubscript{2} current ratio at 200°C. At higher temperatures a limiting cathodic current was observed for all of the cobaltite’s, except La\textsubscript{0.50}Sr\textsubscript{0.50}CoO\textsubscript{3-}, in both air and the nitric oxide containing atmosphere. This was attributed to a rate limiting chemical step (i.e. dissociation of oxygen or nitric oxide) in the reaction sequence.

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Electrochemical removal of NOx with porous cell stacks

In this study porous cell stacks were investigated for their ability to remove NOx electrochemically. The cell stacks were made from laminated tapes of porous electrolyte Ce0.9Gd0.1O1.95 and composite electrodes of La1–xSrxFexMnO3 (x = 0.15, and 0.5) and ceria doped with Gd or Pr. The cell stacks were infiltrated with nano-particles of pure ceria, Ce0.9Gd0.1O1.95 and Ce0.8Pr0.2O2–δ after sintering. A gas stream containing NO were sent through the cell stack. When the cell stacks were polarised with 0.75 V per cell then it was possible to remove some of the NOx in the temperature interval of 250–400 °C. The cell stacks infiltrated with ceria showed the highest activity, while the ones infiltrated with Ce0.9Gd0.1O1.95 had the highest selectivity towards NO compared to O2. When the cell stack was polarised with 1.5 V for each cell it was possible to remove up to 35% of NO present.

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Electrochemical Removal of NOx-Gasses by Use of LSM and LSF Cathodes Impregnated with NOx-Storage Compounds

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Electrochemical Removal of NOx-Gasses by Use of LSM-Cathodes Impregnated with a NOx Storage Compound

Electrochemical decomposition of NO on La0.85Sr0.15MnO3-- Ce0.90Gd0.10O1.95 electrodes with and without KNO3 impregnation is investigated. The KNO3 is added as this compound is expected to work as a NOx-storage compound. Measurements are made in the temperature range 300-400 degree C and in three different atmospheres: 1000 ppm NO, 10% O2 and 1000 ppm NO + 10% O2. Electrochemical impedance spectroscopy and cyclic voltammetry are used for electrochemical characterization of the electrodes, and the conversion of NO and O2 is measured on a mass-spectrometer. The results show a significant increase in the NO-conversion, when KNO3 is added to the La0.85Sr0.15MnO3-- Ce0.90Gd0.10O1.95 electrodes. ©2010 COPYRIGHT ECS - The Electrochemical Society

High Performance Fe-Co Based SOFC Cathodes

With the aim of reducing the temperature of the solid oxide fuel cell (SOFC), a new high-performance perovskite cathode has been developed. An area-specific resistance (ASR) as low as 0.12 Ωcm2 at 600 °C was measured by electrochemical impedance spectroscopy (EIS) on symmetrical cells. The cathode is a composite between (Gd0.6Sr0.4)0.99Fe0.8Co0.203-δ (GSFC) and Ce0.9Gd0.1O1.95 (CGO10). Examination of the microstructure of the cathodes by scanning electron microscopy (SEM) revealed a possibility of further optimisation of the microstructure in order to increase the performance of the cathodes. It also seems that an adjustment of the sintering temperature will make a lowering of the ASR value possible. The cathodes were compatible with ceria-based electrolytes but reacted to some extent with zirconia-based electrolytes depending on the sintering temperature.
NiCr (x) Fe2-x O-4 as cathode materials for electrochemical reduction of NO (x)
Solid solutions of spinel-type oxides with the composition NiCr x Fe2-x O4 (x=0.0, 0.5, 1.0, 1.5, 2.0) were prepared with the glycine–nitrate combustion synthesis. Four-point DC resistivity measurements show an increase in the conductivity as more Cr is introduced into the structure, whereas dilatometer measurements show that the linear thermal expansion decreases with increasing Cr content. The oxides were used as electrode materials in a pseudo-three-electrode setup in the temperature range of 300–600 °C. Cyclic voltammetry and electrochemical impedance spectroscopy were used to characterize the electrochemical behavior in 1% NO, 1% NO2, and 10% O2. NiCr2O4 shows high activity in NO and NO2 relative to O2 and can therefore be considered as a possible electrode material. Peaks were detected in the voltammograms recorded on NiCr2O4 in 1% NO. The origin of the peaks seems to be related to the oxidation of Cr or the formation of nitrogen-containing species formed on the surface of the electrode.
Sintering effect on material properties of electrochemical reactors used for removal of nitrogen oxides and soot particles emitted from diesel engines

In the present work, 12-layered electrochemical reactors (comprising five cells) with a novel configuration including supporting layer lanthanum strontium manganate (LSM)-yttria stabilised zirconia (YSZ), electrode layer LSM-gadolinia-doped cerium oxide (CGO) and electrolyte layer CGO were fabricated via the processes of slurry preparation, tape casting and lamination and sintering. The parameters of porosity, pore size, pore size distribution, shrinkage, flow rate of the sintered reactors and the electrical conductivities of the supporting layer and the electrode in the sintered reactors were characterised. The effect of sintering temperature on microstructures and properties of the sintered samples was discussed, and 1,250 °C was determined as the appropriate sintering temperature for reactor production based on the performance requirements for applications. Using the present ceramic processing route, porous, flat and crack-free electrochemical reactors were successfully achieved. The produced electrochemical reactors have the potential application in the removal of NOx and soot particles emitted from the diesel engines.
Solid State Electrochemical DeNOx: An overview
The literature on direct electrochemical reduction of NOx in a solid state cell has been reviewed. It is shown that the reduction of nitric oxide either occurs on the electrode or on the electrolyte if F-centers are formed. It is also shown that some oxide based electrodes has a high apparent selectivity towards the reduction of nitric oxide in a net oxidizing atmosphere. The activity and apparent selectivity of oxide based electrodes is strongly dependent on the composition of the oxide. The selectivity of noble metal based electrodes is generally very low. However, coating of noble metal based electrodes can give current efficiencies of up to 20 % even in the presence of large amounts of oxygen.

The Effect of a CGO Barrier Layer on the Performance of LSM/YSZ SOFC Cathodes
LSM/YSZ (Strontium substituted Lanthanum Manganite/Yttria Stabilised Zirconia) SOFC (Solid Oxide Fuel Cell) composite electrodes were fabricated with slurry spraying on both sides on either pure YSZ electrolyte foils or YSZ electrolyte foils with a CGO (Cerium-Gadolinium Oxide) barrier layer made by spin coating. Electrochemical impedance spectroscopy (EIS) was used to evaluate the performance of the LSM/YSZ composite electrodes. It was shown that the CGO barrier layer affects both the performance of the LSM/YSZ composite electrodes and the series resistance of the cells. This indicates that the cathode-electrolyte interface and the barrier layer-electrolyte interface have a large influence on the performance of LSM/YSZ composite electrodes.
The effect of A-site deficiency on the performance of La$_{1-x}$Fe$_{0.4}$Ni$_{0.6}$O$_{3-\delta}$ cathodes

The effect of lowering the A-site stoichiometry of La-Fe-Ni based perovskite solid oxide fuel cell cathodes was investigated with electrochemical impedance spectroscopy on cone-shaped electrodes using a Ce$_0.9$Gd$_{0.1}$O$_{1.95}$ electrolyte. It was shown that a lowering of the A-site stoichiometry lowers the amount of Ni in the perovskite phase, as powder XRD revealed that NiO was expelled from the perovskite lattice when the A-site stoichiometry was decreased. NiO inhibits the reduction of oxygen as the activity of a nominally A-site deficient La$_{1-x}$Fe$_{0.4}$Ni$_{0.6}$O$_{3-\delta}$ perovskite was worse than the activity of the corresponding LaFe$_{0.4+x}$Ni$_{0.6-x}$O$_3$ perovskite without NiO. NiO is therefore poison for the reduction of oxygen at the cathode in a solid oxide fuel cell.

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An EIS study of La\(_{2-x}\)Sr\(_x\)NiO\(_{4+\delta}\) SOFC cathodes
La\(_{2-x}\)Sr\(_x\)NiO\(_{4+\delta}\) materials were investigated as cathodes for the electrochemical reduction of oxygen on a Ce\(_{1.9}\)Gd\(_{0.1}\)O\(_{1.95}\) (CGO10) electrolyte using cone-shaped electrodes together with electrochemical impedance spectroscopy. The area-specific resistance (ASR) of the La\(_{2-x}\)Sr\(_x\)NiO\(_{4+\delta}\) nickelates towards the reduction of oxygen is equal to the ASR of perovskites; however, it is not as low as for the best Fe–Co-based perovskites. The lowest ASR is found for the compound La\(_{1.75}\)Sr\(_{0.25}\)NiO\(_{4+\delta}\) with an ASR of 23.8 \(\Omega\)cm\(^2\) measured on a cone-shaped electrode in air at 800 °C. It is suggested that difference in oxide ionic conductivity of the nickelates is the main cause for the different activity of the nickelates towards the electrochemical reduction of oxygen.

A-Site Deficient (Pr\(_{0.6}\)Sr\(_{0.4}\))(1-s)Fe\(_{0.8}\)Co\(_{0.2}\)O\(_{3-\delta}\) Perovskites as Solid Oxide Fuel Cell Cathodes
Five A-site deficient (Pr\(_{0.6}\)Sr\(_{0.4}\))[1-s]Fe\(_{0.8}\)Co\(_{0.2}\)O\(_{3-\delta}\) perovskites (s=0.01, 0.05, 0.10, 0.15, and 0.20) were synthesized using the glycine-nitrate process. The perovskites were characterized with powder X-ray diffraction (XRD), dilatometry, four-point dc conductivity measurements, and electrochemical impedance spectroscopy using cone-shaped electrodes on a Ce\(_{0.9}\)Gd\(_{0.1}\)O\(_{1.95}\) electrolyte. XRD revealed that only the compounds with s=0.01 and 0.05 were of single phase. The other compounds contained an additional phase in the form of a Co–Fe spinel. The thermal expansion coefficient decreased systematically with an increase in s. The total conductivity followed the small polaron hopping process below a certain characteristic temperature. The total conductivity decreased with increasing s. It was likewise shown that the polarization resistance found a minimum for the compound with s=0.05, with a total area specific resistance more than 3 times lower than the weakly A-site deficient (Pr\(_{0.6}\)Sr\(_{0.4}\))[0.99]Fe\(_{0.8}\)Co\(_{0.2}\)O\(_{3-\delta}\) perovskite. ©2009 The Electrochemical Society
Characterization of MgMnxFe2-xO4 as a possible cathode material for electrochemical reduction of NOx

Spinel-type oxides of MgMn x Fe2−x O4, x = 0.0, 0.2, 0.4, 0.6, 0.8, 1.0, were synthesized as a solid state reaction and characterized with dilatometry and resistivity measurements up to 1000 °C. Results showed a general decrease of the linear expansion and an increase in conductivity as a function of the Mn content. Point electrodes were analyzed from 300 to 600 °C in a pseudo-three-electrode setup in 1% NO, 1% NO2, and 10% O2 using cyclic voltammetry. The activities in O2 were in general very low whereas the activities in NO were slightly higher. The activities in NO2 were for all materials much higher than the activities in O2. Even though Mn tends to decrease the activity of the materials, current ratios of have relatively high values in both NO and NO2.
Electrochemical characterization and redox behavior of Nb-doped SrTiO₃

Sr-vacancy compensated Nb-doped SrTiO₃ with the nominal composition Sr₀.₉₄Ti₀.₉Nb₀.₁O₃ has been evaluated as a solid oxide fuel cell (SOFC) anode material in terms of redox stability and electrochemical properties. Sr₀.₉₄Ti₀.₉Nb₀.₁O₃ has been synthesized with a recently developed modified glycine-nitrate process. The phase purity and redox behavior have been analyzed with XRD and TGA. The electrochemical properties of Sr₀.₉₄Ti₀.₉Nb₀.₁O₃ and a composite electrode of Sr₀.₉₄Ti₀.₉Nb₀.₁O₃/YSZ have been investigated by electrochemical impedance spectroscopy (EIS) on cone shaped electrodes and on electrodes in a symmetrical cell configuration. The experiments indicated that the Nb-doped SrTiO₃ electrodes were redox stable and showed a potential ability to be used as a part of a SOFC anode. The electrochemical activity appeared to be governed by the concentration of defect species (especially Ti³⁺ and V-O(++)+) in the vicinity of the triple phase boundaries. However, the electrocatalytic activity of the materials was not sufficient and it needs to be improved if Nb-doped SrTiO₃ based materials are to be realized as SOFC anodes. (C) 2008 Elsevier B.V. All rights reserved.

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Web of Science (2009): Indexed yes
Original language: English
DOIs: 10.1016/j.ssi.2008.10.011
Source: orbit
Source-ID: 242666
Research output: Contribution to journal › Journal article – Annual report year: 2009 › Research › peer-review

Electrochemical reduction of NO on La₂₋ₓSrₓNiO₄ based electrodes

The series La₂₋ₓSrₓNiO₄ (x=0.0, 0.05, 0.15, 0.25, 0.35, and 1.0) was tested for functionality as electrode materials for direct electrochemical reduction of NO. The materials were tested using cyclic voltammetry in 1% NO and 10% O₂ in Ar on a cone-shaped electrode. The best materials for the electrochemical reduction of NO are La₂NiO₄ and LaSrNiO₄, which have current densities for NO reduction 1.82 and 7.09 times higher, respectively, than for O₂ at 400 °C. Increasing the temperature decreased the ability to reduce NO before O₂ while the activity increased. The adsorbed species during direct decomposition was attempted, clarified using X-ray absorption near-edge structure experiments and thermogravimetry, but no conclusive results were obtained.

General information
Publication status: Published
Organisations: Electroceramics, Fuel Cells and Solid State Chemistry Division, Rise National Laboratory for Sustainable Energy, Electrochemistry
Contributors: Simonsen, V. L. E., Nørskov, L., Hagen, A., Kammer Hansen, K.
Pages: 1529-1534
Publication date: 2009
Peer-reviewed: Yes

Publication information
Journal: Journal of Solid State Electrochemistry
Volume: 13
Issue number: 10
Processing and characterization of porous electrochemical cells for flue gas purification

In the present work, porous electrode materials lanthanum strontium manganate (LSM)-gadolinium-doped cerium oxide (CGO) and electrochemical cells LSM-CGO+CGO were fabricated via the processes of slurry preparation, tape casting and lamination, and sintering. Graphite, wheat starch, and polyamide powders were used as pore formers, respectively, in order to investigate their effects on shrinkage, flow rate, porosity, and average pore size of the sintered samples. The observation from scanning electronic microscope (SEM) revealed different microstructures caused by different pore formers. Porous, flat, and crack-free electrochemical cells were successfully achieved using the present ceramic processing route. The produced cells could potentially be used for flue gas purification.

General information
Publication status: Published
Organisations: Ceramic processing, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electrochemistry
Contributors: He, Z., Andersen, K. B., Keel, L., Nygaard, F. B., Menon, M., Kammer Hansen, K.
Pages: 427-431
Publication date: 2009
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Publication information
Journal: Ionics
Volume: 15
Issue number: 4
ISSN (Print): 0947-7047
Ratings:
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 0.55 SNIP 0.619
Web of Science (2009): Indexed yes
Keywords: Fuel Cells and hydrogen, Flue gas purification
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http://www.springerlink.com/content/3073434t10823532/
Source: orbit
Source-ID: 233534
Research output: Contribution to journal › Journal article – Annual report year: 2009 › Research › peer-review

The NiFe2O4-MgFe2O4 series as electrode materials for electrochemical reduction of NOx

Solid solutions of spinel-type oxides with the composition \((x=0.0, 0.3, 0.5, 0.6, 1.0)\) were prepared with the glycine-nitrate combustion synthesis \((x=0.0, 0.3, 0.5, 0.6)\) and the citric-acid combustion synthesis \((x=1.0)\). The oxides were used as electrode materials in a pseudo-three-electrode setup in the temperature range of 400–600 °C. Cyclic voltammetry and electrochemical impedance spectroscopy were used to characterize the electrochemical behavior in 1% NO and 10% O2. Measurements show that NiFe2O4 has relatively high cathodic activity in both NO and O2, whereas MgFe2O4 shows much higher activity in NO compared to O2. MgFe2O4 was also measured with cyclic voltammetry in 1% NO2 and different gas mixtures of NO and O2 at 300 and 400 °C. Results show that the cathodic activities (−0.6 V) are relatively high with current ratios, ranging from 10.1–167.7 and with a maximum at 400 °C. Dilatometry measurements were performed on the materials in air up to 1,000 °C, and they showed that the Curie temperature could be detected for all samples. Four-point DC resistivity measurements at elevated temperatures show that Ni0.4Mg0.6Fe2O4 has the highest conductivity, whereas Ni0.7Mg0.3Fe2O4 and NiFe2O4 have the highest conductivity at lower temperatures.
Defect and electrical transport properties of Nb-doped SrTiO$_3$

This study reports the defect and electrical transport properties of Nb-doped SrTiO$_3$. Samples with various A/B-ratios were synthesized by a modified glycine-nitrate combustion process and evaluated as a constituent in a SOFC anode. The phase purity and defect structure of the materials have been analyzed with SEM, XRD, TGA, and XANES. The electrical conductivity of Nb-doped strontium titanate (Sr$_{0.94}$Ti$_{0.9}$Nb$_{0.1}$O$_3$ - sintered in 9% H$_2$/N$_2$ at 1400 degrees C for 12 h) decreased with increasing temperature and showed a phonon scattering conduction mechanism with ($\sigma>120$ S/cm at 1000 degrees C (in 9% H$_2$/N$_2$). The results were in agreement with the defect chemistry model of donor-doped SrTiO$_3$ where the charge compensation changes from Sr vacancy compensation to the electronic type when samples are sintered in reducing atmosphere. XANES in combination with TGA indicated that Ti is the only species that is reduced to a lower oxidation state (from Ti$^{4+}$ to Ti$^{3+}$). The pre-edge fine structure (PEFS) from the XANES results indicated that Nb improved the overlap of the Ti atomic orbitals and thereby provided one more explanation for the positive effect of Nb on the electronic conductivity of Nb-doped SrTiO$_3$. (C) 2008 Elsevier B.V. All rights reserved.
Electrochemical cell for removing NOx and soot from diesel exhaust

General information
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Organisations: Ceramic processing, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electrochemistry
Contributors: Andersen, K. B., He, Z., Werchmeister, R. M. L., Keel, L., Nygaard, F. B., Menon, M., Kammer Hansen, K.
Pages: 169-169
Publication date: 2008

Host publication information
Title of host publication: Book of abstracts
Publisher: American Ceramic Society
Source: orbit
Source-ID: 232442
Research output: Chapter in Book/Report/Conference proceeding – Conference abstract in proceedings – Annual report year: 2008 › Research

Electrochemical reduction of NO and O2 on La2-xSrxCuO4-based electrodes

General information
Publication status: Published
Organisations: Electroceramics, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electrochemistry
Contributors: Simonsen, V. L. E., Nørskov, L., Kammer Hansen, K.
Pages: 1573-1577
Publication date: 2008
Peer-reviewed: Yes

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Volume: 12
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Ratings:
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Scopus rating (2008): SJR 0.75 SNIP 0.738
Web of Science (2008): Indexed yes
Original language: English
DOIs: 10.1007/s10008-008-0514-6
Source: orbit
Source-ID: 227871
Research output: Contribution to journal › Journal article – Annual report year: 2008 › Research › peer-review

Electrochemical reduction of O2 and NO on Ni, Pt and Au

General information
Publication status: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Contributors: Kammer Hansen, K.
Pages: 591-595
Publication date: 2008
Peer-reviewed: Yes

Publication information
Journal: Journal of Applied Electrochemistry
Volume: 38
Evaluation of LSF based SOFC Cathodes using Cone-shaped Electrodes

Seven La$_{1-x}$Sr$_x$FeO$_{3-\delta}$ ($x = 0, 0.05, 0.15, 0.25, 0.35, 0.50, 0.70$) based perovskites were synthesized using the glycine-nitrate method. The La$_{1-x}$Sr$_x$FeO$_{3-\delta}$ compounds were characterized with powder X-ray diffraction and electrochemical impedance spectroscopy on cone-shaped electrodes using a Ce$_{1.9}$Gd$_{0.1}$O$_{1.95}$ electrolyte. The activity of the La$_{1-x}$Sr$_x$FeO$_{3-\delta}$ based perovskites towards the electrochemical reduction of oxygen was strongly dependent of the strontium content, the activity being highest for the composition La$_{0.85}$Sr$_{0.15}$FeO$_{3-\delta}$. The results indicates that Fe(III) is the catalytic active specie towards the electrochemical reduction of oxygen in a solid oxide fuel cell on La$_{1-x}$Sr$_x$FeO$_{3-\delta}$ compounds. The results also show that oxide ion vacancies in the perovskite structure are important for the electrochemical reduction of oxygen. However, the effect of ordering of oxide ion vacancies for the strontium rich compounds can not be excluded.
Niobium-doped strontium titanates as SOFC anodes
Sr-vacancy compensated Nb-doped SrTiO(3) with the nominal composition Sr(0.94)Ti(0.9)Nb(0.1)O(3) has been evaluated as part of a solid oxide fuel cell (SOFC) anode material in terms of redox stability, electrical conductivity, as well as electrochemical properties. Sr(0.94)Ti(0.9)Nb(0.1)O(3) has been synthesized with a recently developed modified glycine-nitrate process. The synthesized powders have been calcined and sintered in air or in 9% H(2) / N(2) between 800 - 1400 degrees C. After calcination the samples were single phase Nb-doped strontium titanate with grain sizes of less than 100 nm in diameter on average. The phase purity, defect structure, and microstructure of the materials have been analyzed with SEM, XRD, and TGA. The electrical conductivity of the Nb-doped titanate decreased with increasing temperature and showed a phonon scattering conduction mechanism with sigma > 120 S/cm at 1000 degrees C (in 9% H(2) / N(2)). The electrochemical properties of Sr(0.94)Ti(0.9)Nb(0.1)O(3) and a composite electrode of Sr(0.94)Ti(0.9)Nb(0.1)O(3)/YSZ have been investigated by using a symmetrical cell configuration. The tests indicated that the electrodes were redox stable and showed a potential ability of the Nb-doped titanates to be used as a part of a SOFC anode. However, the catalytic activity of the materials was not sufficient and it needs to be improved if titanate based materials are to be realized as constituents in SOFC anodes.

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Organisations: Ceramic processing, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electrochemistry
Contributors: Blennow Tullmar, P., Kammer Hansen, K., Wallenberg, L. R., Mogensen, M. B.
Pages: 203-214
Publication date: 2008
Peer-reviewed: No

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Web of Science (2008): Indexed yes
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Research output: Contribution to journal » Conference article – Annual report year: 2008 » Research

Strontium Titanate-based Composite Anodes for Solid Oxide Fuel Cells
Surfactant-assisted infiltration of Gd-doped ceria (CGO) in Nb-doped SrTiO3 (STN) was investigated as a potential fuel electrode for solid oxide fuel cells (SOFC). An electronically conductive backbone structure of STN was first fabricated at high temperatures and then combined with the mixed conducting and electrochemically active nano-sized CGO phase at low temperatures. Symmetrical cell measurements at open circuit voltage (OCV), showed that the electrochemical activity was maintained or even improved compared to Ni/YSZ fuel electrodes. The novel electrode had an electrode polarization resistance of 0.12 Omega cm2 and 0.44 Omega cm2 in humidified H2 at 850 ºC and 650 ºC, respectively. In addition, the ceramic composite electrode was shown to be redox stable. The electrode was actually activated with redox cycles at 650 ºC. The ceramic electrode structure thus presents a potential solution to overcome some of the major limitations of the current Ni-YSZ cermet SOFC anodes.

General information
Publication status: Published
Organisations: Ceramic processing, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electrochemistry
Contributors: Blennow Tullmar, P., Kammer Hansen, K., Wallenberg, L., Mogensen, M. B.
Pages: 181-194
Publication date: 2008
Peer-reviewed: Yes

Publication information
Temperature dependence of the cation distribution in ZnFe2O4 measured with high temperature neutron diffraction

General information
Publication status: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Contributors: Braestrup, F. R., Hauback, B., Kammer Hansen, K.
Pages: 2364-2369
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Publication information
Journal: Journal of Solid State Chemistry
Volume: 181
Issue number: 9
ISSN (Print): 0022-4596
Ratings:
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 0.982 SNIP 1.171
Web of Science (2008): Indexed yes
Original language: English
DOIs:
10.1016/j.jssc.2008.05.028
Source: orbit
Source-ID: 227943
Research output: Contribution to journal › Journal article – Annual report year: 2008 › Research › peer-review

The action of nano-particles in SOFC electrodes

General information
Publication status: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electroceramics, Ceramic processing
Contributors: Mogensen, M. B., Søgaard, M., Blennow Tullmar, P., Kammer Hansen, K.
Publication date: 2008

Host publication information
Title of host publication: Proceedings (on CD-ROM)
Publisher: European solid oxide fuel cell forum
Source: orbit
A-site deficient (La0.6Sr0.4)1-xFe0.8Co0.2O3-delta perovskites as SOFC cathodes

General information
Publication status: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Contributors: Kammer Hansen, K., Hansen, K. V.
Pages: 1379-1384
Publication date: 2007
Peer-reviewed: Yes

Publication information
Journal: Solid State Ionics
Volume: 178
ISSN (Print): 0167-2738
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Scopus rating (2007): SJR 1.295 SNIP 1.392
Web of Science (2007): Indexed yes
Original language: English
DOIs:
10.1016/j.ssi.2007.07.012
Source: orbit
Source-ID: 215812

Conductivity and electrochemical characterization of PrFe1-xNixO3-δ at high temperature
PrFe(1-x)NixO3-δ (x=0.4-0.6) compounds were synthesized and characterized by powder XRD, electrical conductivity and electrochemical impedance spectroscopy on point electrodes on a Ce0.9Gd0.1O2-delta (CGO10) electrolyte. As a reference, the electrochemical performance of LaFe(0.4)AM(0.6)O3-delta was also measured. The main phase in the PrFe1-xNixO3-delta series was perovskite-type structure and belonged to the orthorhombic crystal system. The conductivities are fairly high, e.g. around 220 S cm(-1) at 873 K for the x = 0.4 compound. The electrochemical performance of the PrFe(1-x)NixO3(3-delta) series is similar to that of La0.6Sr0.4Fe0.8Co0.2O3-delta, which is a good candidate as a cathode material for SOFCs operating at intermediate temperature. The electrochemical performance of LaFe0.4Ni0.6O3-delta was even higher than that of the PrFe1-xNixO3-delta, series and the polarization resistance was 0.14 Omega cm(2) at 1073 K measured on a point electrode. (c) 2006 Elsevier B.V. All rights reserved.

General information
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy, Electrochemistry, Fuel Cells and Solid State Chemistry Division, Electroceramics
Contributors: Hashimoto, S., Kammer Hansen, K., Poulsen, F. W., Mogensen, M. B.
Pages: 256-261
Publication date: 2007
Peer-reviewed: Yes

Publication information
Journal: Journal of Alloys and Compounds
Volume: 428
Issue number: 1-2
ISSN (Print): 0925-8388
Ratings:
Scopus rating (2007): SJR 0.87 SNIP 1.209
Web of Science (2007): Indexed yes
Original language: English
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Source: orbit
Source-ID: 216614

Research output: Contribution to journal › Journal article – Annual report year: 2007 › Research › peer-review
Electrochemical reduction of NO\textsubscript{2} studied by the use of cone-shaped electrodes

General information
Publication status: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Contributors: Kammer Hansen, K.
Pages: 2721-2724
Publication date: 2007
Peer-reviewed: Yes

Gd\textsubscript{0.6}Sr\textsubscript{0.4}Fe\textsubscript{0.8}Co\textsubscript{0.2}O\textsubscript{3-δ}: A novel type of SOFC cathode

The fabrication and electrochemical activity of a type of solid oxide fuel cell (SOFC) cathode is described in this paper. In search of new cathodes a Gd\textsubscript{0.6}Sr\textsubscript{0.4}Fe\textsubscript{0.8}Co\textsubscript{0.2}O\textsubscript{3-δ} compound was synthesized using the glycine-nitrate method. It turned out that this was a two-phase compound consisting of two perovskite phases, a cubic and an orthorhombic phase, as shown by Rietveld refinements. These two phases were synthesized and a cone-shaped electrode study was undertaken. It was shown that the composite cathode had an electrochemical activity superior to that of the two single-phase perovskites, indicating that the unique microstructure of this type of cathode is essential for achieving high electrochemical activity toward the reduction of oxygen in a SOFC.

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Publication status: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electroceramics
Contributors: Kammer Hansen, K., Søgaard, M., Mogensen, M. B.
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High performance ceramic composite anodes for solid oxide fuel cells

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Publication status: Published
Organisations: Ceramic processing, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electrochemistry
Contributors: Blennow Tullmar, P., Hansen, K., Wallenberg, L., Mogensen, M. B.
Publication date: 2007
Peer-reviewed: No
Event: Abstract from Conference on Electrochemical Science and Technology, Århus, Denmark.
URLs:
Source: orbit
Source-ID: 215894
Research output: Contribution to conference › Conference abstract for conference – Annual report year: 2007 › Research

Influence of BaO in perovskite electrodes for the electrochemical reduction of NOx
Using the point electrode method, the effect of BaO on electrochemical reduction of NO (x) was investigated using the perovskites La0.85Sr0.15MnO3 (LSM15) and La0.85Sr0.15CoO3 (LSCO15) as electrode materials. The experiments were carried out in the temperature range 400-600 degrees C in 1% NO and 10% O-2, respectively. For the LSM15 electrode the ability to reduce NO compared to O-2 was increased when applying 20 wt% BaO in the electrode while the current density of the electrode was decreased approximately a decade. For the pure LSM15 electrode the highest current density ratio of NO and O-2 was obtained at 400 degrees C while the optimal temperature in term of current density ratios was 500 degrees C for the LSM15 + BaO and LSM15 + BaO + Pt electrodes. The activity of the electrode decreases when applying BaO or BaO + Pt to the electrode except for the LSM15 + BaO + Pt electrode at 500 degrees C where the activity is approximately the same as for the LSM15 electrode. The formation of Ba(NO3)(2) was clearly seen when applying 16.7 wt% BaO and 2 wt% Pt to the electrode. Generally it was observed that at 400 degrees C the activity of the electrodes were low, while at 600 degrees C the kinetics favored oxygen reduction compared to reduction of nitric oxide. The LSCO15 electrode containing BaO reacted to form a K2NiF4-structure and was not tested further.

New methods for removal of soot and NOx

General information
Publication status: Published
Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy

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Original language: English
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Source: orbit
Source-ID: 216030
Research output: Contribution to journal › Journal article – Annual report year: 2007 › Research › peer-review
Niobium-doped strontium titanates as SOFC anodes

General information
Publication status: Published
Organisations: Ceramic processing, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electrochemistry
Contributors: Blennow Tullmar, P., Hansen, K., Wallenberg, L., Mogensen, M. B.
Publication date: 2007
Peer-reviewed: No
Event: Abstract from 32. International Cocoa Beach conference and exposition on advanced ceramics and composites, Cocoa Beach, FL (US).
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Research output: Contribution to conference › Conference abstract for conference – Annual report year: 2007 › Research

Processing of Ce1-xGdxO2-delta (GDC) thin films from precursors for application in solid oxide fuel cells

General information
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Rose, L., Menon, M., Kammer Hansen, K., Kesler, O., Larsen, P.
Pages: 293-298
Publication date: 2007
Peer-reviewed: Yes
Publication information
Volume: 15-17
Original language: English
Source: orbit
Source-ID: 309677
Research output: Contribution to journal › Journal article – Annual report year: 2006 › Research › peer-review

Spinels as cathodes for the electrochemical reduction of O2 and NO
Spinels were synthesised and investigated as electro-catalyst for the electrochemical reduction of oxygen and nitric oxide using cyclic voltammetry and cone shaped electrodes. The following four spinels were investigated: CoFe2O4, NiFe2O4, CuFe2O4 and Co3O4. The composition CuFe2O4 revealed the largest difference in activity between reduction of oxygen and the reduction of nitric oxide, the activity being highest for the reduction of nitric oxide. The material is probably not stable when polarised cathodically. However it seems that the electrode material can be regenerated upon oxidation. NiFe2O4 is also more active for the reduction of nitric oxide than for the reduction of oxygen, whereas the cobalt containing spinels have a higher activity for the reduction of oxygen than for the reduction of nitric oxide.

General information
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy, Electrochemistry, Fuel Cells and Solid State Chemistry Division
Contributors: Simonsen, V. L. E., Find, D., Lilliedal, M., Petersen, R., Kammer Hansen, K.
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Publication information
Journal: Topics in Catalysis
Spinell-type electrode materials for purification of exhaust gasses from diesel fired engines

General information
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Organisations: Electrochemistry, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Contributors: Bræstrup, F. R., Kammer Hansen, K.
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Research output: Contribution to journal › Journal article – Annual report year: 2007 › Research › peer-review

Synthesis of Nb-doped SrTiO3 by a modified glycine-nitrate process
The objective of the present investigation was to develop a technique to synthesize submicronic particles of Nb-doped strontium titanate with a homogeneous composition. This was achieved by a modified glycine-nitrate process, using Ti-lactate, Nb-oxalate, and Sr(NO3)2 as starting materials. A combination of both citric acid and glycine was needed in order to integrate the useful features of both complexation and combustion natures of citric acid and glycine, respectively. The amount of citric acid, glycine, and nitrates in the starting solution, as well as the source for extra nitrates, and the uniformity of heating during the thermal dehydration step were found to have significant influence on the final phase purity of the material. Calcination at 1100 degrees C in 7% H-2 in N-2 produced single phase Nb-doped strontium titanate with grain sizes of about 100 nm in diameter on average. (c) 2007 Elsevier Ltd. All rights reserved.

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Publication status: Published
Organisations: Ceramic processing, Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, Electrochemistry
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Journal: Journal of the European Ceramic Society
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Web of Science (2007): Indexed yes
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DOIs:
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Research output: Contribution to journal › Journal article – Annual report year: 2007 › Research › peer-review
Electrical and electro-chemical characterisation of La0.99Fe1−xNixO3−δ perovskites

Electrochemical reduction of NOx on spinel-type materials

Novel ceramic anodes for solid oxide fuel cells investigated by cone shaped electrodes
Studies of Fe-Co based perovskite cathodes with different A-site cations
Iron-cobalt based perovskite cathodes with different A-site cations ((Ln(0.6)Sr(0.4))(0.99)Fe0.8Co0.2O3-delta, where Ln is La, Pr, Sm or Gd) have been synthesised, characterised by a powder XRD, dilatometry, 4-point DC conductivity measurements, and electrochemical impedance spectroscopy (EIS) on cone shaped electrodes. In addition to this scanning electron microscopy (SEM) was used to characterise the bars. XRD revealed that only the La-containing perovskite was hexagonal. The Pr and Sm perovskites were orthorhombic. The gadolinium-based perovskite was a two phase system consisting of an orthorhombic and a cubic perovskite phase. The thermal expansion coefficient (TEC) increased systematically with a decrease in the size of the A-site cation until the gadolinium-containing perovskite where the TEC decreases abruptly. The total electric conductivity was the highest for the La-based perovskite and the lowest for the Gd-based perovskite as determined by 4-point DC conductivity measurements on bars. A clear correlation between the size of the A-site cation and the electrochemical performance was revealed, as the area specific resistance (ASR) was the lowest for the compounds with the smallest A-site cation. This might be explained on the background of the creation of a two-phase structure with a unique microstructure when the size of the A-site cation is lowered, or that one of the phases has a high electro-catalytic activity towards the electrochemical reduction of oxygen. (c) 2006 Elsevier B.V. All rights reserved.

Synthesis of Nb-doped SrTiO3 by a modified glycine-nitrate process
A study of Pr$_{0.7}$Sr$_{0.3}$Fe$_{1-x}$Ni$_x$O$_{3-\delta}$ as a cathode material for SOFCs with intermediate operating temperature

Pr$_{0.7}$Sr$_{0.3}$Fe$_{1-x}$Ni$_x$O$_{3-\delta}$ (PSFN; x=0.2-0.5) compounds were synthesized and characterized by powder X-ray diffraction (XRD), thermal expansion coefficient (TEC) measurements, electrical conductivity and electrochemical impedance spectroscopy on cone-shaped electrodes on a Ce$_{0.9}$Gd$_{0.1}$O$_{1.95}$ (CGO10) electrolyte. The main phase of the powders belongs to the orthorhombic crystal system. The conductivities are fairly high, e.g., around 450 scm$^{-1}$ at 600 °C for the x=0.3 compound. The TEC of the compounds is close to the values of ceria-based electrolytes. PSEN showed hysteresis in the temperature dependence of the conductivity, TEC and area-specific polarization resistance $R_{pol}$. It is considered that the hysteresis was caused by relatively slow adjustment of the oxygen stoichiometry. The electrochemical La$_{0.6}$Sr$_{0.4}$Fe$_{0.8}$Co$_{0.2}$O$_{3-\delta}$. © 2004 Elsevier B.V. All rights reserved.

General information
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy, Electrochemistry, Fuel Cells and Solid State Chemistry Division
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Web of Science (2005): Indexed yes
Original language: English
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Research output: Contribution to journal → Journal article – Annual report year: 2005 → Research → peer-review

Ceria revisited: Electrolyte or electrode material?

General information
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Mogensen, M. B., Lybye, D., Kammer Hansen, K., Bonanos, N.
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Publication date: 2005

Host publication information
Title of host publication: Proceedings. Vol. 2. Materials
Place of publication: Pennington, NJ
Publisher: The Electrochemical Society
Source: orbit
Source-ID: 308240
Research output: Chapter in Book/Report/Conference proceeding → Article in proceedings – Annual report year: 2005 → Research

Charge disproportionation in (X$_{0.6}$Sr$_{0.4}$)$_{0.99}$Fe$_{0.8}$Co$_{0.2}$O$_{3-\delta}$ perovskites (X = La, Pr, Sm, Gd)
The change in crystal structure and the oxidation state in iron of iron-cobalt-based perovskites with different A-site cations is investigated by the use of powder XRD and Mossbauer spectroscopy. The perovskites investigated are (X$_{0.6}$Sr$_{0.4}$)$_{0.99}$Fe$_{0.8}$Co$_{0.2}$O$_{3-\delta}$, where X is La, Pr, Sm or Gd. It is shown that the crystal structure changes from hexagonal to orthorhombic when the size of the A-site cation is lowered. At room temperature, the oxidation state of iron increases when the size of the A-site cation is lowered. This is explained by a crystal packing effect. At lower temperatures, charge disproportion of Fe(IV) occurs to Fe(III) and Fe(V). The transition temperature depends on the type of the A-site cation. Some of the Fe is reduced to Fe(III) by transference of electrons from Co upon cooling below room temperature.
Electro-catalytic processes studied by the use of point electrodes

General information
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Kammer Hansen, K., Mogensen, M. B.
Pages: 253-260
Publication date: 2005

Host publication information
Title of host publication: Solid state electrochemistry. Proceedings
Place of publication: Roskilde
Publisher: Risø National Laboratory
ISBN (Print): 87-550-3455-1
Source: orbit
Source-ID: 308384
Research output: Chapter in Book/Report/Conference proceeding – Annual report year: 2005

Electrochemical DeNOx in solid electrolyte cells - an overview

General information
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Kammer Hansen, K.
Pages: 33-39
Publication date: 2005
Peer-reviewed: Yes

Publication information
Journal: Applied Catalysis B: Environmental
Volume: 58
ISSN (Print): 0926-3373
Ratings:
Scopus rating (2005): SJR 2.103 SNIP 2.296
Web of Science (2005): Indexed yes
LSFM perovskites as cathodes for the electrochemical reduction of NO
Six La0.6Sr0.4Fe1-xMnO3-delta (x=0.0, 0.2, 0.4, 0.6, 0.8, 1.0) perovskite compounds have been synthesised by the citric-acid route. The perovskites have been characterised by powder XRD and are shown to belong to the hexagonal crystal system. The perovskites are also evaluated by TG-measurements in 50% oxygen in nitrogen and in nitrogen. From this the redox capacity is calculated and shown to be highest for the iron rich compounds. Cyclic voltammetry has been recorded on cone shaped electrodes of the LSFM compounds in atmospheres containing either NO or O2, at temperatures of 300, 400 and 500 degrees C on a ceria based electrolyte. Only La0.6Sr0.4Fe0.8Mn0.2O3-delta (LSFM020) and La0.6Sr0.4FeO3-delta (LSFM000) show significant activity for the reduction of NO. This can probably be related to the high redox capacity of these compounds. The activity of the perovskites for the reduction of oxygen increases systematically with increasing iron content. The selectivity of the perovskites towards the reduction of NO with regard to the reduction of O2 is highest at the lowest temperatures. (c) 2004 Published by Elsevier B.V.

Oxidation of methane and hydrogen on Ce1-xGdxO2-δ flourrites
The oxidation of methane and hydrogen was studied on cone shaped electrodes with the composition Ce1-xGdxO2-δ (x equals 0, 0.1, 0.2, 0.4). It was shown that the area specific resistance values measured at open-circuit voltage (OCV) for the oxidation of both methane and hydrogen is lowest for the composition Ce0.9Gd0.1O2-δ. The OCV in wet methane was shown to depend on the material composition. It was shown that stable operation in wet methane could be achieved as long as the temperature was kept below 750 degrees C. (C) 2004 The Electrochemical Society.
Perovskites as electrodes and catalysts

General information
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Kammer Hansen, K.
Publication date: 2005
Peer-reviewed: No
Source: orbit
Source-ID: 308055
Research output: Contribution to conference › Conference abstract for conference – Annual report year: 2005 › Research

Solid state electrochemistry. Proceedings

General information
Publication status: Published
Organisations: Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy
Number of pages: 388
Publication date: 2005

Historical publication information
Place of publication: Roskilde
Publisher: Risø National Laboratory
ISBN (Print): 87-550-3455-1
Original language: English
Source: orbit
Source-ID: 308373
Research output: Book/Report › Book – Annual report year: 2005 › Research › peer-review

Structural characterization of A-site deficient strontium titanate by the use of cone shaped electrodes, electron microscopy, and XRD

General information
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Blennow Tullmar, P., Kammer Hansen, K., Mogensen, M. B.
Publication date: 2005

Host publication information
Title of host publication: Book of abstracts
Place of publication: Lausanne (CH)
Publisher: International Society of Electrochemistry
Source: orbit
Source-ID: 308426
Research output: Chapter in Book/Report/Conference proceeding › Conference abstract in proceedings – Annual report year: 2005 › Research

A study of Pr_{0.7}Sr_{0.3}Fe_{1-x}Ni_{x}O_{3-δ} as a cathode material for intermediate temperature operating SOFCs

General information
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy
A study of Pr$_{0.7}$Sr$_{0.3}$Fe$_{1-x}$Ni$_x$O$_{3-\delta}$ as an SOFC cathode material

General information
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Hashimoto, S., Kammer Hansen, K., Larsen, P., Poulsen, F., Mogensen, M. B.
Pages: 42-43
Publication date: 2004

Host publication information
Title of host publication: Extended abstracts
Place of publication: Tokyo
Publisher: Solid State Ionics Society of Japan
Source: orbit
Source-ID: 307047
Research output: Chapter in Book/Report/Conference proceeding › Conference abstract in proceedings – Annual report year: 2004 › Research

Fastoxid-brændselscellen - en elektrokemisk reaktor

General information
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Kammer Hansen, K., Barfod, R., Mogensen, M. B.
Pages: 99-100
Publication date: 2004

Host publication information
Title of host publication: DK2
Place of publication: Lyngby
Publisher: Danmarks Tekniske Universitet, Institut for Kemiteknik
Editors: Johannessen, T., Gani, R., Dam-Johansen, K.
ISBN (Print): 87-91435-05-6
Source: orbit
Source-ID: 310304
Research output: Chapter in Book/Report/Conference proceeding › Conference abstract in proceedings – Annual report year: 2004 › Research

Oxidation of methane on oxides

General information
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Kammer Hansen, K., Mogensen, M. B.
Publication date: 2004
Peer-reviewed: No
Event: Abstract from Final workshop on OSSEP - Optimisation of solid state FC processes for all aspects of hydrocarbon oxidation, Tenerife (ES), 10-14 Nov, .
Source: orbit
Source-ID: 307620
Research output: Contribution to conference › Conference abstract for conference – Annual report year: 2004 › Research
SOFC - an electrochemical reactor

Conversion of hydrocarbons in solid oxide fuel cells
Recently, a number of papers about direct oxidation of methane and hydrocarbon in solid oxide fuel cells (SOFC) at relatively low temperatures (about 700°C) have been published. Even though the conversion of almost dry CH4 at 1000°C on ceramic anodes was demonstrated more than 10 years ago, the reports about high-current densities for methane oxidation at such low temperatures are indeed surprising. Several papers indicate that a catalytic effect (due to the mixed ionic and electronic conductivity) of CeO2-x is partially responsible for this effect. However, this seems to contradict previous reports, and thus this issue deserves further analysis.

Electrodes for oxidation of methane

Improvement of LSM cathode for high power density SOFCs

**General information**
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Wang, W., Barfod, R., Larsen, P., Kammer Hansen, K., Bentzen, J., Hendriksen, P., Mogensen, M. B.
Pages: 400-408
Publication date: 2003

**Host publication information**
Title of host publication: Proceedings
Place of publication: Pennington, NJ
Publisher: The Electrochemical Society
Editors: Singhal, S., Dokiya, M.
Source: orbit
Source-ID: 306410
Research output: Chapter in Book/Report/Conference proceeding – Article in proceedings – Annual report year: 2003 – Research

Development of thin-electrolyte solid oxide fuel cells

**General information**
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Kammer Hansen, K., Larsen, P., Liu, Y., Kindl, B., Mogensen, M. B.
Pages: 875-882
Publication date: 2002

**Host publication information**
Title of host publication: Proceedings. Vol. 2
Place of publication: Oberrohrdorf (CH)
Publisher: European Fuel Cell Forum
Editor: Huijsmans, J.
ISBN (Print): 3-905592-10-X
Source: orbit
Source-ID: 304283
Research output: Chapter in Book/Report/Conference proceeding – Article in proceedings – Annual report year: 2002 – Research

Electrochemical gas cleaning

**General information**
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Kammer Hansen, K.
Publication date: 2002
Peer-reviewed: No
Source: orbit
Source-ID: 304843
Research output: Contribution to conference – Conference abstract for conference – Annual report year: 2002 – Research

Strategies for testing of solid oxide fuel cells and electrodes

**General information**
Publication status: Published
Organisations: Risø National Laboratory for Sustainable Energy
Contributors: Mogensen, M. B., Hendriksen, P., Kammer Hansen, K.
Pages: 893-902
Publication date: 2002

**Host publication information**
Working electrode for electrochemical reactor comprises electric conductive ceramic oxide material

Perovskites as catalysts for the selective catalytic reduction of nitric oxide with propene: Relationship between solid state properties and catalytic activity

The influence of the solid state properties of manganese- and iron-based perovskites on the activity for the selective catalytic reduction of nitric oxide with propene under oxidizing conditions has been investigated. By substituting manganese for iron in the solid solution series $La_{0.6}Sr_{0.4}Fe_{1-y}Mn_yO_{3+δ}$ (0≤y≤1) and by substituting strontium for lanthanum in the solid solution series $La_{1-x}Sr_xMnO_{3+δ}$ (0.05≤x≤0.50) it is shown that the catalytic activity of the perovskites depends mainly on the redox properties of the transition metal and on the number of oxygen vacancies. © 2001 Academic Press
Electrochemical Exhaust Gas Purification
This text is divided into two parts; the first part gives a short recapitulation of the development status of the electrochemical reactor for soot removal, while the second part gives a description of activities on electrochemical NOx reduction. The Electrochemical Reactor for filtration and continuous combustion of soot from diesel exhaust gas has been described earlier [1, 2]. The reactor size has been increased by stacking flat plate reactors, and full size test has been performed on a diesel engine in a test bench. The soot removal efficiency is better than 90%, but the efficiency for gas oxidation (conversion of carbon monoxide (CO) and hydrocarbons (CH)) is still low, of the order of 50%. The packing and mounting of the reactor is under development to avoid mechanical breakage by engine vibrations. This has improved the durability significantly, and on road vehicle test is under initiation. Optimisation of reactor shape and materials are going on continuously to improve efficiency and durability. In this phase various production methods are under consideration. Activity to extend the electrochemical principle to lean NOx removal is increasing. In this work a concept for electrochemical removal of NOx under lean conditions is outlined. The main obstacles to the design of an effective and energy efficient electrochemical DeNOx reactor are presented. The use of a selective electrode in combination with the need to be able to control the applied potential and, following from this, the need for a simple geometry of the reactor is stressed. A selective electrode for nitric oxide reduction has been identified.

General information
Publication status: Published
Organisations: Dinex A/S, Technical University of Denmark, University of Southern Denmark
Number of pages: 5
Publication date: 2000

Host publication information
Title of host publication: Diesel Exhaust Aftertreatment 2000 : SAE 2000 World Congress
Publisher: Society of Automotive Engineers (S A E Technical Papers; No. 2000-01-0478).
Source: dtu
Source-ID: u::10599
Research output: Chapter in Book/Report/Conference proceeding › Article in proceedings – Annual report year: 2000 › Research › peer-review

Electrochemical Reduction of NO and O2 on Cu/CuO

General information
Publication status: Published
Organisations: Department of Chemistry
Contributors: Kammer Hansen, K., Christensen, H., Skou, E. M., Skaarup, S.
Pages: 193-200
Publication date: 2000
Peer-reviewed: Yes

Publication information
Journal: Journal of Applied Electrochemistry
Volume: 30
ISSN (Print): 0021-891X
Ratings:
Scopus rating (2000): SJR 0.685 SNIP 1.007
Web of Science (2000): Indexed yes
Original language: English
Source: orbit
Source-ID: 176768
Research output: Contribution to journal › Journal article – Annual report year: 2000 › Research › peer-review

Electrochemical Reduction of NO and O2 on Oxide Based Electrodes
Cyclic voltammetry has been used to investigate the electrocatalytic properties of oxide based electrodes for the reduction of NO and oxygen. Perovskites of compositions La$_{x}$Sr$_{1-x}$MnO$_{3+δ}$ (LSMx), with 0.05 ≤ x ≤ 0.5 and La$_{0.6}$Sr$_{0.4}$Fe$_{0.8}$Mn$_{0.2}$O$_{3-δ}$ (LSFM), the spinels Co$_{3}$O$_{4}$ and CuCr$_{2}$O$_{4}$ and the mixed oxide V$_{2}$O$_{5}$ have been selected and investigated in the temperature range 300 - 500 ºC. The materials show good catalytic properties against both NO oxidation and NO reduction. The catalytic activity as well as the selectivity is dependent both on the redox behavior of the compound and on the oxygen defect concentration.

General information
Publication status: Published
Organisations: Technical University of Denmark, Dinex A/S, University of Southern Denmark
Perovskites as Cathodes for Nitric Oxide Reduction
Using cone shaped electrodes, the electrochemical reduction of nitric oxide and oxygen has been investigated by cyclic
voltammetry on an oxygen overstoichiometric (La$_{0.85}$Sr$_{0.15}$MnO$_{3+δ}$), and an oxygen stoichiometric (La$_{0.85}$Sr$_{0.15}$CoO$_{3-δ}$)
perovskite over the temperature range 300-500°C. An oxygen ion-conducting 10% gadolinium-doped cerium oxide is
used as electrolyte. It is shown that the reduction of nitric oxide proceeds rapidly on La$_{0.85}$Sr$_{0.15}$MnO$_{3+δ}$ compared to
the oxygen reduction while the oxygen reduction on La$_{0.85}$Sr$_{0.15}$CoO$_{3-δ}$ is faster than the nitric oxide reduction. © 2000
The Electrochemical Society. S0013-4651(99)09-054-0. All rights reserved.

Electrochemical Reactor for Exhaust Gas Purification
A concept for an electrochemical reactor acting as a trap for the removal of soot particles from diesel exhaust gas has
been developed and presented earlier [1]. Only small scale flat plate samples tested with synthetic exhaust gas was
presented. Since then, the sample size has been increased, and test on a diesel engine in a test bench has been carried
out. Various concepts for the establishment of a sufficient filtering surface and for the electrical connections have been
tested, and the construction of a muffler with an electrochemical reactor installed has been initiated. This is to be on a
passenger car for on-road test. The preliminary bench test indicates a soot removal efficiency of 75-90% with no
accumulation of soot at the reactor, at temperatures above 250°C. A separate project has been started to evaluate the
possibilities of lean NOx removal on a similar reactor. Results from this will be reported separately.
Mechanochemical synthesis of Fe-S materials.
Powder mixtures of metallic iron and sulfur have been milled in a high-energy planetary ball mill and the formation of iron sulfides has been studied by x-ray diffraction, Mossbauer spectroscopy, and scanning electron microscopy. For Fe:S ratios of 1:1 and 1:2 the final products are FeS with the modified NiAs structure and FeS2 (pyrite), respectively. No other iron sulfides were formed for any of the Fe:S ratios studied. The FeS phase has been tested as an electrode material in lithium batteries.

General information
Publication status: Published
Organisations: Department of Physics, Department of Chemistry, Department of Chemistry
Contributors: Jiang, J., Larsen, R., Lin, R., Mørup, S., Chorkendorff, I., Nielsen, K., Kammer Hansen, K., West, K.
Pages: 114-125
Publication date: 1998
Peer-reviewed: Yes

Lithium Insertion into Iron Sulfides

General information
Publication status: Published
Organisations: Department of Chemistry
Contributors: Kammer Hansen, K., West, K.
Pages: 124-132
Publication date: 1997

Host publication information
Title of host publication: Proceedings of the Symposium on Batteries for Portable Applications and Electric Vehicles
Place of publication: Paris
Publisher: The Electrochemical Society
Source: orbit
Source-ID: 168552
Research output: Contribution to journal › Journal article – Annual report year: 1997 › Research

Projects:

Durable thin ceramic films for improvement of Proton Exchange Membrane (PEM) electrolysis
Fenini, F., PhD Student, Department of Energy Conversion and Storage
Mogensen, M. B., Main Supervisor
Kammer Hansen, K., Supervisor
Hendriksen, P. V., Supervisor
Li, Q., Examiner
Carmo, M., Examiner
Friedrich, K. A., Examiner
Forskningsrådsfinansiering  
15/10/2015 → 10/12/2018  
Award relations: Durable thin ceramic films for improvement of Proton Exchange Membrane (PEM) electrolysis  
Project: PhD

Elektrokatalytisk gasrensning  
Kammer Hansen, K., PhD Student, Department of Chemistry  
Skaarup, S., Main Supervisor  
Zachau-Christiansen, B., Supervisor  
Mogensen, M. B., Examiner  
Kilner, J., Examiner  
Koksbang, R., Examiner  
Innovationsfonden  
01/12/1997 → 05/07/2001  
Award relations: Elektrokatalytisk gasrensning  
Project: PhD

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

Selective NO Conversion Electrocatalysts  
Werchmeister, R. M. L., PhD Student, Risø National Laboratory for Sustainable Energy  
Mogensen, M. B., Main Supervisor  
Kammer Hansen, K., Supervisor  
Rossmeisl, J., Supervisor  
Skaarup, S., Examiner  
Metcalf, I. S., Examiner  
Primdahl, S., Examiner  
Christensen, H., Supervisor  
Grisen, G., Supervisor  
Forskningsrådsstipendium  
01/03/2007 → 04/08/2010  
Award relations: Selective NO Conversion Electrocatalysts  
Project: PhD

Direct conversion of carbon to electricity in a hybrid solid oxide fuel cell  
Deleebeeck, L., PhD Student, Department of Energy Conversion and Storage  
Kammer Hansen, K., Main Supervisor  
Mogensen, M. B., Supervisor  
Hagen, A., Examiner  
Lagergren, C., Examiner  
Grisen, G., Examiner  
EU-finansieret  
01/03/2012 → 03/06/2015  
Award relations: Direct conversion of carbon to electricity in a hybrid solid oxide fuel cell  
Project: PhD

Oxygen Membranes for Biomass Gasification and Cement Industry  
Cheng, S., PhD Student, Department of Energy Conversion and Storage  
Hendriksen, P. V., Main Supervisor  
Kaiser, A., Supervisor  
Kammer Hansen, K., Examiner  
Yu, J. H., Examiner  
Guillon, O., Examiner
**Low Temperature oxidation of hydrocarbons using an electrochemical reactor**
Ippolito, D., PhD Student, Department of Energy Conversion and Storage
Kammer Hansen, K., Main Supervisor
Bonanos, N., Examiner
Marnellos, G. E., Examiner
Christensen, H., Supervisor
Grisen, G., Examiner
01/05/2010 → 19/08/2013
Award relations: Low Temperature oxidation of hydrocarbons using an electrochemical reactor
Project: PhD

**Experimental tape casting of multilayer for flue gas purification**
Schmidt, C. G., PhD Student, Department of Energy Conversion and Storage
Kaiser, A., Main Supervisor
Kammer Hansen, K., Supervisor
Chatzichristodoulou, C., Examiner
Görsman, C. F., Examiner
Graule, T. J., Examiner
Andersen, K. B., Supervisor
15/05/2011 → 30/09/2015
Award relations: Experimental tape casting of multilayer for flue gas purification
Project: PhD

**Electrochemical Reduction of NOx**
Traulsen, M. L., PhD Student, Department of Energy Conversion and Storage
Kammer Hansen, K., Main Supervisor
Holtappels, P., Examiner
Rasmussen, S. B., Examiner
Grisen, G., Examiner
01/05/2009 → 27/06/2012
Award relations: Electrochemical Reduction of NOx
Project: PhD