Complementary analyses of aging in a commercial LiFePO$_4$/graphite 26650 cell

In this work we investigate the electrode degradation mechanisms in a commercial 2.5 Ah LiFePO$_4$/graphite 26650 cylindrical cell. Aged and fresh electrode samples were prepared by cycling two cells respectively five and 22 k times. Subsequently the cells were disassembled in a glovebox and the electrode samples were prepared for electrochemical testing in a 3-electrode setup, and for characterization with XRD, XPS and low-kV FIB/SEM tomography. A 1 μm thick CEI (cathode electrolyte interface) layer was observed at the electrode/electrolyte interface of the aged LiFePO$_4$ electrode. Relative to the fresh LiFePO$_4$ electrode, the aged electrode exhibited a larger series resistance which indicates the observed degradation layer increases the ionic resistance. In addition, micron-sized agglomerates, probably a mixture of carbonaceous material and decomposition products from the electrolyte, were observed at the electrode/electrolyte interface of the aged graphite electrode. These layers may contribute significantly to the loss of lithium inventory (LLI) in the cell, and to the loss of active material (LAM) in the graphite electrode. Low-voltage FIB/SEM tomography was used to detect local charging effects of graphite particles in the carbon electrode, an effect of poor dissipation of the electric charge to the ground after the sample interaction with the electron beam. The charging effects were primarily observed in the aged electrode and most of the locally charged particles were found to be close to the electrode/electrolyte interface, indicating a poorly percolating graphite network near this interface.
Compared to conventional preparation methods for supported heterogeneous catalysts, the use of colloidal nanoparticles (NPs) allows for a precise control over size, size distribution, and distribution/location of the NPs on the support. However, common colloidal syntheses have restrictions that limit their applicability for industrial catalyst preparation. We present a
simple, surfactant-free, and scalable preparation method for colloidal NPs to overcome these restrictions. We demonstrate how precious-metal NPs are prepared in alkaline methanol, how the particle size can be tuned, and how supported catalysts are obtained. The potential of these colloids in the preparation of improved catalysts is demonstrated by two examples from heterogeneous catalysis and electrocatalysis.

**General information**
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Organisations: Department of Energy Conversion and Storage, Imaging and Structural Analysis, University of Bremen, University of Copenhagen, University of Bern
Authors: Quinson, J. (Ekstern), Neumann, S. (Ekstern), Wannmacher, T. (Ekstern), Kacenaukaite, L. (Ekstern), Inaba, M. (Ekstern), Bucher, J. (Ekstern), Bizzotto, F. (Ekstern), Simonsen, S. B. (Intern), Theil Kuhn, L. (Intern), Bujak, D. (Ekstern), Zana, A. (Ekstern), Arenz, M. (Ekstern), Kunz, S. (Ekstern)
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ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
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BFI (2011): BFI-level 2
Scopus rating (2011): SJR 6.063 SNIP 2.361 CiteScore 10.75
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Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 4.528 SNIP 1.888
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Continuous Hydrothermal Flow Synthesis of LaCrO3 in Supercritical Water and Its Application in Dual-Phase Oxygen Transport Membranes

The continuous production of LaCrO3 particles (average edge size 639 nm, cube-shaped) by continuous hydrothermal flow synthesis using supercritical water is reported for the first time. By varying the reaction conditions, it was possible to suggest a reaction mechanism for the formation of this perovskite material. Moreover, dual-phase oxygen transport membranes were manufactured from the as-synthesized LaCrO3 particles and (ZrO2)0.89(Y2O3)0.01(Sc2O3)0.10 (10Sc1YSZ), and oxygen permeation fluxes up to 5 × 10–8 mol cm–2 s–1 were measured on a 1 mm thick membrane.

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Authors: Xu, Y. (Intern), Pirou, S. (Intern), Zielke, P. (Intern), Simonsen, S. B. (Intern), Norby, P. (Intern), Hendriksen, P. V. (Intern), Kiebach, R. (Intern)
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Web of Science (2016): Indexed yes
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Scopus rating (2015): SJR 0.938 SNIP 1.145 CiteScore 2.87
Web of Science (2015): Indexed yes
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Scopus rating (2014): SJR 1.009 SNIP 1.287 CiteScore 2.85
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Investigating particle size effects in catalysis by applying a size controlled and surfactant-free synthesis of colloidal nanoparticles in alkaline ethylene glycol - The case study of the oxygen reduction reaction on Pt

Colloidal platinum nanoparticles are obtained via a surfactant-free polyol process in alkaline ethylene glycol. In this popular synthesis, ethylene glycol functions as solvent and reducing agent. The preparation procedure is known for its reproducibility to obtain 1-2 nm nanoparticles, but at the same time the controlled preparation of larger nanoparticles is challenging. A reliable size control without the use of surfactants is a fundamental yet missing achievement for systematic investigations. In this work it is demonstrated how the molar ratio between NaOH and the platinum precursor determines the final particle size and how this knowledge can be used to prepare and study in a systematic way supported catalysts with defined size and metal loading. Using small-angle X-ray scattering, transmission electron microscopy, infra-red spectroscopy, X-ray absorption spectroscopy, pair distribution function and electrochemical analysis it is shown that changing the NaOH/Pt molar ratio from 25 to 3, the Pt nanoparticle size is tuned from 1 to 5 nm. This size range is of interest for various catalytic applications, such as the oxygen reduction reaction (ORR). Supporting the nanoparticles onto a high surface area carbon, we demonstrate how the particle size effect can be studied keeping the catalyst loading.
constant, an important aspect that in previous studies could not be accomplished.

**General information**

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**Organisations:** Department of Energy Conversion and Storage, Imaging and Structural Analysis, University of Copenhagen, University of Oldenburg, University of Bremen, University of Bern

**Authors:** Quinson, J. (Ekstern), Inaba, M. (Ekstern), Neumann, S. (Ekstern), Swane, A. A. (Ekstern), Bucher, J. (Ekstern), Simonsen, S. B. (Intern), Theil Kuhn, L. (Intern), Kirkensgaard, J. J. K. (Ekstern), Jensen, K. M. (Ekstern), Oezaslan, M. (Ekstern), Kunz, S. (Ekstern), Arenz, M. (Ekstern)

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- Scopus rating (2016): CiteScore 10.3 SJR 4.367 SNIP 2.081
- Web of Science (2016): Indexed yes
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- Web of Science (2015): Indexed yes
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**A toolbox to study precious metal nano- catalysts: surfactant free synthesis, characterization and catalytic activity**

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**Organisations:** Department of Energy Conversion and Storage, Imaging and Structural Analysis, University of Copenhagen, University of Oldenburg, University of Bremen

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**Number of pages:** 1

**Publication date:** 2017
Coarsening of carbon black supported Pt nanoparticles in hydrogen

This study addresses coarsening mechanisms of Pt nanoparticles supported on carbon black in hydrogen. By means of in situ transmission electron microscopy (TEM), Pt nanoparticle coarsening was monitored in 6 mbar 20 % H2/Ar while ramping up the temperature to almost 1000 °C. Time-resolved TEM images directly reveal that separated ca. 3 nm sized Pt nanoparticles in a hydrogen environment are stable up to ca. 800 °C at a heating rate of 10 °C/min. The coarsening above this temperature is dominated by the particle migration and coalescence mechanism. However, for agglomerated Pt nanoparticles, coalescence events were observed already above 200 °C. The temperature-dependency of particle sizes and the observed migration distances are described and found to be consistent with simple early models for the migration and coalescence.

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Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
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Web of Science (2014): Indexed yes
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ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): SJR 1.861 SNIP 1.307 CiteScore 3.34
ISI indexed (2012): ISI indexed yes
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BFI (2011): BFI-level 1
Scopus rating (2011): SJR 1.899 SNIP 1.451 CiteScore 3.86
Continuous Hydrothermal Flow Synthesis of Functional Oxide Nanomaterials Used in Energy Conversion Devices

Continuous hydrothermal flow synthesis (CHFS) was used to prepare functional oxide nanoparticles. Materials synthesized include NiO, Y-doped ZrO2, Gd-doped CeO2, LaCrO3 and Ni-substituted CoFe2O4. These types of oxides can be applied in several energy conversion devices, e.g. as active materials in solid oxide fuel cells (SOFCs), oxygen transport membranes (OTMs), and water electrolysis processes. Compared with other chemical synthesis methods, CHFS is advantageous for preparing nanosized materials with a narrow size distribution and a high phase purity. Moreover, CHFS has a high throughput as materials are continuously produced, and the technology can be scaled-up to an industrial-relevant production capacity.

The thesis starts with investigating the most appropriate mixer design for a novel two-stage reactor by computational fluid dynamics modelling. Based on the modelling results, a two-stage CHFS reactor was constructed, and proof-of-concept syntheses of NiO and Y-doped ZrO2 (YSZ), in both one-stage and two-stage modes, were conducted.

Secondly, Gd-doped CeO2 (GDC) nanoparticles (6 – 40 nm) were synthesized, and the effect of the pH on their size, morphology and composition was studied. An up-scaled synthesis of Gd0.2Ce0.8O2-δ nanoparticles was made, and the as-synthesized particles were processed into inks that displayed a good printability in inkjet printing of electrolytes on both green NiO-GDC and pre-sintered NiO-YSZ substrates. Particularly on the pre-sintered NiO-YSZ substrate, dense continuous layers (< 2 μm in thickness) composed of GDC and YSZ that fully covered the substrate were obtained after firing at 1300 °C.

In addition, (La0.6Sr0.4)0.99CoO3 – Gd0.2Ce0.8O2-δ (LSC-GDC) core-shell type particles were prepared and their high-temperature microstructural evolution was studied in two different sintering processes, i.e. spark plasma sintering and conventional sintering. By conventional hydrothermal batch-type synthesis, a core-shell structure was realized by precipitating ~6 nm large GDC particles on the surface of LSC particles under a mild hydrothermal condition of 100 °C resulting in an integral GDC shell around the LSC core. It was found that by spark plasma sintering, a fine microstructure containing nanograins could be obtained and the graded core-shell architecture could be partially maintained.

For the first time, phase-pure LaCrO3 was obtained by CHFS without any post treatments. A continuous production of cube-shaped LaCrO3 particles (639 nm) was achieved. Processing parameters (temperature and alkali concentration)
were found to affect the phase purity of the obtained particles significantly. The synthesized LaCrO\(_3\) particles were used to prepare 10Sc\(_1\)YSZ – LaCrO\(_3\) dual-phase oxygen transport membranes. A density of ~90 % was achieved after firing at 1400 °C, which is below normal sintering temperatures for LaCrO\(_3\)-based ceramics. Oxygen permeation fluxes up to 5 \times 10^{-8} \text{ mol cm}^{-2} \text{ s}^{-1} were obtained with a 1 mm thick membrane tested in air/N\(_2\) at 900 °C. Finally, CoFe\(_2\)O\(_4\) and Ni-substituted CoFe\(_2\)O\(_4\) nanoparticles were prepared by CHFS and their catalytic properties were evaluated. The CoFe\(_2\)O\(_4\) was found to be active for catalytic CO oxidation. A 50 % conversion of CO at 223 °C and a complete conversion at 310 °C was reached. By CHFS, Ni-substituted CoFe\(_2\)O\(_4\) nanoparticles with controlled Ni contents were synthesized, and their activity as catalysts for the oxygen evolution reaction (the half reaction of water electrolysis) was evaluated. Whereas no simple correlation between the activity and the Ni content was detected, a remarkable improvement of the activity was observed for the sample with 30 at% (in atomic percent) Ni-substituted CoFe\(_2\)O\(_4\) (Co0.7Ni0.3Fe2O4) compared to all other investigated compositions.

Overall, the thesis demonstrates the versatility of the CHFS route for preparing functional oxides in nano particulate form, and documents the properties of the synthesized materials in a number of specific applications (fuel cells, membranes and catalysis).

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Continuous Hydrothermal Flow Synthesis of Functional Oxide Nanomaterials Used in Energy Conversion Devices

**In situ TEM analysis of a symmetric solid oxide cell in oxygen and vacuum–cation diffusion observations**
In order to establish the use of solid oxide fuel/electrolysis cells (SOFC/SOEC) in the energy market, a deeper understanding of degradation effects during operation is necessary. This study apply in situ transmission electron microscopy (TEM) of a symmetric model cell composed by two oxygen electrodes of La\(_{0.6}\)Sr\(_{0.4}\)CoO\(_{3-\delta}\) (LSC) and an electrolyte, ZrO\(_2\); 8% mol Y\(_2\)O\(_3\) (8YSZ), deposited on 1% Nb doped SrTiO\(_3\)\(_{\delta}\) (STN) single crystal substrate by pulsed laser deposition. The results showed a high cation mobility of the electrodes when exposed to 900 °C. Cobalt is found to agglomerate at the interface between LSC and STN. Strontium depletion is observed in both electrodes. Finally, a faster grain growth occurs in LSC for the cell exposed to oxygen in comparison with the cell in vacuum.

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Organisations: Department of Energy Conversion and Storage, Imaging and Structural Analysis, Center for Electron Nanoscopy, Electrofunctional materials, Nagoya University
Authors: Gualandris, F. (Intern), Simonsen, S. B. (Intern), Wagner, J. B. (Intern), Sanna, S. (Intern), Muto, S. (Ekstern), Kuhn, L. T. (Intern)
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In situ TEM study of the coarsening of carbon black supported Pt nanoparticles in hydrogen

The control of sizes and shapes of nanostructures is of tremendous importance for the catalytic activity in electrochemistry and in catalysis more generally. However, due to relatively large surface free energies, nanostructures often sinter to form coarser and more stable structures that may not have the intended physicochemical properties.

Pt is known to be a very active catalyst in several chemical reactions and for example as carbon supported nanoparticles in fuel cells.

The presentation focusses on coarsening mechanisms of Pt nanoparticles supported on carbon black during exposure to hydrogen. By means of in situ transmission electron microscopy (TEM), Pt nanoparticle coarsening was monitored in 6 mbar 20 % H₂/Ar while ramping up the temperature to ca. 900 °C. Time-resolved TEM images directly reveal that separated ca. 3 nm sized Pt nanoparticles in the pure hydrogen environment are stable during constant temperature ramping by 10°C/min up to ca. 800 °C. The coarsening above this temperature is fully dominated by the particle migration and coalescence mechanism. This is contrary to supported Pt nanoparticles in oxygen, where the coarsening is fully dominated by Ostwald ripening. For agglomerated Pt nanoparticles, coalescence events were observed already at ca. 200 °C. The temperature-dependency of particle sizes and the observed migration distances are consistent with simple early models for the migration and coalescence.

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In Situ Transmission Electron Microscopy in Materials Science - Possibilities and Prospects

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On the Properties and Long-Term Stability of Infiltrated Lanthanum Cobalt Nickelates (LCN) in Solid Oxide Fuel Cell Cathodes

Infiltration as a fabrication method for solid oxide fuel cells (SOFC) electrodes is offering significant improvements in cell performance at reduced materials and fabrication costs, especially when combined with co-sintering. However, important questions regarding the long-term performance and microstructural stability remain unanswered. Here, we present the results of a three-year project, where large footprint anode-supported SOFCs with a co-sintered cathode backbone and infiltrated La0.95Co0.4Ni0.6O3 (LCN) cathodes were developed and thoroughly characterized. The initial long-term performance and stability of this new cell type was investigated for 1500+ hours, coupled with STEM-EDS investigation of the microstructural changes in the infiltrated electrodes. Additionally, electrodes were further aged at elevated temperatures (750 - 900°C) for periods reaching up to 5000 hours, while following changes in the electrode properties using SEM, BET area, and in-plane conductivity measurements. Finally, the mechanical properties of co-sintered cathode backbone cells were determined in four-point bending tests carried out both at room temperature and at 800°C in air. Based on these results, degradation mechanisms were identified and recommendation for safe operation conditions in real life application could be formulated.

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Scopus rating (2016): CiteScore 2.97 SJR 1.222 SNIP 0.963
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 1.115 SNIP 1.066 CiteScore 3.17
Web of Science (2015): Indexed yes
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Scopus rating (2014): SJR 1.213 SNIP 1.25 CiteScore 3.36
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 1.169 SNIP 1.309 CiteScore 2.92
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BFI (2012): BFI-level 1
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Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): SJR 1.331 SNIP 1.335 CiteScore 2.74
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Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 1.418 SNIP 1.304
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Scopus rating (2009): SJR 1.442 SNIP 1.27
Web of Science (2009): Indexed yes
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Scopus rating (2008): SJR 1.595 SNIP 1.41
Web of Science (2008): Indexed yes
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Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 1.608 SNIP 1.535
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 1.523 SNIP 1.481
Web of Science (2005): Indexed yes
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Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 1.971 SNIP 1.677
Web of Science (2003): Indexed yes
Scopus rating (2002): SJR 2.036 SNIP 1.618
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Web of Science (2000): Indexed yes
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Original language: English
Structural evolution during calcination and sintering of a \((\text{La}_{0.6}\text{Sr}_{0.4})_{0.99}\text{CoO}_{3-\delta}\) nanofiber prepared by electrospinning

Design of 3-dimensional metal oxide nanofibers by electrospinning is being widely explored. However, the impacts of calcination and sintering on the resulting morphology remain unknown. For the first time, \((\text{La}_{0.6}\text{Sr}_{0.4})_{0.99}\text{CoO}_{3-\delta}\) (LSC) nanofiber, which is among the most promising electrode materials for solid oxide fuel cells, was synthesized by sol-gel electrospinning. By elevating the temperature in oxygen using in situ transmission electron microscopy, we discovered the structural transitions from nanofibers to nanotubes and then to nano-pearl strings. This facile and up-scaleable method can be widely applied to design metal oxide one-dimensional nanomaterials with precise control in both geometry (nanofiber, nanotube and nano-pearl string) and surface area (by varying grain size).
A TEM study of morphological and structural degradation phenomena in LiFePO$_4$-CB cathodes: Morphological and structural degradation in LiFePO$_4$-CB cathodes

LiFePO$_4$-based cathodes suffer from various degradation mechanisms, which influences the battery performance. In this paper, morphological and structural degradation phenomena in laboratory cathodes made of LiFePO$_4$ mixed with carbon black (CB) in a 1 mol/L LiPF$_6$ in EC:DMC (1:1 by weight) electrolyte are investigated by transmission electron microscopy at various preparation, assembling, storage, and cycling stages. High-resolution transmission electron microscopy imaging shows that continuous SEI layers are formed on the LiFePO$_4$ particles and that both storage and cycling affect the formation. Additionally, loss of CB crystallinity, CB aggregation, and agglomeration is observed. Charge–discharge curves and impedance spectra measured during cycling confirm that these degradation mechanisms reduce the cathode conductivity and capacity.

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Coarsening of Pd nanoparticles in an oxidizing atmosphere studied by in situ TEM

The coarsening of supported palladium nanoparticles in an oxidizing atmosphere was studied in situ by means of transmission electron microscopy (TEM). Specifically, the Pd nanoparticles were dispersed on a planar and amorphous Al$_2$O$_3$ support and were observed during the exposure to 10 mbar technical air at 650 °C. Time-resolved TEM image series reveal that the Pd nanoparticles were immobile and that a few percent of the nanoparticles grew or shrunk, indicating a coarsening process mediated by the Ostwald ripening mechanism. The TEM image contrast suggests that the largest nanoparticles tended to wet the Al$_2$O$_3$ support to a higher degree than the smaller nanoparticles and that the distribution of projected particle sizes consequently broadens by the appearance of an asymmetric tail toward the larger particle sizes. A comparison with computer simulations based on a simple mean-field model for the Ostwald ripening process indicates that the observed change in the particle size distribution can be accounted for by wetting of the Al$_2$O$_3$ support by the larger Pd nanoparticles.

General information
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Organisations: Department of Physics, Experimental Surface and Nanomaterials Physics, Center for Individual Nanoparticle Functionality, Haldor Topsoe AS, Chalmers University of Technology
Authors: Simonsen, S. B. (Intern), Chorkendorff, I. (Intern), Dahl, S. (Ekstern), Skoglundh, M. (Ekstern), Helveg, S. (Ekstern)
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Scopus rating (2016): CiteScore 1.85 SJR 0.746 SNIP 0.834
Web of Science (2016): Indexed yes
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Scopus rating (2015): SJR 0.747 SNIP 0.804 CiteScore 1.85
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 0.818 SNIP 0.864 CiteScore 1.81
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
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ISI indexed (2013): ISI indexed yes
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BFI (2012): BFI-level 1
Scopus rating (2012): SJR 1.096 SNIP 0.878 CiteScore 1.91
ISI indexed (2012): ISI indexed yes
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BFI (2011): BFI-level 1
Scopus rating (2011): SJR 1.076 SNIP 0.906 CiteScore 1.88
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 1.228 SNIP 0.858
Web of Science (2010): Indexed yes
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Scopus rating (2009): SJR 1.129 SNIP 0.896
Comparison of ultramicrotomy and focused-ion-beam for the preparation of TEM and STEM cross section of organic solar cells

The challenge of preparing cross sections of organic photovoltaics (OPV) suitable for transmission electron microscopy (TEM) and scanning TEM (STEM) is addressed. The samples were polymer solar cells fabricated using roll-to-roll (R2R) processing methods on a flexible polyethylene terephthalate (PET) substrate. Focused ion beam (FIB) and ultramicrotomy were used to prepare the cross sections. The differences between the samples prepared by ultramicrotomy and FIB are addressed, focusing on the advantages and disadvantages of each technique. The sample prepared by ultramicrotomy yielded good resolution, enabling further studies of phase separation of P3HT:PCBM by means of energy filtered TEM (EFTEM). The sample prepared by FIB shows good structure preservation, but reduced resolution due to non-optimal thicknesses achieved after treatment. Degradation studies of samples prepared by ultramicrotomy are further discussed, which reveal particular effects of the ISOS-L-3 aging test (85 °C, 50% R.H., 0.7 Sun) onto the sample, especially pronounced in the silver layer.

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Effects of strong cathodic polarization of the Ni-YSZ interface

Long-term strong cathodic polarization experiments of down to -2.4 V vs. $E^\circ(O_2)$ of the Ni-YSZ interface were performed at 900°C in 97% H₂/3% H₂O on model electrodes. The Ni-YSZ interface underwent extensive changes and a large affected volume with a complex microstructure and phase distribution resulted. Impedance spectroscopy shows initial decrease but later increase in the series resistance and polarization resistance during the 140-160 h of polarization, and significant inductive behavior. An intermetallic Ni-Zr phase that formed during polarization was preserved when the polarization was kept during cooling, and was identified post-mortem by transmission electron microscopy as Ni₇Zr₂. ZrO₂ nanoparticles were formed on the Ni-gas surface next to the Ni-YSZ-gas triple phase boundary. Explanations of the observed features are offered based on electron microscopy and impedance spectroscopy.

General information

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Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 2.97 SJR 1.222 SNIP 0.963
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 1.115 SNIP 1.066 CiteScore 3.17
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 1.213 SNIP 1.25 CiteScore 3.36
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ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): SJR 1.329 SNIP 1.281 CiteScore 2.61
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): SJR 1.331 SNIP 1.335 CiteScore 2.74
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 1.418 SNIP 1.304
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 1
Electron microscopy investigations of changes in morphology and conductivity of LiFePO4/C electrodes

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

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Degradation mechanism, Focused ion beam scanning electron microscopy, Loss in electron percolation, Low accelerating voltage, Three-dimensional analysis of LiFePO4/Carbon electrode
High-Performance Microchanneled Asymmetric Gd_{0.1}Ce_{0.9}O_{1.95-\delta}-La_{0.6}Sr_{0.4}FeO_{3-\delta}-Based Membranes for Oxygen Separation

A microchanneled asymmetric dual phase composite membrane of 70 vol % Gd_{0.1}Ce_{0.9}O_{1.95-\delta}-30 vol % La_{0.6}Sr_{0.4}FeO_{3-\delta} (CGO-LSF) was fabricated by a "one step" phase-inversion tape casting. The sample consists of a thin dense membrane (100 μm) and a porous substrate including "finger-like" microchannels. The oxygen permeation flux through the membrane with and without catalytic surface layers was investigated under a variety of oxygen partial pressure gradients. At 900 degrees C, the oxygen permeation flux of the bare membrane was 1.6 (STP) ml cm(-2) min(-1) for the air/He-case and 10.10 (STP) ml cm(-2) min(-1) for the air/CO-case. Oxygen, flux measurements as well as electrical conductivity relaxation show that the oxygen flux through the bare membrane without catalyst is limited by the oxygen surface exchange. The surface exchange can be enhanced by introduction of catalyst on the membrane surface. An increase of the 50 150 oxygen flux of-4.49 (STP) mL cm(-2) min(-1) at 900 degrees C was observed when catalyst is added for the air/He-case. Mass transfer polarization through the finger-like support was confirmed to be negligible, which benefits the overall performance. A stable flux of 7.00 (STP) ml cm(-2) min(-1) was observed between air/CO/CO2 over 200 h at 850 degrees C. Partial surface decomposition was observed on the permeate side exposed to CO, in line with predictions from thermodynamic calculations. In a mixture of CO, CO2, H-2, and H2O at similar oxygen activity the material will according to the calculation not decompose. The microchanneled asymmetric CGO-LSF membranes show high oxygen permeability and chemical stability under a range of technologically relevant oxygen potential gradients.
In-Situ Transmission Electron Microscopy on Operating Electrochemical Cells

Solid oxide cells (SOC) have the potential of playing a significant role in the future efficient energy systems scenario. In order to become widely commercially available, an improved performance and durability of the cell has to be achieved [1]. Conventional scanning and transmission SEM and TEM have been often used for ex-situ post-mortem characterization of SOFCs and SOECs [2,3]. However, in order to get fundamental insight of the microstructural development of SOFC/SOEC during operation conditions in-situ studies are necessary [4]. The development of advanced TEM chips and holders makes it possible to undertake analysis during exposure to the SOFC/SOEC sample of reactive gas flow, elevated temperatures and electrical biasing in combination. This allows the study of nanostructure development under temperature and electrode polarization conditions similar to operation conditions. In this work, we have for the first time performed in-situ analysis of a symmetric cell inside a TEM under different configurations. In order to be able to perform in-situ experiments while drawing a current through the sample, we used a homemade TEM chip [5,6] and an 80-300 kV Titan ETEM (FEI Company) equipped with an image corrector and a differential pumping system. A symmetric cell was prepared by depositing a cell consisting of three thin films on a strontium titanate (STO) single crystal substrate by pulsed laser deposition (PLD). Lanthanum strontium cobaltite La$_{0.6}$Sr$_{0.4}$CoO$_3$-$\delta$ (LSC) was chosen as electrode and yttria stabilized zirconia ZrO$_2$: 8% mol Y$_2$O$_3$ (YSZ) as electrolyte, see figure 1. High-resolution TEM analysis on PLD samples after the deposition, did not reveal any second phase formation at the interface between YSZ and LSC. The in-situ experiment was firstly conducted in vacuum at temperature between 250°C and 900°C. Secondly, it was repeated in presence of oxygen with an oxygen partial pressure of about 2 mbar and a maximum temperature of 750°C. Subsequently, the symmetric cell was exposed to oxygen at 600°C and 1 V overpotential within the ETEM. In order to do that, a symmetric cell has been placed on the chip with the use of a focus ion beam (FIB) microscope, see figure 2. To do so, a lamella was first extracted by the bulk sample and attached to a conventional TEM grid. Afterwards, the grid was tilted by 90 degrees and the lamella was detached once again and placed on the chip. STEM-EDS investigation was used for ex-situ post-mortem analysis. Finally, a bulk symmetric cell, coming from the same batch as the in-situ treated TEM samples, was tested in a furnace with similar environmental conditions. This comparison is vital for distinguishing possible surface diffusion effects caused by having a thin lamella for in-situ TEM analysis. Electrochemical properties were also investigated by electrochemical impedance spectroscopy (EIS). In the figure 3 the cell was heat treated at 400°C in vacuum, whereas in figure 4, the cell was treated at the same temperature but in presence of oxygen, with PO$_2$ of 2 mbar. Comparing the two figures, the cell exposed to oxygen showed structural changes in the LSC thin film in comparison with the sample heated in vacuum. These changes refer to the formation of grains as is confirmed by electron diffraction patterns.
In SITU Transmission Electron Microscopy on Operating Electrochemical CELLS

Solid oxide cells (SOC) have the potential of playing a significant role in the future efficient energy system scenario. In order to become widely commercially available, an improved performance and durability of the cells has to be achieved [1]. Conventional scanning and transmission SEM and TEM have been often used for ex-situ post mortem characterization of SOFCs and SOECs [2,3]. However, in order to get fundamental insight of the microstructural development of SOFC/SOEC during operation conditions in situ studies are necessary [4].

Large CZTS Nanoparticles Synthesized by Hot-injection for Thin Film Solar Cells

Infiltration is a fabrication method that is offering potentially significant improvements in cell performance at reduced materials and fabrication costs, especially when combined with co-sintering. However, important questions regarding the long-term performance and microstructural stability of infiltrated electrodes, as well as the mechanical properties of such electrodes have remained unanswered.
Nanocomposite YSZ-NiO Particles with Tailored Structure Synthesized in a Two-Stage Continuous Hydrothermal Flow Reactor

The increasing amount of fluctuating electricity generation from renewable sources requires a flexible energy system and storage technologies to ensure that energy services can be covered in a stable and affordable manner. In order to become truly independent from fossil fuels, increasing the performance of energy storage and conversion devices such as fuel cells, electrolyzers and batteries is important. One promising approach to further improve these devices is the use of carefully structured nanosized materials. Nano-composite particles combining different materials in advanced geometries like core-shell structures or surface decorated particles could exhibit better performance compared with single phase materials. To obtain such advanced structures is the aim of the ProEco project (www.proeco.dk). In this project, a two-stage continuous reactor is built and used to synthesize such nano-composites.

Here we report on the design of the two-stage continuous hydrothermal flow synthesis reactor and first results on obtaining structured nano-composite consisting of yttria-stabilized zirconia (YSZ) and NiO materials. These materials are commonly applied in the fuel electrodes of today’s state-of-the-art solid oxide fuel and electrolysis cells. The prepared particles were characterized by X-ray powder diffraction, (high resolution) transmission electron microscopy, scanning tunnel transmission microscopy and Raman spectroscopy in order to determine crystal structure, particle size, surface morphology and element distribution.

Scandium-doped zinc cadmium oxide as a new stable n-type oxide thermoelectric material

Scandium-doped zinc cadmium oxide (Sc-doped ZnCdO) is proposed as a new n-type oxide thermoelectric material. The material is sintered in air to maintain the oxygen stoichiometry and avoid instability issues. The successful alloying of CdO with ZnO at a molar ratio of 1 : 9 significantly reduced the thermal conductivity by up to 7-fold at room temperature. By carefully selecting the Sc-dopant concentrations, a high power factor of $7.1 \times 10^{-4}$ W m$^{-1}$ K$^{-2}$ at 1173 K could be obtained. Therefore, the highest ZT = 0.3 at 1173 K was achieved for the $\text{Zn}_{0.9}\text{Cd}_{0.1}\text{Sc}_{0.01}\text{O}_{1.015}$ sample, and it has so far one of the highest ZT values among those reported for ZnO based thermoelectric materials over the temperature range, e.g., its ZT value at 300 K, which is 0.012, is over 1 order of magnitude higher than that of the state-of-the-art nanostructured Al-doped ZnO, which is 0.0013. It suggests that this material is a good candidate for improving the overall conversion efficiencies in oxide thermoelectric modules. Meanwhile, Sc-doped ZnCdO is robust in air at high temperatures, whereas other n-type materials, such as Al-doped ZnO, will experience rapid degradation of their electrical conductivity and ZT.
Simulation, design and proof-of-concept of a two-stage continuous hydrothermal flow synthesis reactor for synthesis of functionalized nano-sized inorganic composite materials

Computational fluid dynamics simulations were employed to evaluate several mixer geometries for a novel two-stage continuous hydrothermal flow synthesis reactor. The addition of a second stage holds the promise of allowing the synthesis of functionalized nano-materials as for example core-shell or decorated particles. Based on the simulation results, a reactor system employing a confined jet mixer in the first and a counter-flow mixer in the second stage was designed and built. The two-stage functionality and synthesis capacity is shown on the example of single- and two-stage syntheses of pure and mixed-phase NiO and YSZ particles.
Synthesis of ligand-free CZTS nanoparticles via a facile hot injection route

Single-phase, ligand-free Cu2ZnSnS4 (CZTS) nanoparticles that can be dispersed in polar solvents are desirable for thin film solar cell fabrication, since water can be used as the solvent for the nanoparticle ink. In this work, ligand-free nanoparticles were synthesized using a simple hot injection method and the precursor concentration in the reaction medium was tuned to control the final product. The as-synthesized nanoparticles were characterized using various techniques, and were found to have a near-stoichiometric composition and a phase-pure kesterite crystal structure. No secondary phases were detected with Raman spectroscopy or scanning transmission electron microscopy energy dispersive x-ray spectroscopy. Furthermore, high resolution transmission electron microscopy showed large-sized nanoparticles with an average diameter of 23 nm ± 11 nm. This approach avoids all organic materials and toxic solvents that otherwise could hinder grain growth and limit the deposition techniques. In addition the synthesis route presented here results in nanoparticles of a large size compared to other ligand-free CZTS nanoparticles, due to the high boiling point of the solvents selected. Large particle size in CZTS nanoparticle solar cells may lead to a promising device performance. The results obtained demonstrate the suitability of the synthesized nanoparticles for application in low cost thin film solar cells.

General information
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Organisations: Department of Photonics Engineering, Optical Microsensors and Micromaterials, Department of Micro- and Nanotechnology, Silicon Microtechnology, Department of Energy Conversion and Storage, Imaging and Structural Analysis, Department of Physics, Experimental Surface and Nanomaterials Physics, Center for Individual Nanoparticle Functionality, Nanyang Technological University
Authors: Mirbagheri, N. (Intern), Engberg, S. L. J. (Intern), Crovetto, A. (Intern), Simonsen, S. B. (Intern), Hansen, O. (Intern), Lam, Y. M. (Ekstern), Schou, J. (Intern)
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Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
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Scopus rating (2015): SJR 1.257 SNIP 1.035 CiteScore 3.07
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): SJR 1.497 SNIP 1.269 CiteScore 3.09
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Scopus rating (2013): SJR 1.602 SNIP 1.231 CiteScore 2.74
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
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Scopus rating (2012): SJR 1.861 SNIP 1.307 CiteScore 3.34
Analysis of the Interphase on Carbon Black Formed in High Voltage Batteries

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

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Authors: Younesi, R. (Intern), Christiansen, A. S. (Intern), Scipioni, R. (Intern), Ngo, D. (Intern), Simonsen, S. B. (Intern), Edström, K. (Ekstern), Hjelm, J. (Intern), Norby, P. (Intern)
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Eliminating degradation in solid oxide electrochemical cells by reversible operation

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Authors: Graves, C. R. (Intern), Ebbesen, S. D. (Intern), Jensen, S. H. (Intern), Simonsen, S. B. (Intern), Mogensen, M. B. (Intern)
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Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Web of Science (2015): Indexed yes
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Scopus rating (2014): SJR 14.956 SNIP 8.905 CiteScore 23.23
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): SJR 16.726 SNIP 9.171 CiteScore 23.3
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): SJR 18.482 SNIP 8.331 CiteScore 21.29
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): SJR 17.79 SNIP 7.876 CiteScore 19.84
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 2
Scopus rating (2010): SJR 18.404 SNIP 7.654
Web of Science (2010): Indexed yes
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Web of Science (2009): Indexed yes
BFI (2008): BFI-level 2
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 10.274 SNIP 5.685
Enhancement of the chemical stability in confined δ-Bi₂O₃

Bismuth-oxide-based materials are the building blocks for modern ferroelectrics¹, multiferroics², gas sensors³, light photocatalysts⁴ and fuel cells⁵,⁶. Although the cubic fluorite δ-phase of bismuth oxide (δ-Bi₂O₃) exhibits the highest conductivity of known solid-state oxygen ion conductors⁵, its instability prevents use at low temperature⁷–¹⁰. Here we demonstrate the possibility of stabilizing δ-Bi₂O₃ using highly coherent interfaces of alternating layers of Er₂O₃-stabilized δ-Bi₂O₃ and Gd₂O₃-doped CeO₂. Remarkably, an exceptionally high chemical stability in reducing conditions and redox cycles at high temperature, usually unattainable for Bi₂O₃-based materials, is achieved. Even more interestingly, at low oxygen partial pressure the layered material shows anomalous high conductivity, equal or superior to pure δ-Bi₂O₃ in air. This suggests a strategy to design and stabilize new materials that are comprised of intrinsically unstable but high-performing component materials.
Environmental TEM study of the dynamic nanoscaled morphology of NiO/YSZ during reduction

The reduction of a metal oxide is often a critical preparation step for activating catalytic behaviour. This study addresses the reduction process of NiO in pure form and in a composite of NiO/yttria-stabilized zirconia (YSZ) in hydrogen relevant for solid oxide electrochemical cells by comparing results from environmental transmission electron microscopy (ETEM) with thermogravimetric analysis (TGA). The temperature dependent reduction profiles obtained from TGA confirm an inhibitive effect from YSZ on the NiO reduction. The ETEM images show the growth of Ni in decaying NiO and reveal the nanoscale morphological changes such as pore formation in NiO above 280°C and densification and collapse of the pore structures above 400°C. The accelerated Ni front in NiO illustrates the auto catalysis of the reaction. A rapid temperature ramping from room temperature to 780°C in hydrogen in 1 second resulted in immediate morphological changes at the nanoscale from dense NiO to dense Ni. The analysis suggests that the inhibitive effect of YSZ on the NiO reduction reaction is not due to a direct local interaction between YSZ and NiO, but instead due to gas and/or mass transport limitations.© 2014 Elsevier B.V. All rights reserved.
Long Term Stability Investigation of Solid Oxide Electrolysis Cell with Infiltrated Porous YSZ Air Electrode Under High Current

The increased interest in stable and low cost electrodes for solid oxide cells (SOC) has driven the research of electrode preparation to infiltration of catalyst material into porous backbone material. The infiltration method enables a reduction of amount of catalyst material and increases its activity, due to high surface area of catalyst nano particles. Advantage of infiltration is also separate production of electrolyte backbone structure with good ionic connectivity and mechanical properties. With this study we present the results of a solid oxide cell with infiltrated porous yttria stabilised zirconia (YSZ) backbone air electrode and Ni/YSZ cermet fuel electrode. The SOC was tested at electrolysis conditions under high current (up to -1 A/cm²). The porous YSZ electrodes was infiltrated with gadolinium-doped ceria oxide (CGO), to act as a barrier layer between the catalyst and the backbone, and perovskite catalyst material. Cobalt doped lanthanum nickelate was used as the perovskite catalyst due to its excellent performance. The cell was tested in steam electrolysis for at least 2000h. This initial test indicate that a stable air electrode was formed, and that the cell performance and stability matches that of a state-of-the-art SOC.

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Authors: Veltzé, S. (Intern), Ovtar, S. (Intern), Simonsen, S. B. (Intern), Thydén, K. T. S. (Intern), Kiebach, W. (Intern), Küngas, R. (Ekstern)
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Low - voltage FIB/SEM Tomography for 3D Microstructure Evolution of LiFePO4/C Electrode

Li-ion batteries find widespread use in many electricity storage applications, from portable devices to electric vehicles [1-3], and LiFePO4 (LFP) is one of the most common cathodes because of its long durability and high safety [4, 5] but, since its low ionic and electronic conduction, it is always mixed with carbon black (CB) additives to increase electronic percolation in the electrode. Focused Ion Beam (FIB)/Scanning Electron Microscopy (SEM) Tomography is one of the most used techniques for the study of the three-dimensional microstructure of porous electrodes [6-8]. Imaging at low-kV has been shown to be an excellent technique for studying electron percolation in Ni-network in solid oxide fuel cells [9]. In this work we study the degradation process that occurs in a LFP/C electrode by Low-kV FIB/SEM Tomography, using the low-voltage percolation technique to identify compositional changes in the CB network in three-dimensions. FIB/SEM images of a fresh and degraded cathode are compared and LFP grains are seen to crack with cycling, resulting in the formation of secondary disconnected particles with increased ionic resistivity; CB particles are instead observed to agglomerate, reducing the electrochemically active surface area. Using low voltage imaging (1 kV) a significant fraction of the large carbon agglomerates found in the aged electrode show a higher secondary electron yield compared to the fresh CB particles at low accelerating voltage. This suggests that degradation occurs both due to morphological changes and due to amorphous-crystalline phase transitions in the carbon network, resulting in non-percolating CB agglomerates. Figure 1 shows lateral Everhart – Thornley (E-T) and in-lens detector images of fresh (a, b) and degraded cathode (c, d) collected after FIB slicing. White grains are LFP, black particles are CB additive and grey regions are pores infiltrated with silicon resin to improve phase contrast [7]. The CB network appears entirely dark in the in-lens detector image of the fresh electrode (Fig. 1b). It is possible to notice some charging effects from the insulating silicon resin. The in-lens detector image of the degraded electrode (Fig. 1d) is instead characterized by the presence of big carbon agglomerates (red rings) which are brighter because they charge as the electron beam hit them. This indicates a lower electric conductivity. 3D reconstruction of the entire network (Fig. 2) revealed that the carbon phase with lower electric conductivity accounted for approximately 25% of the volume of the total carbon in the sample. This of course affects electrode capacity since a

General information
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Organisations: Department of Energy Conversion and Storage, Applied Electrochemistry, Imaging and Structural Analysis , Atomic scale modelling and materials
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Main Research Area: Technical/natural sciences

Low-voltage FIB/SEM Tomography for 3D Microstructure Evolution of LiFePO4/C Electrode
This work presents an investigation of the degradation mechanisms that occur in LiFePO4/C battery electrodes during charge/discharge cycling. Impedance spectra were measured on a fresh electrode and an electrode aged by cycling. The spectra were modeled with an equivalent circuit which indicates that both the ionic and electronic pathways in the electrode were negatively affected by the cycling. Focused Ion Beam/Scanning Electron Microscopy (FIB/SEM) tomography of both electrodes shows that cycling causes agglomerations of Carbon black (CB). In addition to this, Low-voltage FIB/SEM revealed non-conductive CB in the aged electrode.

General information
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Main Research Area: Technical/natural sciences
Structure and crystallinity of water dispersible photoactive nanoparticles for organic solar cells

Water based inks would be a strong advantage for large scale production of organic photovoltaic devices. Formation of water dispersible nanoparticles produced by the Landfester method is a promising route to achieve such inks. We provide new insights into the key ink properties of poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) nanoparticles such as the internal structure and crystallinity of the dispersed nanoparticles and the previously unreported drastic changes that occur when the inks are cast into a film. We observe through transmission electron microscopy (TEM) and small angle X-ray scattering (SAXS) that the nanoparticles in dispersion are spherical with the nanodomains of P3HT being partly crystalline. When wet processed and dried into films, the nanoparticles lose their spherical shape and become flattened into oblate shapes with a large aspect ratio. Most particles are observed to have a diameter 13 times of the particle height. After casting into a film, the crystal domains adopt a preferred orientation with the majority of the nanocrystals (68%) with face-on orientation to the substrate. We propose that low substrate surface energy is responsible for particle deformation and texturing.

General information
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Organisations: Department of Energy Conversion and Storage, Imaging and Structural Analysis, Functional organic materials, University of Copenhagen, Aalborg University, Changchun Institute of Applied Chemistry
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Main Research Area: Technical/natural sciences

Publication information
Journal: Journal of Materials Chemistry A
Volume: 3
Issue number: 33
ISSN (Print): 2050-7488
Ratings:
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
NiO/YSZ Reduction for SOFC/SOEC Studied In Situ by Environmental Transmission Electron Microscopy

SOFCs/SOECs are typically composed of ceramic materials, which are highly complex at the nano-scale. Scanning and transmission electron microscopy (SEM and TEM) are routinely applied for studying these nano-scaled structures post mortem, but only few SOFC/SOEC studies have applied environmental TEM (ETEM). ETEM offer the possibility to record image series (movies) of the ceramic nanostructures with atomic scale resolution during exposure to a reactive gas environment at elevated temperatures. The present contribution focuses on the typical reduction preparation step for the state-of-the-art Ni/YSZ (YSZ = Y2O3-stabilized ZrO2) based anodes for SOFC and cathodes for SOEC. Specifically, the reduction of nickel oxide to form the catalytically active nickel surface is monitored directly at the nano- and atomic scale by using an ETEM. The reduction process was followed while exposing NiO/YSZ and pure NiO to H2 at temperatures from room temperature to ca. 800°C. The NiO/YSZ was prepared by crushing down a tape casted and sintered model anode/cathode into a fine powder. Previous studies based on averaging techniques have shown that the reduction of pure NiO is a relatively rapid process, while the reduction of NiO/YSZ is slower, which indicates that the presence of YSZ inhibits the reduction of NiO. In the presnt in situ experiments the temperature dependent reduction profile are found similar for the both nano-scaled NiO and NiO/YSZ sample. The apparent inhibitive effect of YSZ on NiO reduction is therefore not caused by a direct interaction between NiO and YSZ, but is an indirect effect depending on the NiO being integrated in a macroscopic network of NiO/YSZ. A Titan E-Cell 80-300ST TEM was used for the in situ work in combination with the chip-based heating holder from Protochips which facilitated rapid temperature ramping for example from room temperature to 800°C in only 1 s. The ETEM results are compared to complementary averaging techniques such as thermo-gravimetric analysis (TGA) and X-ray diffraction analysis (XRD). The figure presents a TEM image series of NiO during exposure to 2 mbar H2 and constant temperature ramping rate of 1°C/min. The NiO observed in the first image at 320°C is dense. From the lower left corner a front of porous Ni is progressing until full reduction at 340°C.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Imaging and Structural Analysis, Mixed Conductors, Department of Chemistry, Center for Electron Nanoscopy
Number of pages: 1
Pages: 981
Publication date: 2014
Conference: 226th Meeting of the Electrochemical Society (ECS) and 7th Meeting of the Mexico Section of the Electrochemical Society ECS and SMEQ Joint International Meeting, Cancun, Mexico, 05/10/2014 - 05/10/2014
Main Research Area: Technical/natural sciences

Publication information
Journal: Electrochemical Society. Meeting Abstracts (Online)
Volume: MA2014-02
Issue number: 20
NiO/YSZ Reduction for SOFC/SOEC Studied In Situ by Environmental Transmission Electron Microscopy

A typical anode for solid oxide fuel cells (SOFC) or cathode for solid oxide electrolysis cells (SOEC) is a complex porous structure of Ni and yttria-stabilized zirconia (YSZ). The porous Ni/YSZ is usually prepared from powder mixtures of NiO and YSZ, tape casted and sintered into a dense structure and finally reduced during start-up of the SOFC/SOEC in H₂ at the operating temperature of the cell (ca. 800 °C). This contribution presents environmental transmission electron microscopy (ETEM) nanoscale observations of the reduction process of a NiO/YSZ powder in H₂ at temperatures up to almost 1000 °C. The study focusses on the temperature dependent dynamical morphology of the NiO/YSZ and on the possible influence of YSZ on the NiO reduction.

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Conference: 226th Meeting of the Electrochemical Society (ECS) and 7th Meeting of the Mexico Section of the Electrochemical Society ECS and SMEQ Joint International Meeting, Cancun, Mexico, 05/10/2014 - 05/10/2014
Main Research Area: Technical/natural sciences

Publication information
Journal: ECS Transactions
Volume: 64
Issue number: 2
ISSN (Print): 1938-5862
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BFI (2018): BFI-level 1
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 0.44 SJR 0.225 SNIP 0.252
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 0.4 SJR 0.228 SNIP 0.253
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 0.211 SNIP 0.244 CiteScore 0.36
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 0.212 SNIP 0.234 CiteScore 0.36
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 0.192 SNIP 0.231 CiteScore 0.27
ISI indexed (2013): ISI indexed no
BFI (2012): BFI-level 1
Scopus rating (2012): SJR 0.241 SNIP 0.26 CiteScore 0.29
ISI indexed (2012): ISI indexed no
BFI (2011): BFI-level 1
Scopus rating (2011): SJR 0.261 SNIP 0.28 CiteScore 0.36
ISI indexed (2011): ISI indexed no
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 0.249 SNIP 0.251
BFI (2009): BFI-level 1
SOFC anode reduction studied by in situ TEM

The Solid Oxide Fuel Cell (SOFC) is a promising part of future energy approaches due to a relatively high energy conversion efficiency and low environmental pollution. SOFCs are typically composed of ceramic materials which are highly complex at the nanoscale. TEM is routinely applied ex situ for studying these nanoscale structures, but only few SOFC studies have applied in situ TEM to observe the ceramic nanostructures in a reactive gas environment at elevated temperatures.

The present contribution focuses on the reduction of an SOFC anode which is a necessary process to form the catalytically active Ni surface before operating the fuel cells. The reduction process was followed in the TEM while exposing a NiO/YSZ (YSZ = Y2O3-stabilized ZrO2) model anode to H2 at T = 250-1000°C. Pure NiO was used in reference experiments. Previous studies have shown that the reduction of pure NiO is a relatively rapid autocatalytic process. On the contrary, the reduction of NiO/YSZ is significantly slower, which indicates that the presence of YSZ inhibits the reduction of NiO. This study aims to obtain fundamental insight into this reduction mechanism and to explain the inhibitive influence of YSZ.

A Titan E-Cell 80-300ST TEM was used for the in situ work in combination with the chip-based Aduro heating holder from Protochips. Since the chip-based heating holder does not allow internal temperature measurements, and since the temperature of the chips are only calibrated in vacuum, part of the presentation will focus on temperature calibration.

Alloyed Ni-Fe nanoparticles as catalysts for NH3 decomposition

A rational design approach was used to develop an alloyed Ni-Fe/Al2O3 catalyst for decomposition of ammonia. The dependence of the catalytic activity is tested as a function of the Ni-to-Fe ratio, the type of Ni-Fe alloy phase, the metal loading and the type of oxide support. In the tests with high temperatures and a low NH3-to-H2 ratio, the catalytic activity of the best Ni-Fe/Al2O3 catalyst was found to be comparable or even better than that of a more expensive Ru-based catalyst. Small Ni-Fe nanoparticle sizes are crucial for an optimal overall NH3 conversion because of a structural effect favoring the smallest particles in terms of catalytic activity per active site. Compared with SiO2, ZrO2 and TiO2, the support materials Al2O3 or Mg-Al-spinel give the highest performance in the high temperature range.

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Event: Abstract from Scandem 2013 - Annual Meeting of the Nordic Microscopy Society, Copenhagen, Denmark.
Main Research Area: Technical/natural sciences
Electronic versions: prod21375094843033.2013_Simonsen_abstract_Scandem.pdf
Source: dtu
Source-ID: u::8159
Publication: Research - peer-review › Conference abstract for conference – Annual report year: 2013

General information

State: Published
Organisations: Department of Energy Conversion and Storage, Imaging and Structural Analysis, Department of Physics, Experimental Surface and Nanomaterials Physics, Amminex Emissions Technology A/S
Authors: Simonsen, S. B. (Intern), Chakraborty, D. (Ekstern), Chorkendorff, I. (Intern), Dahl, S. (Intern)
Pages: 22-31
Publication date: 2012
Main Research Area: Technical/natural sciences
Effect of Particle Morphology on the Ripening of Supported Pt Nanoparticles

To improve the understanding of sintering in diesel and lean-burn engine exhaust after-treatment catalysts, we examined oxygen-induced sintering in a model catalyst consisting of Pt nanoparticles supported on a planar, amorphous Al2O3 substrate. After aging at increasing temperatures, a transmission electron microscopy analysis reveals that a highly monodispersed ensemble of nanoparticles transformed into an ensemble with bimodal and subsequently Lifshitz–Slyozov–Wagner particle size distribution. Moreover, scanning transmission electron microscopy and atomic force microscopy analyses suggest that the Pt nanoparticle had size-dependent morphologies after sintering in the oxidizing environment. The evolution of the particle sizes is described by a simple kinetic model for ripening, and the size-dependent particle morphology is proposed as an explanation for the observed bimodal particle size distribution shape.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Imaging and Structural Analysis, Department of Physics, Experimental Surface and Nanomaterials Physics, Chalmers University of Technology, Aarhus University, Haldor Topsoe AS
Authors: Simonsen, S. B. (Intern), Chorkendorff, I. (Intern), Dahl, S. (Intern), Skoglundh, M. (Ekstern), Meinander, K. (Forskerdatabase), Jensen, T. N. (Ekstern), Lauritsen, J. V. (Forskerdatabase), Helveg, S. (Ekstern)
Pages: 5646-5653
Publication date: 2012
Main Research Area: Technical/natural sciences

Publication information
Journal: The Journal of Physical Chemistry Part C
Volume: 116
Issue number: 9
ISSN (Print): 1932-7447
Ratings:
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 4.58 SJR 2.135 SNIP 1.147
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 4.48 SJR 1.964 SNIP 1.195
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): SJR 1.886 SNIP 1.26 CiteScore 4.68
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): SJR 2.032 SNIP 1.447 CiteScore 5.08
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): SJR 2.143 SNIP 1.445 CiteScore 5.14
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): SJR 2.529 SNIP 1.461 CiteScore 4.98
ISI indexed (2012): ISI indexed yes
Quantification of tip-broadening in non-contact atomic force microscopy with carbon nanotube tips
Carbon nanotube terminated atomic force microscopy (AFM) probes have been used for the imaging of 5 nm wide surface supported Pt nanoclusters by non-contact (dynamic mode) AFM in an ultra-high vacuum. The results are compared to AFM measurements done with conventional Si-tips, as well as with transmission electron microscopy images, which give accurate measures for cluster widths. Despite their ideal aspect ratio, tip-broadening is concluded to be a severe problem even when imaging with carbon nanotube tips, which overestimates the cluster width by several times the nominal width of the nanotube tip. This broadening is attributed to a bending of the carbon nanotubes, and not to pure geometrical factors, which coincidentally results in a significant improvement for relative height measurements of tightly spaced high aspect ratio structures, as compared to what can be achieved with geometrically limited conventional probes. Superior durability also stands out as a defining feature of carbon nanotube terminated probes, allowing them to give results with a greatly enhanced reproducibility.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Imaging and Structural Analysis, Aarhus University, Haldor Topsoe AS, Interdisciplinary Nanoscience Center
Authors: Meinander, K. (Forskerdatabase), Jensen, T. N. (Ekstern), Simonsen, S. B. (Intern), Helveg, S. (Ekstern), Lauritsen, J. V. (Forskerdatabase)
Pages: -
Publication date: 2012
Main Research Area: Technical/natural sciences

Publication information
Journal: Nanotechnology
Volume: 23
Issue number: 40
ISSN (Print): 0957-4484
Ratings:
Structure–activity relationships of Pt/Al₂O₃ catalysts for CO and NO oxidation at diesel exhaust conditions
Structure–performance relationships for Pt/Al₂O₃ catalysts with mean Pt particle sizes of 1, 2, 3, 5 and 10nm are investigated for the catalytic oxidation of CO and NO under lean-burning diesel exhaust conditions. The most active catalysts for CO oxidation exhibit Pt particles of 2–3nm, having a large fraction of low-coordinated and reactive surface Pt atoms. Exploiting in situ XAFS, we find that a reversible Pt surface oxidation is connected to high CO conversion. NO oxidation is most efficient over the catalysts with the largest Pt particles mainly exhibiting surface Pt atoms on planar facets. An irreversible Pt oxide formation observed during NO oxidation is a possible deactivation route and we suggest that the most active sites for NO oxidation are the ones least prone to surface oxidation. When both CO and NO are present in the reaction mixture, activity is increased for both reactions, suggesting that CO oxidation actively regenerates the Pt surface for NO oxidation and vice versa. The effect is strongest for the NO oxidation activity.

General information
State: Published
Organisations: Department of Physics, Experimental Surface and Nanomaterials Physics, Department of Energy Conversion and Storage, Imaging and Structural Analysis, Haldor Topsoe AS, Karlsruher Institut für Technologie, University of Copenhagen
Authors: Boubnov, A. (Ekstern), Dahl, S. (Intern), Johnson, E. (Ekstern), Molina, A. P. (Ekstern), Simonsen, S. B. (Intern), Cano, F. M. (Ekstern), Helveg, S. (Ekstern), Lemus-Yegres, L. J. (Ekstern), Grunwaldt, J. (Ekstern)
Pages: 315-325
Publication date: 2012
Main Research Area: Technical/natural sciences

Publication information
Journal: Applied Catalysis B: Environmental
Volume: 126
ISSN (Print): 0926-3373
Ratings:
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 10.92 SJR 3.152 SNIP 2.359
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 8.86 SJR 2.693 SNIP 2.185
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): SJR 2.326 SNIP 2.16 CiteScore 7.72
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): SJR 2.322 SNIP 2.206 CiteScore 6.92
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): SJR 2.391 SNIP 2.154 CiteScore 6.42
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): SJR 2.65 SNIP 2.234 CiteScore 6.08
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): SJR 2.606 SNIP 2.351 CiteScore 6.14
ISI indexed (2011): ISI indexed yes
Ostwald ripening in a Pt/SiO2 model catalyst studied by in situ TEM

Sintering of Pt nanoparticles dispersed on a planar SiO2 support was studied by in situ transmission electron microscopy (TEM). A time-lapsed TEM image series of the Pt nanoparticles, acquired during the exposure to 10mbar synthetic air at 650°C, reveal that the sintering was governed by the Ostwald ripening mechanism. The in situ TEM images also provide information about the temporal evolution of the Pt particle size distribution and of the growth or decay of the individual nanoparticles. The observed Pt nanoparticle changes compare well with predictions made by mean-field kinetic models for ripening, but deviations are revealed for the time-evolution for the individual nanoparticles. A better description of the individual nanoparticle ripening is obtained by kinetic models that include local correlations between neighboring nanoparticles in the atom-exchange process.

General information
State: Published
Organisations: Experimental Surface and Nanomaterials Physics, Department of Physics, Center for Individual Nanoparticle Functionality, Chalmers University of Technology, Haldor Topsoe AS
Authors: Simonsen, S. B. (Intern), Chorkendorff, I. (Intern), Dahl, S. (Intern), Skoglundh, M. (Ekstern), Sehested, J. (Ekstern), Helveg, S. (Ekstern)
Pages: 147-155
Publication date: 2011
Main Research Area: Technical/natural sciences

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Journal: Journal of Catalysis
Volume: 281
Issue number: 1
ISSN (Print): 0021-9517
Ratings:
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 6.99 SJR 2.397 SNIP 1.85
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 7.27 SJR 2.451 SNIP 2.142
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): SJR 2.668 SNIP 2.177 CiteScore 7.23
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): SJR 2.688 SNIP 2.233 CiteScore 6.92
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): SJR 2.553 SNIP 2.091 CiteScore 6.42
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): SJR 3.006 SNIP 2.257 CiteScore 6.17
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): SJR 3.092 SNIP 2.214 CiteScore 6.23
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 2
Scopus rating (2010): SJR 3.38 SNIP 2.193
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 2
Scopus rating (2009): SJR 2.965 SNIP 2.135
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 2
Scopus rating (2008): SJR 3.123 SNIP 2.164
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 3.15 SNIP 2.138
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 3.137 SNIP 2.043
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 2.99 SNIP 2.17
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 2.623 SNIP 2.015
Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 2.407 SNIP 1.783
Web of Science (2003): Indexed yes
Scopus rating (2002): SJR 2.317 SNIP 1.771
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 2.829 SNIP 1.873
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 2.344 SNIP 2.156
Web of Science (2000): Indexed yes
Scopus rating (1999): SJR 2.309 SNIP 1.951

Original language: English
Sintering, Pt, Ostwald ripening, Nanoparticles, In situ TEM, Catalyst deactivation
Electronic versions:
Sintering of oxide-supported Pt and Pd nanoparticles in air studied by in situ TEM

This thesis presents a fundamental study of the sintering of supported nanoparticles in relation to diesel oxidation catalysts. The sintering of supported nanoparticles is an important challenge in relation to this catalyst, as well as many other catalyst systems, and a fundamental understanding of the sintering mechanisms of nanoparticles is important for making improvements to their long term catalytic activity.

Diesel oxidation catalysts are usually composed of noble metal nanoparticles on a complex three-dimensional high surface area oxide. The complex support structure makes it difficult to directly observe dynamical processes such as particle sintering with the present state of the art microscope techniques, and consequently it is difficult to relate experimental observations and theoretical sintering models. To reduce the complexity, the present study uses planar model catalysts. These are composed of Pt, Pd and bimetallic Pt-Pd nanoparticles supported on a flat and homogeneous Al2O3 or SiO2 surface. By using in situ TEM on the planar model catalysts it was possible to directly monitor the detailed dynamical changes of the individual nanoparticles during exposure to oxidizing conditions at elevated temperatures. The time-resolved TEM images are presented and these offer direct insight into the fundamental dynamics of the sintering process at the nano-scale.

For Pt, Pd and bimetallic Pt-Pd nanoparticles it is shown that the sintering process is governed by the Ostwald ripening mechanism in an oxidizing environment. The observations compare well with predictions from mean-field kinetic models for ripening, but deviations are revealed for the time-evolution for the individual nanoparticles. A better description of the individual nanoparticle ripening is obtained by kinetic models that include local correlations between neighbouring nanoparticles in the atom-exchange process. The sintering process was also presented statistically by particle size distributions extracted from the TEM images. The statistical data agreed only partly with the mean-field kinetic models for ripening, but the deviations could be accounted for by including more detailed information into the models, such as an observed size-dependence of the three-dimensional shape of the supported nanoparticles and the local correlations between the nanoparticles.

General information
State: Published
Organisations: Center for Individual Nanoparticle Functionality, Department of Physics, Experimental Surface and Nanomaterials Physics, Haldor Topsoe AS
Authors: Simonsen, S. B. (Intern), Chorkendorff, I. (Intern), Helveg, S. (Ekstern), Dahl, S. (Intern)
Number of pages: 162
Publication date: 2011

Catalytic Soot Oxidation studied by ETEM

General information
State: Published
Organisations: Haldor Topsoe AS, University of Copenhagen
Authors: Simonsen, S. B. (Intern), Dahl, S. (Ekstern), Johnson, E. (Ekstern), Molenbroek, A. M. (Ekstern), Helveg, S. (Ekstern)
Publication date: 2010
Event: Abstract from NACSC, San Francisco, CA, USA, 7-12 June, .
Direct Observations of Oxygen-induced Platinum Nanoparticle Ripening Studied by In Situ TEM

This study addresses the sintering mechanism of Pt nanoparticles dispersed on a planar, amorphous Al2O3 support as a model system for a catalyst for automotive exhaust abatement. By means of in situ transmission electron microscopy (TEM), the model catalyst was monitored during the exposure to 10 mbar air at 650 degrees C. Time-resolved image series unequivocally reveal that the sintering of Pt nanoparticles was mediated by an Ostwald ripening process. A statistical analysis of an ensemble of Pt nanoparticles shows that the particle size distributions change shape from an initial Gaussian distribution via a log-normal distribution to a Lifshitz-Slyozov-Wagner (LSW) distribution. Furthermore, the time-dependency of the ensemble-averaged particle size and particle density is determined. A mean field kinetic description captures the main trends in the observed behavior. However, at the individual nanoparticle level, deviations from the model are observed suggesting in part that the local environment influences the atom exchange process.

General information

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Organisations: Experimental Surface and Nanomaterials Physics, Department of Physics
Authors: Simonsen, S. B. (Intern), Chorkendorff, I. (Intern), Dahl, S. (Intern), Skoglundh, M. (Ekstern), Sehested, J. (Ekstern), Helveg, S. (Ekstern)
Pages: 7968-7975
Publication date: 2010
Main Research Area: Technical/natural sciences

Publication information
Journal: Journal of the American Chemical Society
Volume: 132
Issue number: 23
ISSN (Print): 0002-7863
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BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 14.05 SJR 8.127 SNIP 2.641
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 13.18 SJR 7.492 SNIP 2.596
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): SJR 6.775 SNIP 2.63 CiteScore 12.81
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): SJR 6.294 SNIP 2.587 CiteScore 11.92
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): SJR 5.993 SNIP 2.466 CiteScore 11.38
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): SJR 6.211 SNIP 2.38 CiteScore 10.37
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): SJR 5.478 SNIP 2.321 CiteScore 9.94
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
In situ TEM studies of catalysts for diesel engine exhaust abatement

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Imaging and Structural Analysis, Department of Physics, Experimental Surface and Nanomaterials Physics, Haldor Topsoe AS, Chalmers University of Technology, University of Copenhagen
Authors: Simonsen, S. B. (Intern), Dahl, S. (Ekstern), Chorkendorff, I. (Intern), Johnson, E. (Ekstern), Sehested, J. (Ekstern), Skoglundh, M. (Ekstern), Molenbroek, A. M. (Ekstern), Helveg, S. (Ekstern)
Number of pages: 2
Publication date: 2010
Event: Abstract from 17th International Microscopy Congress, Rio De Janeiro, Brazil.
Main Research Area: Technical/natural sciences
Electronic versions:
2011-Simonsen_imc17_abstract.pdf
Source: dtu
Source-ID: u::8160
Publication: Research - peer-review › Conference abstract for conference – Annual report year: 2010

Size-selective aerobe oxidation by sinter stable gold nanoparticles embedded in zeolite crystals

General information
State: Published
Size-selective Oxidation with Au nanoparticles embedded in Zeolite Crystals

General information
State: Published
Organisations: Department of Physics, Department of Energy Conversion and Storage, Imaging and Structural Analysis, Department of Chemistry, Centre for Catalysis and Sustainable Chemistry
Publication date: 2010
Event: Abstract from 14th Nordic Symposium on Catalysis, Helsingør, Denmark.
Main Research Area: Technical/natural sciences
Publication: Research - peer-review › Conference abstract for conference – Annual report year: 2010

Substrate Size-Selective Catalysis with Zeolite-Encapsulated Gold Nanoparticles

The Dark Crystal: A hybrid material is reported that is comprised of 1-2 nm sized gold nanoparticles, accessible only through zeolite micropores in a silicalite-1 crystal, as shown by three-dimensional TEM tomography (see picture). Calcination experiments indicate that the embedded nanoparticles are highly stable towards sintering.

General information
State: Published
Organisations: Experimental Surface and Nanomaterials Physics, Department of Physics, Sustainable and Green Chemistry, Department of Chemistry, CHEC Research Centre, Department of Chemical and Biochemical Engineering, Administration
Pages: 3504-3507
Publication date: 2010
Main Research Area: Technical/natural sciences
Publication information
Volume: 49
Issue number: 20
ISSN (Print): 1433-7851
Ratings:
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 11.31 SJR 6.155 SNIP 2.165
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 10.8 SJR 5.954 SNIP 2.146
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): SJR 5.888 SNIP 2.225 CiteScore 11.13
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): SJR 5.811 SNIP 2.307 CiteScore 10.84
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): SJR 5.702 SNIP 2.198 CiteScore 10.7
Catalytic Soot Oxidation studied by ETEM

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Imaging and Structural Analysis, Department of Physics, Experimental Surface and Nanomaterials Physics, Haldor Topsoe AS, University of Copenhagen
Authors: Simonsen, S. B. (Intern), Dahl, S. (Ekstern), Johnson, E. (Ekstern), Molenbroeck, A. M. (Ekstern), Chorkendorff, I. (Intern), Helveg, S. (Ekstern)
Publication date: 2009
Event: Abstract from 9th European Congress on Catalysis, Salamanca, Spain.
Main Research Area: Technical/natural sciences
Electronic versions:
prod21375095071849.2009_Simonsen_Extended_Europacat.pdf
Source: dtu
Source-ID: u::8162
Direct observations of CeO$_2$-catalyzed soot oxidation at the nano-scale using environmental transmission electron microscopy

Environmental transmission electron microscopy (ETEM) was used to study in situ the kinetics and dynamics of CeO$_2$-catalyzed soot oxidation at the nano-scale. From time-lapsed ETEM image series, direct insight was obtained into structural changes of soot particles in contact with CeO$_2$ during exposure to oxidizing conditions. Based on the insight, it was shown that catalytic soot oxidation was governed by reactions at the CeO$_2$-soot interface and that this finding was consistent with previous findings. © 2008 SAE International.

General information
State: Published
Organisations: Haldor Topsoe AS, University of Copenhagen
Authors: Simonsen, S. B. (Intern), Dahl, S. (Ekstern), Helveg, S. (Ekstern), Johnson, E. (Ekstern)
Pages: 199-203
Publication date: 2009
Main Research Area: Technical/natural sciences

Publication information
Journal: SAE International Journal of Materials and Manufacturing
Volume: 1
Issue number: 1
ISSN (Print): 1946-3979
Ratings:
Web of Science (2018): Indexed yes
Scopus rating (2017): SNIP 0.853 SJR 0.413 CiteScore 0.79
Web of Science (2017): Indexed yes
Scopus rating (2016): SJR 0.286 SNIP 0.636 CiteScore 0.58
Scopus rating (2015): SJR 0.288 SNIP 0.749 CiteScore 0.59
Scopus rating (2014): SJR 0.359 SNIP 0.781 CiteScore 0.56
Scopus rating (2013): SJR 0.35 SNIP 1.054 CiteScore 0.65
ISI indexed (2013): ISI indexed no
Scopus rating (2012): SJR 0.294 SNIP 0.649 CiteScore 0.34
ISI indexed (2012): ISI indexed no
Scopus rating (2011): SJR 0.237 SNIP 0.558 CiteScore 0.25
ISI indexed (2011): ISI indexed no
Scopus rating (2010): SJR 0.199 SNIP 0.215
Scopus rating (2005): SJR 0.122 SNIP 0
Scopus rating (2004): SJR 0.121 SNIP 0
Scopus rating (2003): SJR 0.121 SNIP 0
Original language: English
Electronic versions:
Direct_observations.pdf
DOIs:
10.4271/2008-01-0418

Bibliographical note
Technical paper presented at SAE World Congress & Exhibition, April 2008
Source: orbit
Source-ID: 314509
Publication: Research - peer-review › Journal article – Annual report year: 2008

Catalytic soot oxidation studied by Environmental Transmission Electron Microscopy

General information
State: Published
Organisations: Technical University of Denmark, Haldor Topsoe AS, University of Copenhagen
Authors: Simonsen, S. B. (Intern), Dahl, S. (Ekstern), Johnson, E. (Ekstern), Helveg, S. (Ekstern)
Publication date: 2008
Event: Abstract from 14th European Microscopy Congress, Aachen, Germany.
Ceria-catalyzed soot oxidation studied by environmental transmission electron microscopy

Environmental transmission electron microscopy (ETEM) was used to monitor in situ ceria-catalyzed oxidation of soot in relation to diesel engine emission control. From time-lapsed ETEM image series of soot particles in contact with CeO2, or with Al2O3 as inert reference, mechanistic and kinetic insight into the catalytic and noncatalytic oxidation mechanisms was obtained. Specifically, the results indicate that the catalytic soot oxidation mechanism involves reaction centers at the soot–CeO2 interface and that the interface reaction kinetic properties are in good agreement with previous macroscopic, averaging measurements.

Keyword: Ceria-based catalysts, Soot oxidation, Diesel exhausts emission control, Environmental transmission electron microscopy

General information
State: Published
Organisations: Haldor Topsoe AS, University of Copenhagen
Authors: Simonsen, S. B. (Intern), Dahl, S. (Ekstern), Johnson, E. (Ekstern), Helveg, S. (Ekstern)
Pages: 1-5
Publication date: 2008
Main Research Area: Technical/natural sciences

Publication information
Journal: Journal of Catalysis
Volume: 255
Issue number: 1
ISSN (Print): 0021-9517
Ratings:
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 6.99 SJR 2.397 SNIP 1.85
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 7.27 SJR 2.451 SNIP 2.142
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): SJR 2.668 SNIP 2.177 CiteScore 7.23
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): SJR 2.688 SNIP 2.233 CiteScore 6.92
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): SJR 2.553 SNIP 2.091 CiteScore 6.42
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): SJR 3.006 SNIP 2.257 CiteScore 6.17
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): SJR 3.092 SNIP 2.214 CiteScore 6.23
ISI indexed (2011): ISI indexed yes
Direct observations of catalytic soot oxidation on a nano-scale

General information
State: Published
Organisations: Haldor Topsoe AS, Technical University of Denmark
Authors: Simonsen, S. B. (Intern), Dahl, S. (Ekstern), Helveg, S. (Ekstern)
Publication date: 2008
Main Research Area: Technical/natural sciences
Electronic versions:
2008-Abstract_SAE.doc
Source: dtu
Source-ID: u::8164
Publication: Research - peer-review › Conference abstract for conference – Annual report year: 2008

In situ studies of catalytic soot oxidation by means of Environmental Transmission Electron Microscopy (ETEM)

General information
State: Published
Organisations: Technical University of Denmark, Haldor Topsoe AS
Authors: Simonsen, S. B. (Intern), Dahl, S. (Ekstern), Johnson, E. (Ekstern), Helveg, S. (Ekstern)
In situ studies of CeO2-catalyzed soot oxidation by means of Environmental Transmission Electron Microscopy

General information
State: Published
Organisations: University of Copenhagen
Authors: Simonsen, S. B. (Intern)
Number of pages: 98
Publication date: 2008

Publication information
Publisher: University of Copenhagen
Original language: English
Main Research Area: Technical/natural sciences
Electronic versions:
Simonsen_Master_thesis.pdf

Bibliographical note
This thesis is submitted to the Faculty of Science, University of Copenhagen, Denmark, in order to fulfill the requirements for obtaining the Master degree in physics. The experimental work presented was carried out in the Characterization department and in cooperation with the Topsøe Automotive project at Haldor Topsøe A/S, Lyngby, Denmark from January 2007 to December 2007

Projects:

3D electron microscopy of nanostructures in energy devices
Department of Energy Conversion and Storage
Period: 01/08/2017 → 31/07/2020
Number of participants: 4
Phd Student:
Colding-Jørgensen, Sofie (Intern)
Supervisor:
Schmidt, Søren (Intern)
Simonsen, Søren Bredmose (Intern)
Main Supervisor:
Kuhn, Luise Theil (Intern)

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

3D imaging center
Department of Physics
Neutrons and X-rays for Materials Physics
Department of Applied Mathematics and Computer Science
Image Analysis & Computer Graphics
Department of Energy Conversion and Storage
Solid oxide fuel/electrolysis cells (SOFC/SOEC) will play an important role in future efficient and environmentally friendly energy systems if a better long term performance of the cells can be achieved. In a solid oxide fuel cell (SOFC) a fuel such as hydrogen, methane, synthesis gas, etc. is oxidized over a solid oxygen ion-conducting electrolyte which physically separates fuel and air.

The electron transfer in this chemical reaction takes place via an external circuit and electricity is therefore a product of the reaction. Catalysts are used on both sides of the ion conducting electrolyte to activate the splitting of fuel and oxygen molecules.

Impressive performance improvements have recently been reported for cells with specific multiphase nano-structures, but structural and compositional nano-scaled changes can also lead to deactivation. On the other hand, deactivation processes due to nano-scaled structural and compositional changes (segregations followed by nano-particle precipitation in some cases) can be observed near the material interfaces in the electrodes. Neither the mechanisms of the initial fast electrode process nor the development and dynamics of these critical structures are understood. Further studies of these dynamical changes are hampered since there are no available methods which offer in situ characterization of operating cells with nano-scale spatial resolution.
The project will develop a method for in situ transmission electron microscopy (TEM) of operating solid oxide electrochemical cells. Until now, knowledge of the nanoscale SOFC/SOEC dynamics is entirely based on observations performed post mortem, when the cells have been cooled down and de-mounted. To obtain direct insight into the dynamics of the active nanostructures of a SOFC/SOEC in operation can therefore have significant importance for our understanding of the cell dynamics during activation and deactivation.

The aim is to record TEM image sequences (movies) with atomic resolution of the active nanostructures in the SOFC/SOEC during operation. To do this, model SOFC/SOEC systems will be developed and integrated into a TEM holder so that heating, electrical currents and exposure to reactive gas environments all are integrated at the same time. In addition the model systems will need to be highly compact and the thickness of the imaged area should be approximately 100 nm. The project will therefore push the limits of in situ TEM experiments.

The goal is direct insight into the nano-scale dynamics of the operating SOFC/SOEC during exposure to elevated temperatures, electrical currents and a reactive gas environment. This will be a key factor in developing more efficient and stable SOFCs/SOECs. Therefore, micro-SOFC will be developed and characterized by thin layer, especially regarding the electrolyte layer which will have a thickness lower than one micrometre. This will allow to work at temperature lower than 600°C.

The project will be carried out as a PhD project in collaboration between the two applicant institutions (DTU Energy Conversion and DTU Cen) and with the in situ group from Nagoya University with Prof. Shunsuke Muto. The work will primarily be carried out by the PhD student with support and supervision from the project team. The principal supervisor is Head of Section Luise T. Kuhn. The TEM work will be carried out under the guidance of Søren Bredmose Simonsen and Prof. Jakob B. Wagner. Prof. Mogens B. Mogensen will contribute to the project as an internal consultant when analyzing and linking the observed physical properties to the electrochemical performance.

Department of Energy Conversion and Storage

Imaging and Structural Analysis

Center for Electron Nanoscopy

DTU Danchip
Period: 01/10/2014 → 30/09/2017
Number of participants: 4
In-situ TEM, fuel cell, electrolysis, electrochemistry, Nanotechnology
Acronym: TEMOC
Number of related Ph.D. students: 1
Project participant:
Kuhn, Luise Theil (Intern)
Simonsen, Søren Bredmose (Intern)
Wagner, Jakob Birkedal (Intern)
Gualandris, Fabrizio (Intern)

Project

In-situ transmission electron microscopy on operating electrochemical cells

Department of Energy Conversion and Storage
Period: 01/10/2014 → 24/01/2018
Number of participants: 7
Phd Student:
Gualandris, Fabrizio (Intern)
Supervisor:
Simonsen, Søren Bredmose (Intern)
Wagner, Jakob Birkedal (Intern)
Main Supervisor:
Kuhn, Luise Theil (Intern)
Examiner:
Bowen, Jacob R. (Intern)
Sharma, Renu (Ekstern)
Wallenberg, Reine (Ekstern)

Financing sources
Source: Internal funding (public)
Name of research programme: Forskningsrådsfinansiering
Green Production of Nanomaterials for Energy Conversion

Department of Energy Conversion and Storage
Period: 01/07/2014 → 30/09/2017
Number of participants: 8
Phd Student:
Xu, Yu (Intern)
Supervisor:
Hendriksen, Peter Vang (Intern)
Norby, Poul (Intern)
Simonsen, Søren Bredmose (Intern)
Main Supervisor:
Kiebach, Wolff-Ragnar (Intern)
Examiner:
Della Negra, Michela (Intern)
Aymonier, Cyril (Ekstern)
Lester, Edward Henry (Ekstern)

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

Relations
Publications:
Continuous Hydrothermal Flow Synthesis of Functional Oxide Nanomaterials Used in Energy Conversion Devices

Ædelmetalkatalysatore til rensning af røg fra dieselmotorer

Department of Physics
Period: 01/02/2008 → 28/09/2011
Number of participants: 7
Phd Student:
Simonsen, Søren Bredmose (Intern)
Supervisor:
Dahl, Søren (Intern)
Helveg, Stig (Ekstern)
Main Supervisor:
Chorkendorff, Ib (Intern)
Examiner:
Hansen, Thomas Willum (Intern)
Crozier, Peter A. (Ekstern)
Olsson, Eva (Ekstern)

Financing sources
Source: Internal funding (public)
Name of research programme: ErhvervsPhD-ordningen VTU

Activities:

In situ TEM study of the coarsening of carbon black supported Pt nanoparticles in hydrogen
Period: 1 Oct 2017 → 5 Oct 2017
Søren Bredmose Simonsen (Speaker)
Department of Energy Conversion and Storage
Imaging and Structural Analysis

Description
The control of sizes and shapes of nanostructures is of tremendous importance for the catalytic activity in electrochemistry and in catalysis more generally. However, due to relatively large surface free energies, nanostructures often sinter to form coarser and more stable structures that may not have the intended physicochemical properties.

Pt is known to be a very active catalyst in several chemical reactions and for example as carbon supported nanoparticles in fuel cells.

The presentation focusses on coarsening mechanisms of Pt nanoparticles supported on carbon black during exposure to hydrogen. By means of in situ transmission electron microscopy (TEM), Pt nanoparticle coarsening was monitored in 6 mbar 20 % H2/Ar while ramping up the temperature to ca. 900 °C. Time-resolved TEM images directly reveal that separated ca. 3 nm sized Pt nanoparticles in the pure hydrogen environment are stable during constant temperature ramping by 10°C/min up to ca. 800 °C. The coarsening above this temperature is fully dominated by the particle migration and coalescence mechanism. This is contrary to supported Pt nanoparticles in oxygen, where the coarsening is fully dominated by Ostwald ripening. For agglomerated Pt nanoparticles, coalescence events were observed already at ca. 200 °C. The temperature-dependency of particle sizes and the observed migration distances are consistent with simple early models for the migration and coalescence.

Degree of recognition: International

Related event

232nd ECS meeting
01/10/2017 → 05/10/2017
National Harbor, Washington, DC, United States
Activity: Talks and presentations › Conference presentations