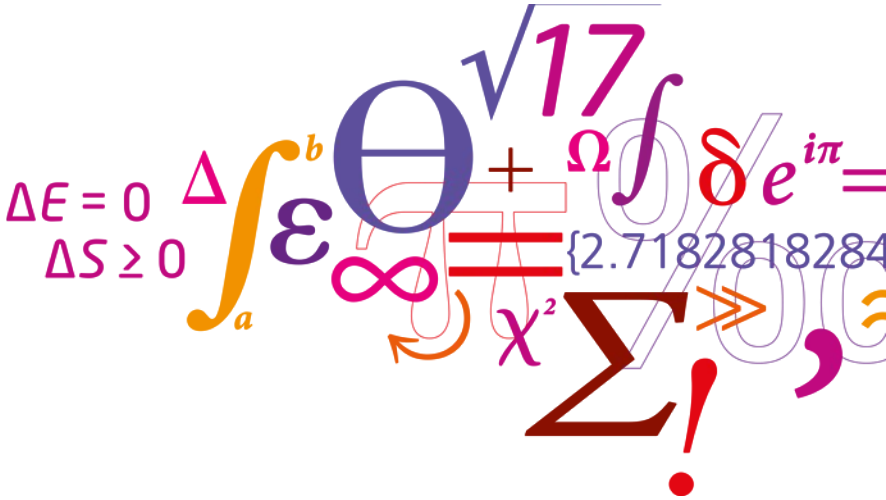
Battery Research at DTU Energy - from atomic scale to battery ~~packs~~ ~~modules~~ cells

Johan Hjelm [johh@dtu.dk], Poul Norby, Didier Blanchard,
Juan Maria Garcia Lastra, Tejs Vegge



DTU Energy
Department of Energy Conversion and Storage

Some history...

DTU Energy
Department of Energy Conversion and Storage

Created 1 January 2012, bringing together research groups from

- Risø DTU National Laboratory for Sustainable Energy (prior to 2007: Risø National Laboratory)
- DTU Chemistry

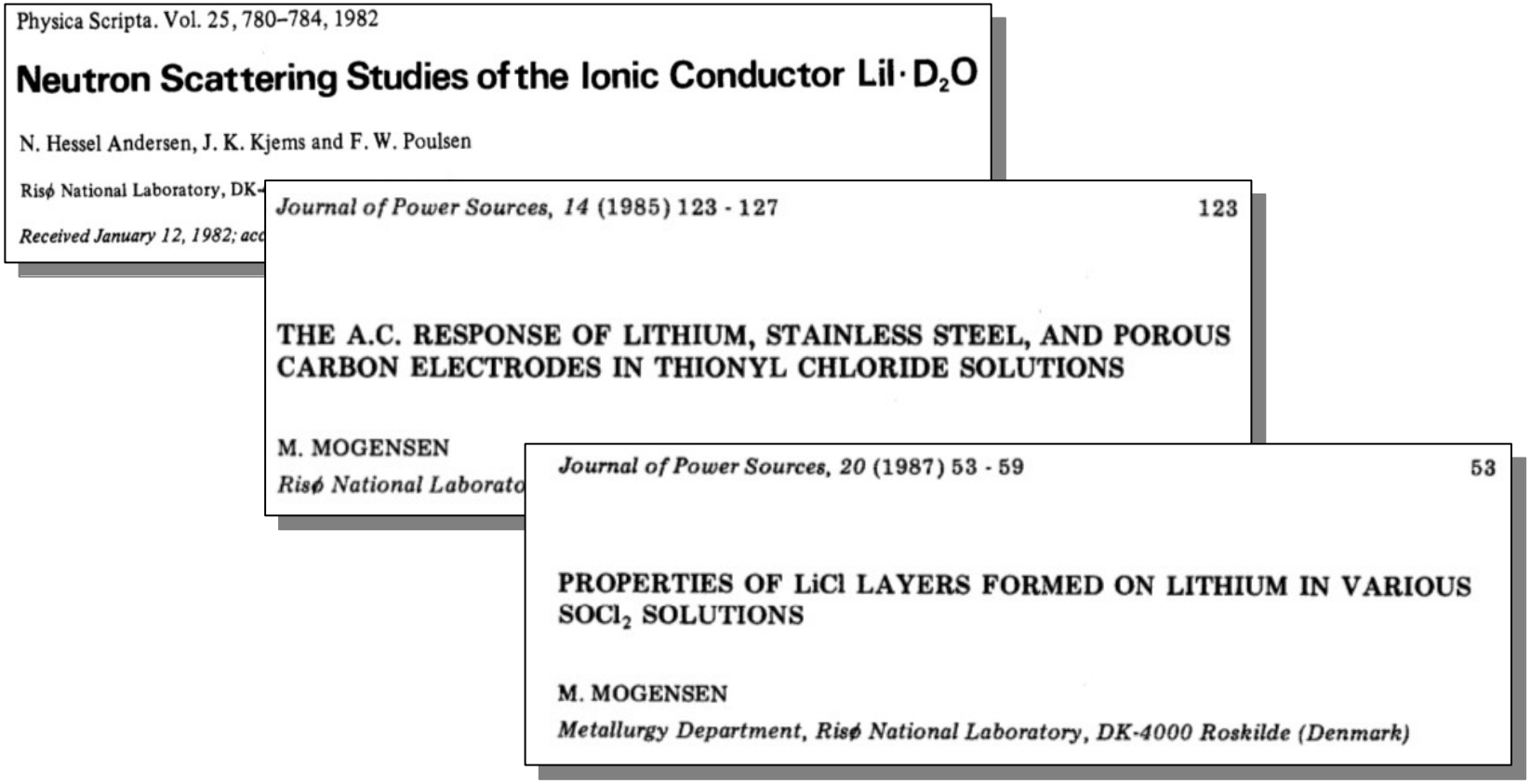
- More than 200 people
- Research spans from fundamental investigations to component manufacture
- Focus on industrial collaboration and industrially relevant processes
- Sustainable technologies for energy conversion and storage

Risø Campus (Roskilde)

Lyngby Campus (Copenhagen)

Early Battery Related Research

1980s - Li-ion conductors and Li-$SOCl_2$ research at Risø National Laboratory



Physica Scripta. Vol. 25, 780–784, 1982

Neutron Scattering Studies of the Ionic Conductor LiI·D_2O

N. Hessel Andersen, J. K. Kjems and F. W. Poulsen

Risø National Laboratory, DK-

Received January 12, 1982; acc

Journal of Power Sources, 14 (1985) 123 - 127 123

THE A.C. RESPONSE OF LITHIUM, STAINLESS STEEL, AND POROUS CARBON ELECTRODES IN THIONYL CHLORIDE SOLUTIONS

M. MOGENSEN

Risø National Laborato

Journal of Power Sources, 20 (1987) 53 - 59 53

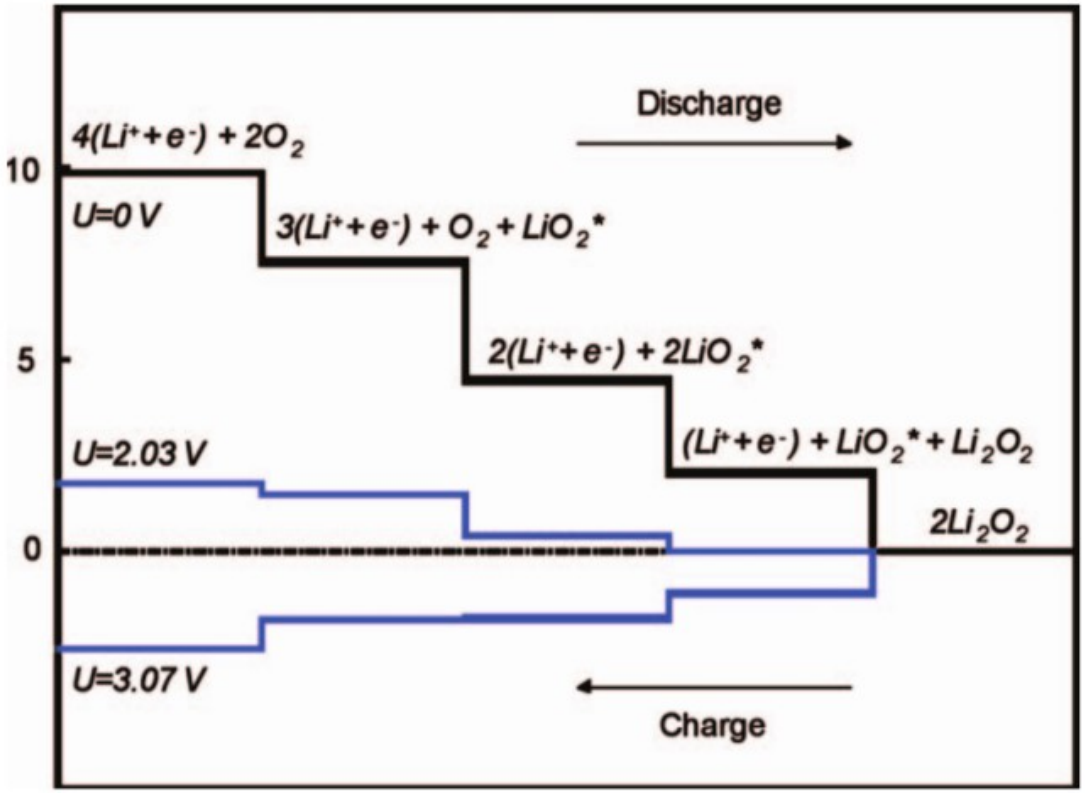
PROPERTIES OF LiCl LAYERS FORMED ON LITHIUM IN VARIOUS $SOCl_2$ SOLUTIONS

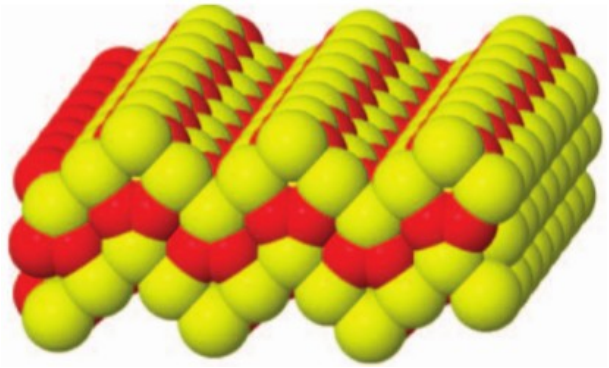
M. MOGENSEN

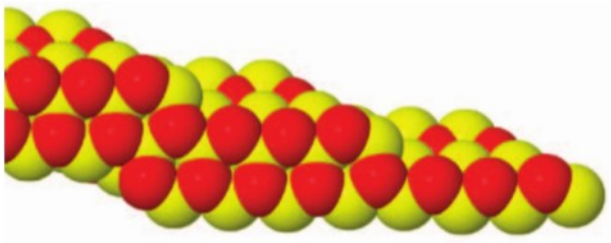
Metallurgy Department, Risø National Laboratory, DK-4000 Roskilde (Denmark)

Battery Research 2.0 - Reboot in 2010

Theoretical work on fundamental overpotentials and properties of Li_2O_2 ($Li\text{-}O_2$ cells)







"Elementary Oxygen Electrode Reactions in the Aprotic Li-Air Battery."
Hummelshøj, J. S.; Blomqvist, J.; Datta, S.; Vegge, T.; Rossmeisl, J.;
Thygesen, K. S.; Luntz, A. C.; Jacobsen, K. W.; Nørskov, J. K.
The Journal of Chemical Physics 2010, 132 (7), 071101. (Communications)

Batteries at DTU Energy: Staff & students

Ane Sælland Christiansen
Yedilfana S. Mekkonen
Mie Møller Storm
Andreas Elkjær Christensen
Kristian Bastholm Knudsen
Simon Loftager
Rolf E.R. Møller-Nielsen*
Steen Lysgaard
Vladimir Tripkovic
Roberto Scipioni
Supti Das
Nicolai Mathiesen
Jessica Lefevre*
Mathias K. Christensen

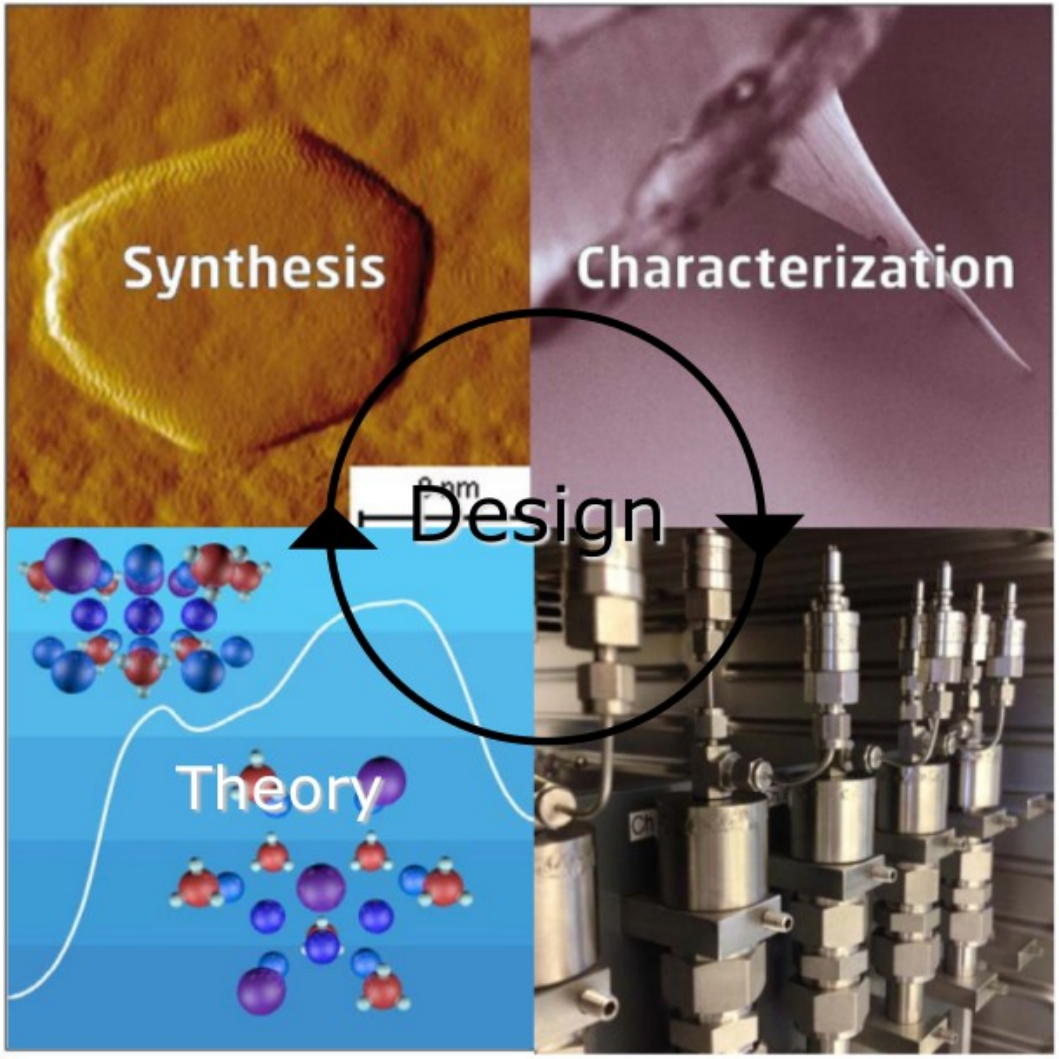
Peter Bjerre Jensen
Marko Melander
Juan Maria Garcia Lastra
Didier Blanchard
Johan Hjelm
Eugen Stamate
Søren Højgaard Jensen
Poul Norby
Tejs Vegge
Peter Holtappels
Allan Schrøder Pedersen
Irina Petrushina
Don Siegel (Uni. Michigan)
Steen Schougaard (U. Quebec)

> 25 staff & students involved (part/full) time

Main Activities

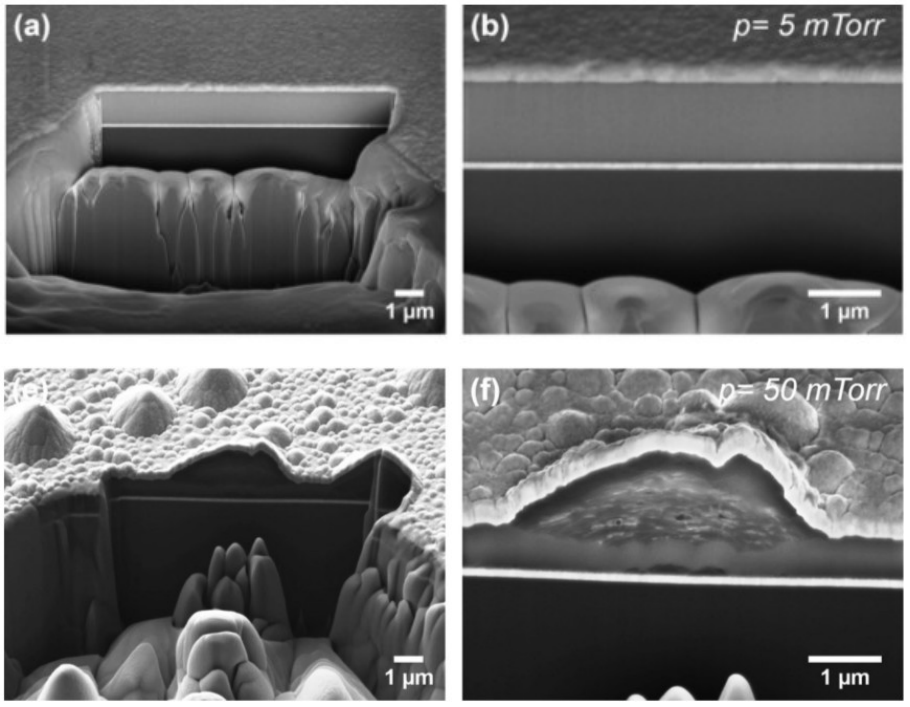
- Metal-air batteries (ReLiable, **ZAS-H2020**, Mat4Bat-YIP)
- Li-ion electrode materials (CCC, **Mat4Bat**-YIP, Nordeafonden)
- **Solid electrolytes** (Villum- and Nordeafonden, COST)
- Battery interfaces (FP7 **Hi-C**)
- Stability and lifetime predictions of batteries (**ALPBES**)
- Applications and integration
- Na-ion batteries (internal/industrial collaboration)
- Nano-confined batteries (internal, COST)
- Organic redox flow batteries (internal/industrial collaboration)

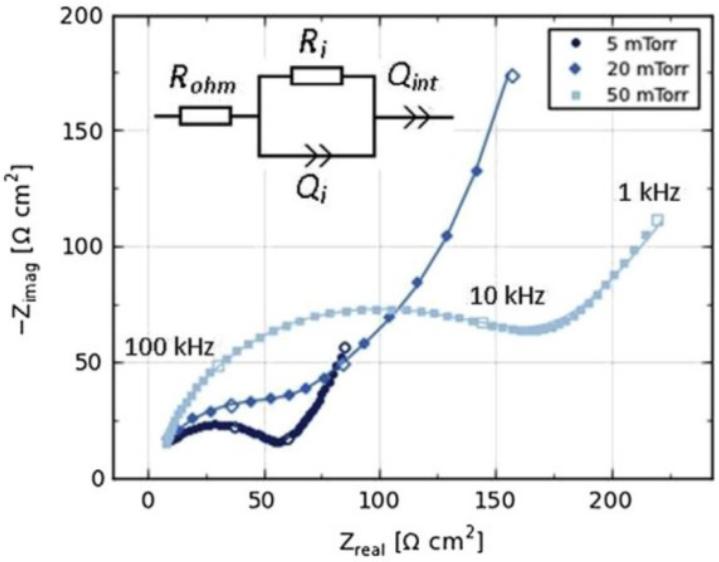
Integrated Design Loop



Solid Li-ion Electrolytes I. - [LiPON]

- LiPON (Li phosphorous oxynitride) produced by reactive RF sputtering
- Investigation of the influence of the plasma properties on the resulting electrolyte layers

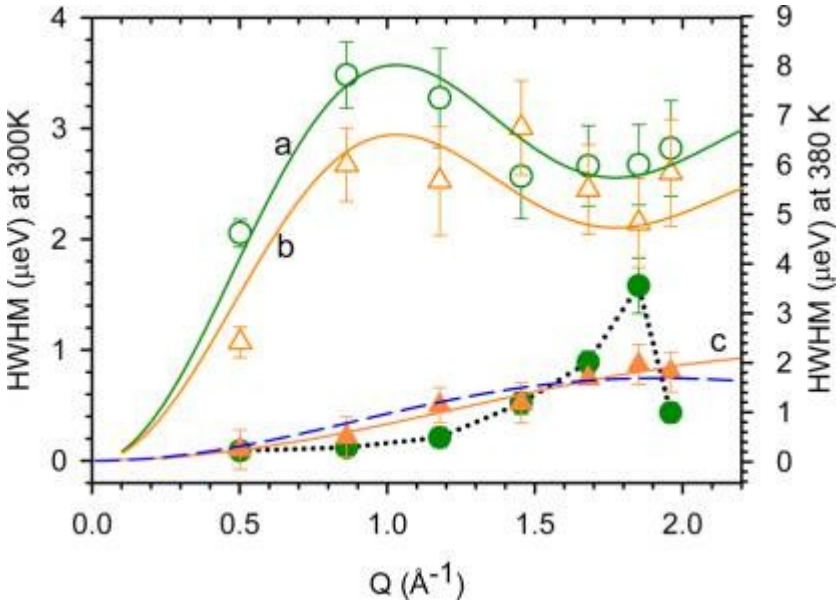


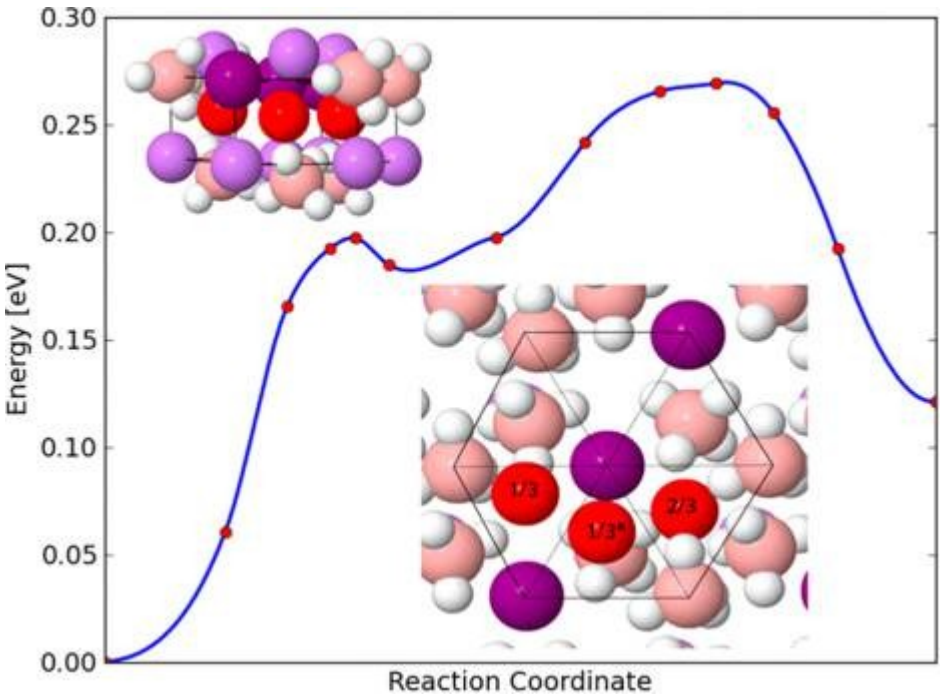


"Plasma Properties during Magnetron Sputtering of Lithium Phosphorous Oxynitride Thin Films."
Christiansen, A. S.; Stamate, E.; Thydén, K.; Younesi, R.; Holtappels, P. Journal of Power Sources 2015, 273, 863–872.

Solid Electrolytes II - [$LiBH_4$-LiI solid solution].

- Quasi-elastic neutron scattering for Li ion diffusion measurements
- DFT - Frenkel pair formation easy (Li^+ in interstitial sites)
- QENS - jump distance & frequency -> 2D diffusion in hexagonal plane

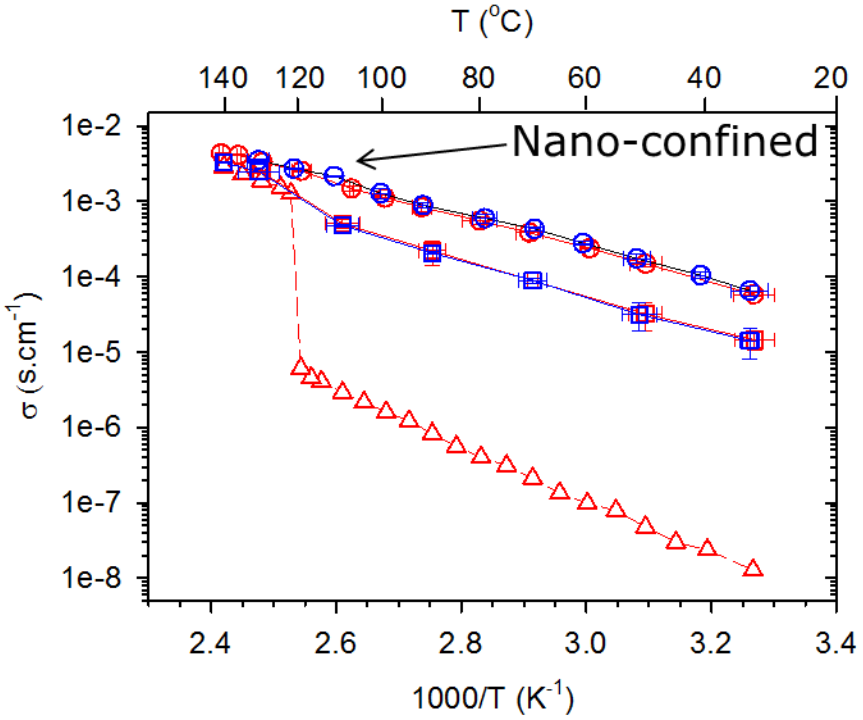


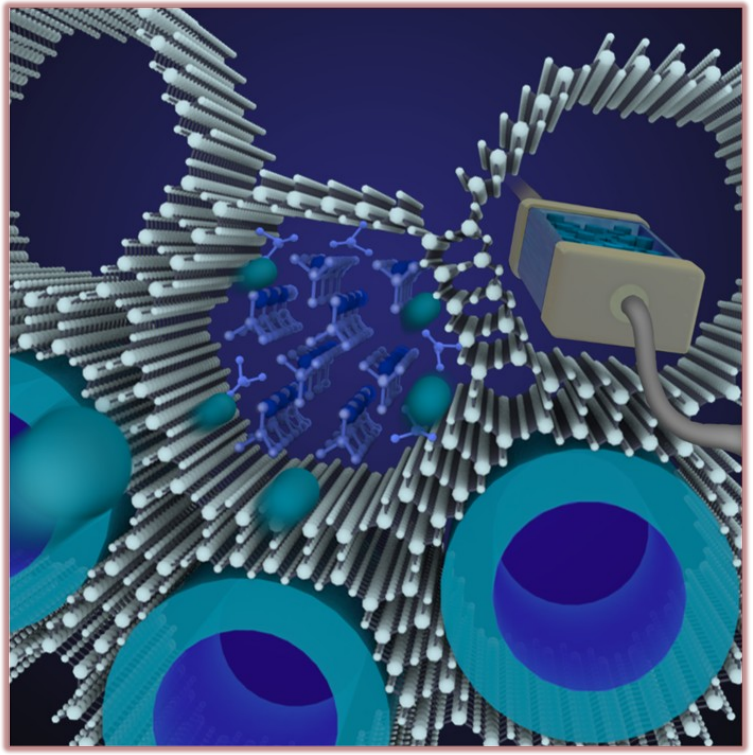


"Li-Ion Conduction in the $LiBH_4$:LiI System from Density Functional Theory Calculations and Quasi-Elastic Neutron Scattering." Myrdal, J. S. G.; Blanchard, D.; Sveinbjörnsson, D.; Vegge, T.; J. Phys. Chem. C 2013, 117 (18), 9084–9091.

Solid Electrolytes III. - [$LiBH_4$]

- Nano-confining $LiBH_4$ yields high conductivity and improved stability
- Nano-confinement in MCM-41 ordered mesoporous silica scaffolds

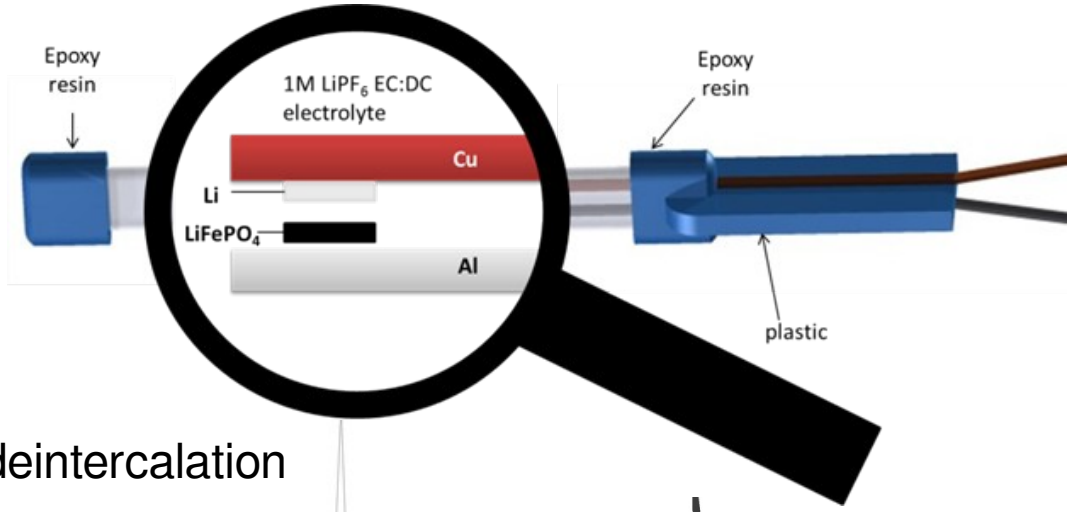


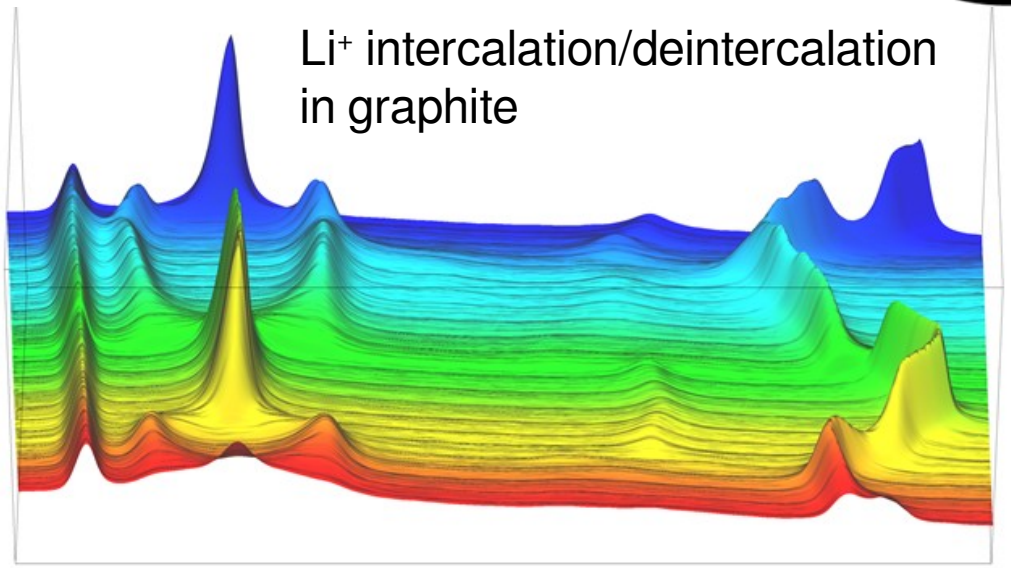


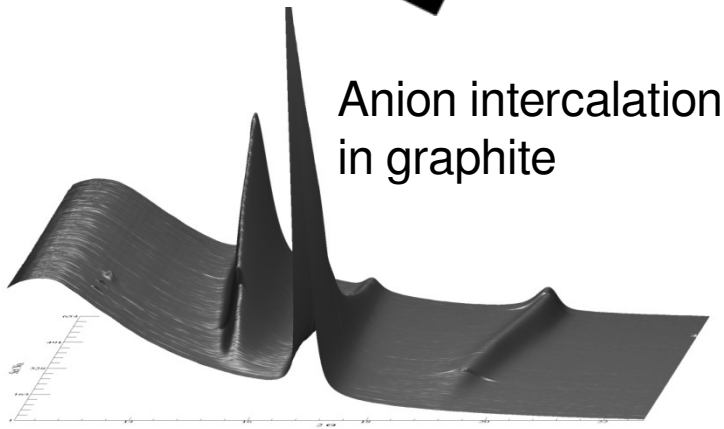
"Nanoconfined LiBH4 as a Fast Lithium Ion Conductor". Adv. Funct. Mater. 2015, 25 (2), 184–192. Blanchard, D.; Nale, A.; Sveinbjörnsson, D.; Eggenhuisen, T. M.; Verkuijlen, M. H. W.; Suwarno; Vegge, T.; Kentgens, A. P. M.; de Jongh, P. E.

In-situ synchrotron X-ray Diffraction I.

- Capillary Microbattery Cell allows in-situ/operando XRD studies of electrode materials

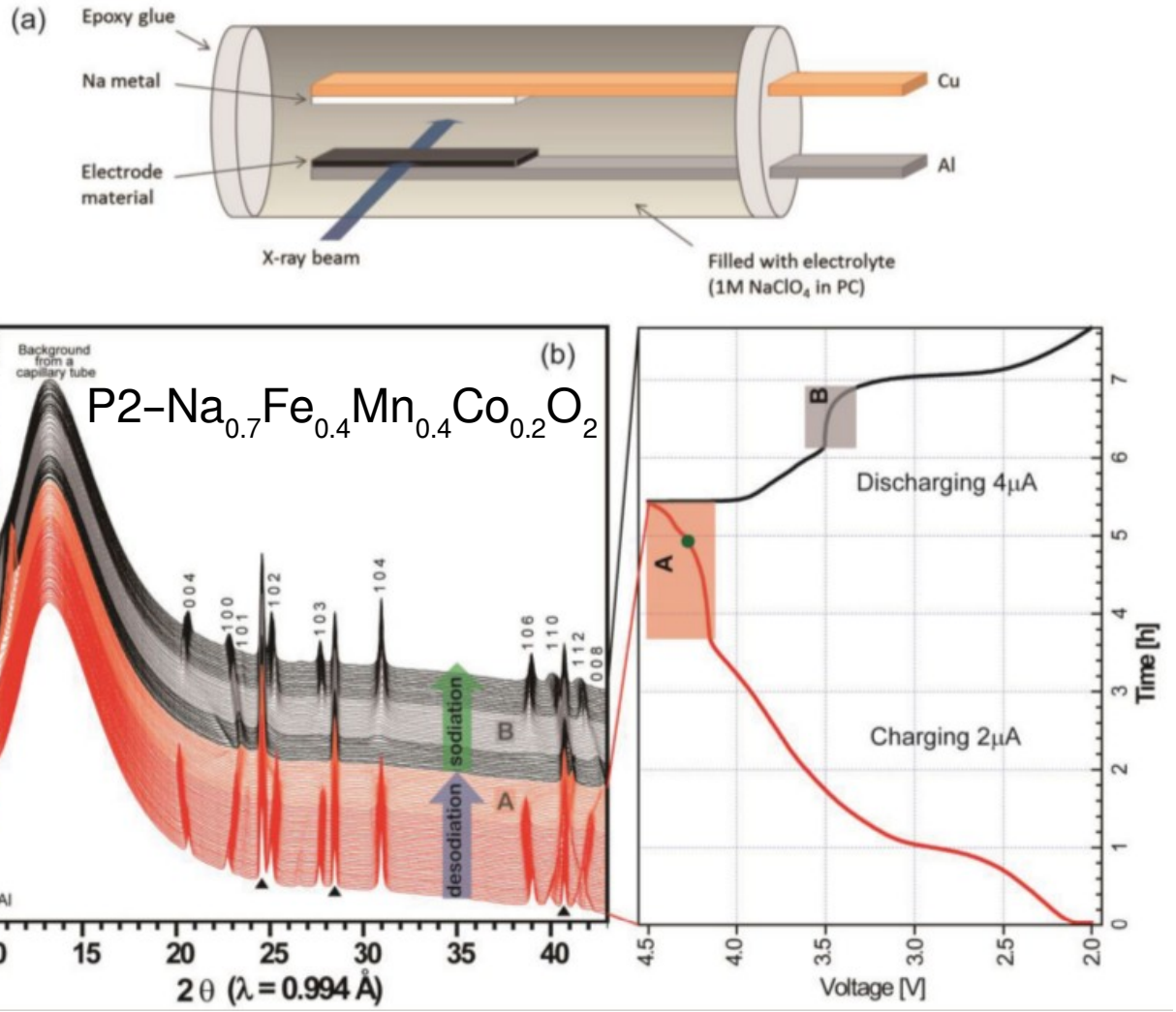






"Capillary-Based Micro-Battery Cell for in Situ X-Ray Powder Diffraction Studies of Working Batteries: A Study of the Initial Intercalation and Deintercalation of Lithium into Graphite."
Johnsen, R. E.; Norby, P.; Journal of Applied Crystallography 2013, 46 (6), 1537–1543.

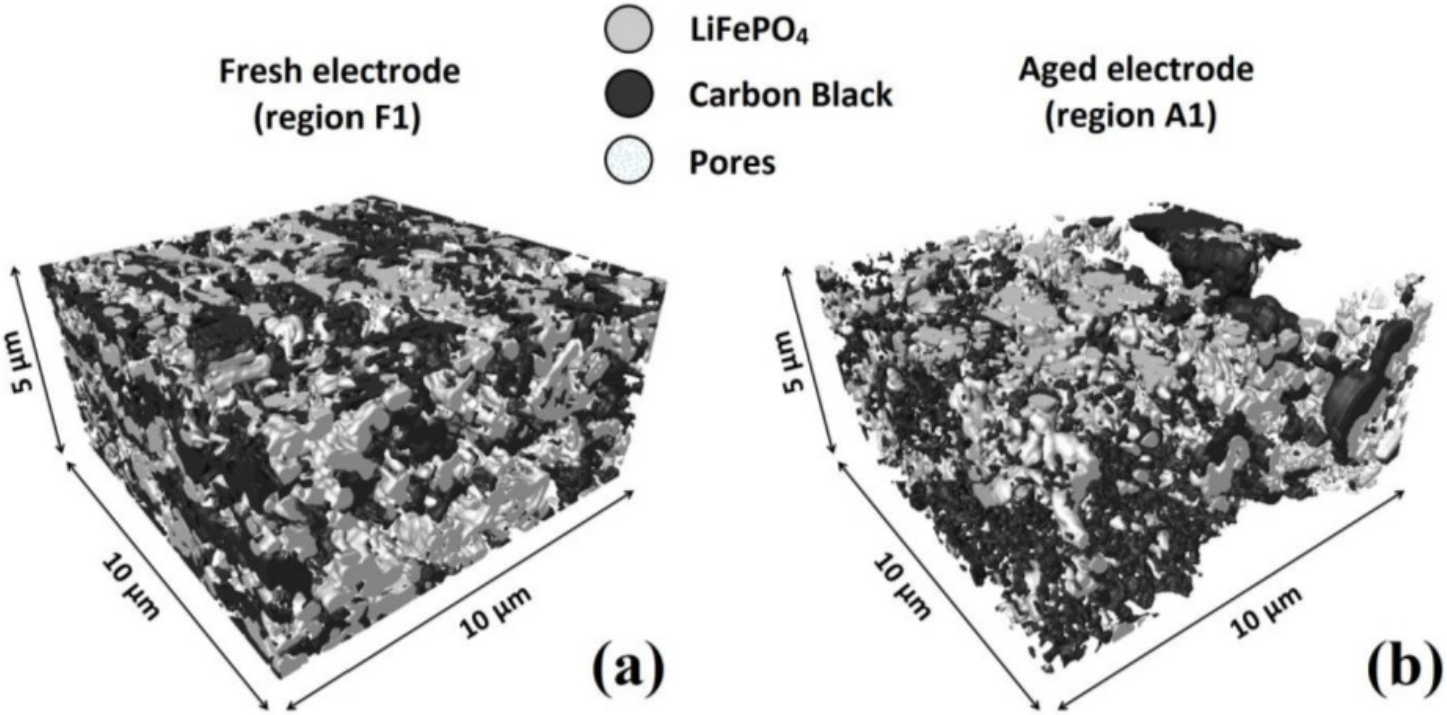
In-situ X-ray Diffraction II. [Na-ion]



Jung, Y. H.; Christiansen, A. S.; Johnsen, R. E.; Norby, P.; Kim, D. K.
"In Situ X-Ray Diffraction Studies on Structural Changes of a P2 Layered Material during Electrochemical Desodiation/Sodiation."
Adv. Funct. Mater. 2015, 25 (21), 3227–3237.

3D Reconstruction via FIB-SEM [Li-ion]

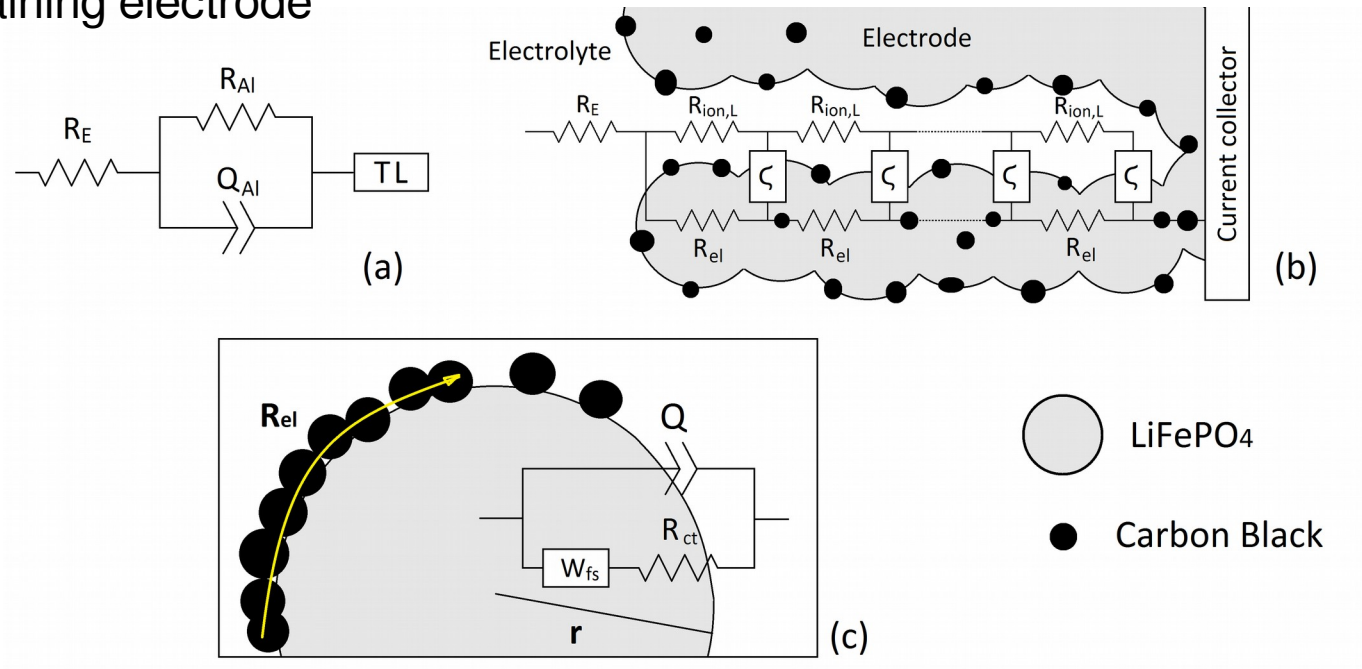
- LFP cathode aging studied using FIB-SEM + serial sectioning + 3D reconstruction
- Aging by cycling at 0.1 C for 100 cycles between 3.0 and 4.0 V -> 30% capacity
- Changes in network of conductive additive (carbon black) network observed



"Electron Microscopy Investigations of Changes in Morphology and Conductivity of $LiFePO_4$/C Electrodes",
Roberto Scipioni, Peter Stanley Jørgensen, Duc-The Ngo, Søren Bredmose Simonsen, Zhao Liu, Kyle Joseph Yakal-Kremski Hongqian Wang, Johan Hjelm, Poul Norby, Scott A. Barnett, Søren Højgaard Jensen, submitted to J. Power Sources

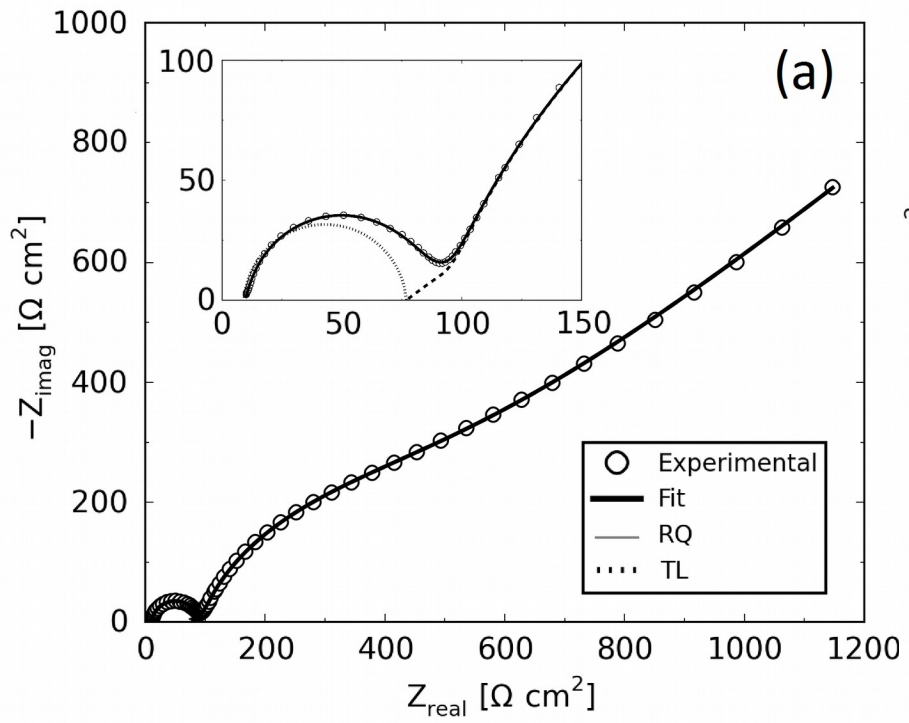
Degradation study by EIS [Li-ion]

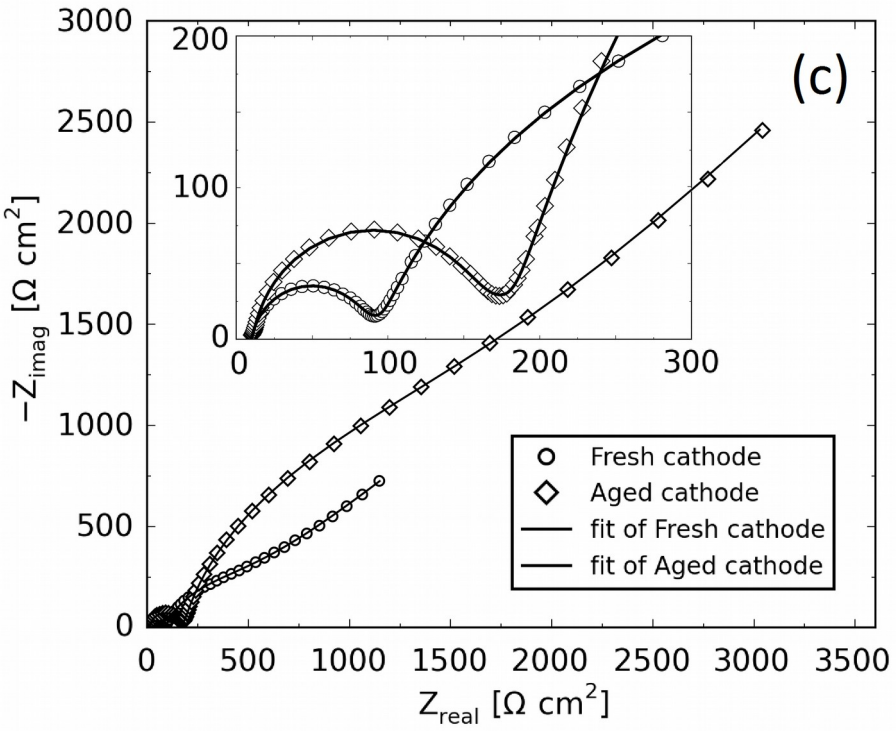
- LFP cathode aging studied in 3-electrode set-up using Impedance Spectroscopy
- The impedance was modeled using an equivalent circuit model including a generalized transmission line element to model the porous LFP containing electrode



"Low-voltage FIB/SEM Tomography for 3D Microstructure Evolution of $LiFePO_4$/C Electrode"
R. Scipioni, P. S. Jørgensen, D. T. Ngo, S. B. Simonsen, J. Hjelm, P. Norby, and S. H. Jensen
ECS Transactions, Accepted for publication (2015)

Degradation study by EIS [Li-ion]

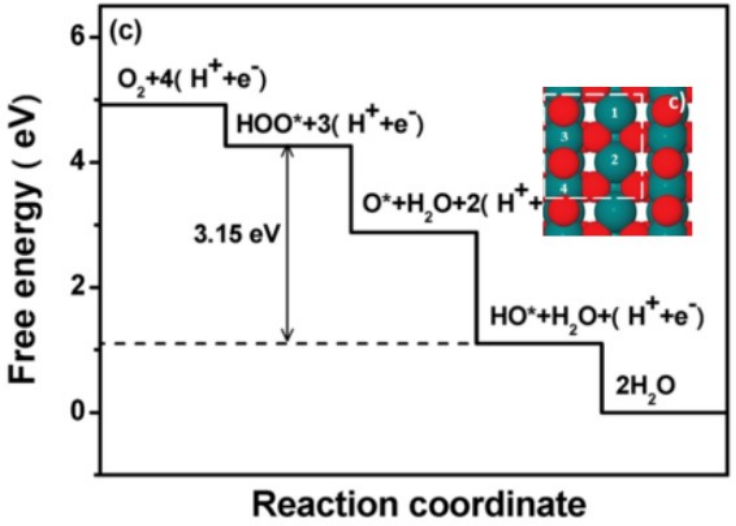


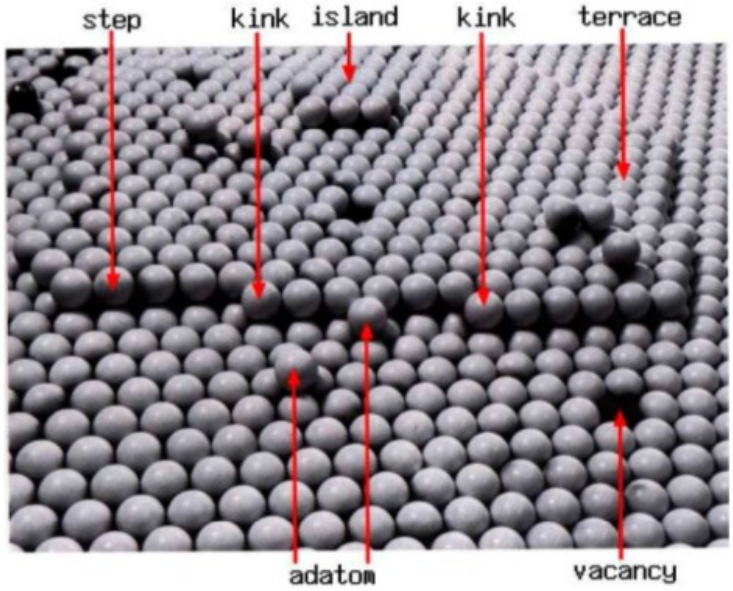


	RQ element		Transmission Line				
	R_{Al} (Ωcm^2)	C_{Al} (μF)	R_{ct} (Ωcm^2)	C_{dl} (F)	D ($cm^2 s^{-1}$)	$R_{ion,L}$ (Ωcm^{-1})	R_{el} (Ωcm^{-1})
Fresh	68	4.2	0.66	0.75	$2.7 \cdot 10^{-11}$	8760	21
Aged	142	3	3.27	0.64	$2.9 \cdot 10^{-11}$	17260	1546

ZAS [Zinc - Air, H2020 project]

- Atomistic studies of oxygen reduction
- Investigations of most stable close-packed surfaces of Zn and Zn-alloys
- Most stable stepped and kinked surfaces in selected electrolyte
- Construction of free energy diagrams
- Calculation of adsorption energies

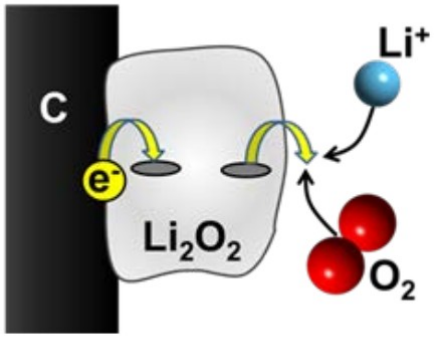




Mat4Bat

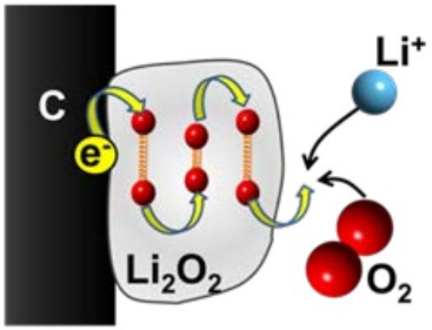
- In silico design of efficient materials for next generation batteries

Electron tunneling



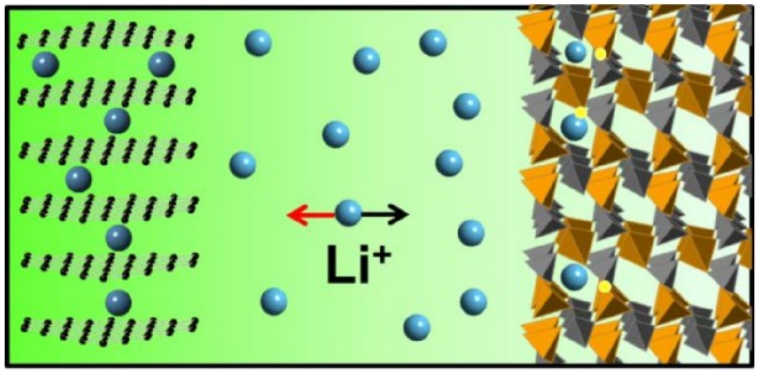
TD-DFT

Polaron hopping

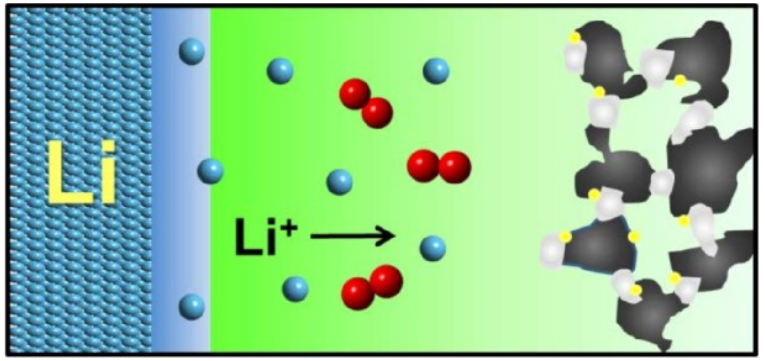


Markus Theory

Significant investment in computational infrastructure



Li - ion

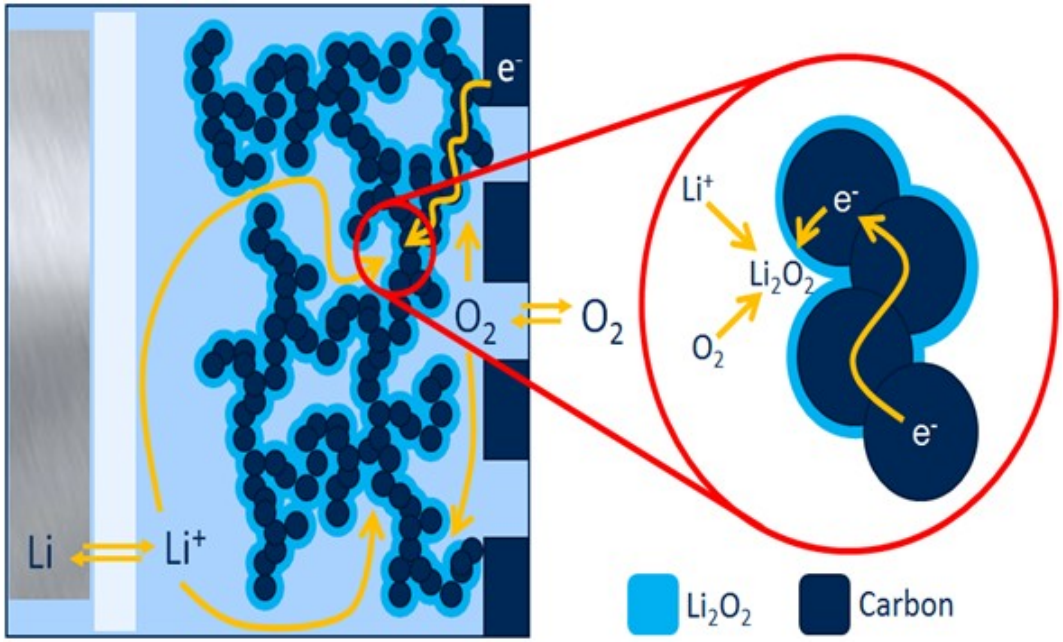


Metal - air

Project headed by Juan Maria García Lastra

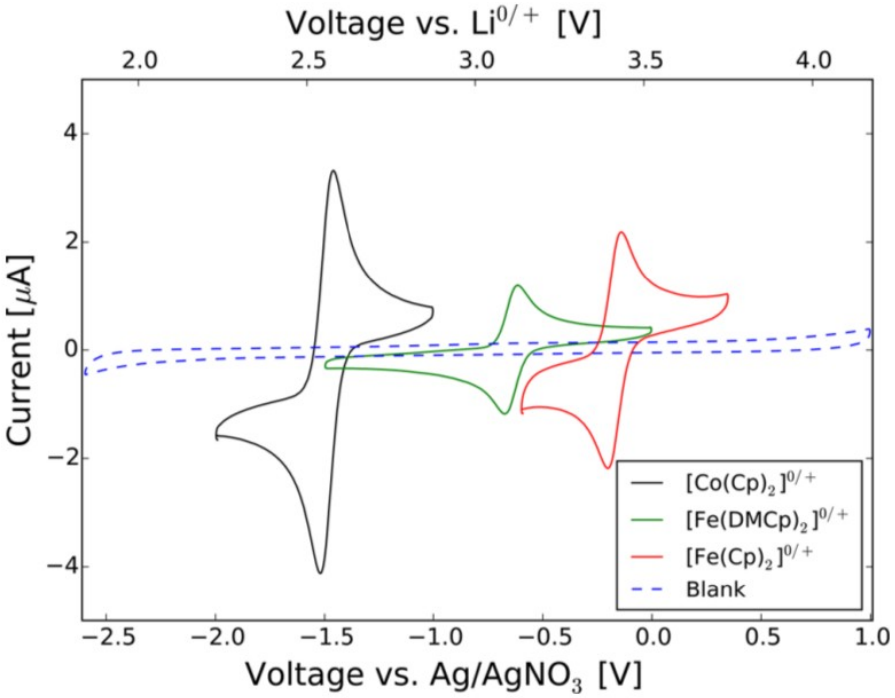
Aprotic Li-Air / Li-O_2 Cells

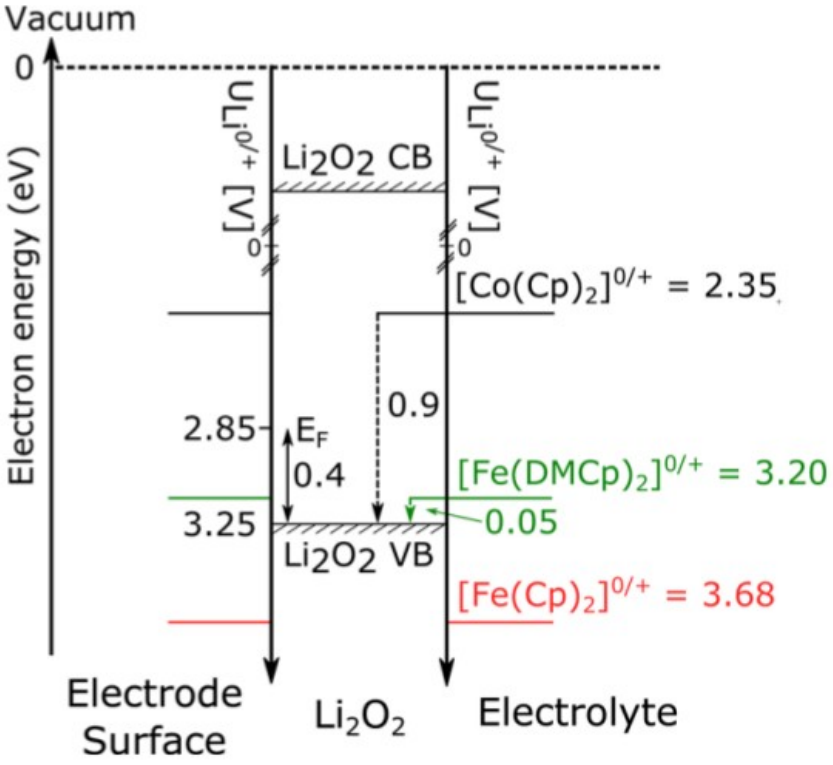
- Electrolyte: LiTFSI in 1,2-dimethoxyethane (DME)
- Under dry conditions Li_2O_2 deposits conformally on the porous carbon cathode
- Electrolyte (and carbon) stability a problem
- Sudden-death observed during discharge, due to poor electronic conductivity of Li_2O_2



Charge Transfer across thin Li_2O_2 films

- Investigation of potential dependence of charge transfer (tunnelling / polaron hopping?)
- Redox probing study using three different redox couples (metallocenes)

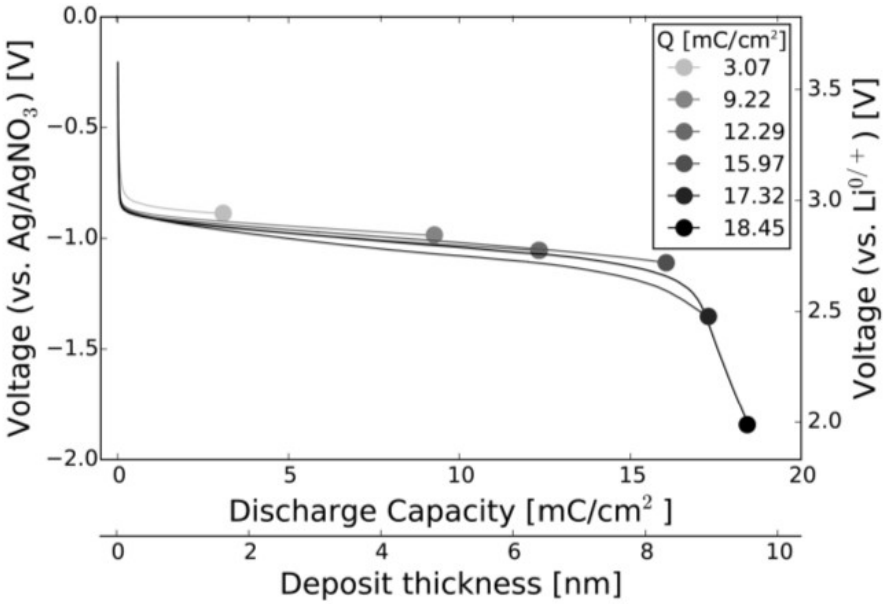


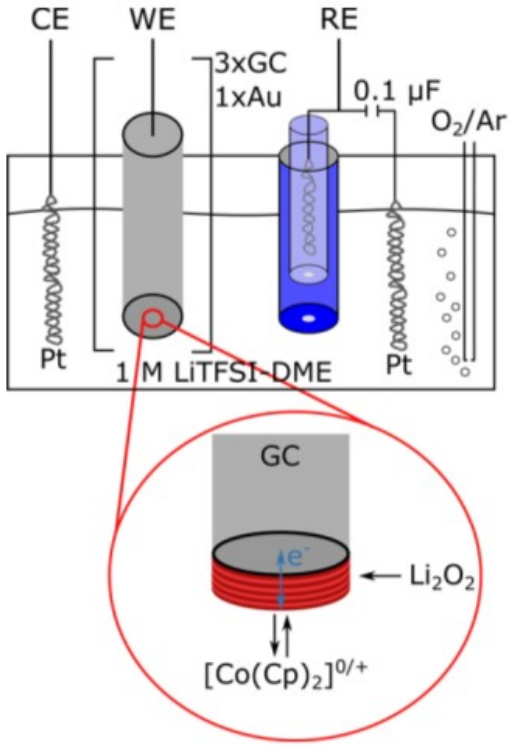


"Redox Probing Study of the Potential Dependence of Charge Transport Through Li_2O_2",
Kristian B. Knudsen, Alan C. Luntz, Søren H. Jensen, Tejs Vegge, Johan Hjelm
Journal of Physical Chemistry C, http://dx.doi.org/10.1021/acs.jpcc.5b08757

Charge Transfer across thin Li_2O_2 films

- Deposition of Li_2O_2 thin films on glassy carbon disk electrodes
- Conventional three-electrode cell used
- After deposition, a redox probe was introduced in the solution

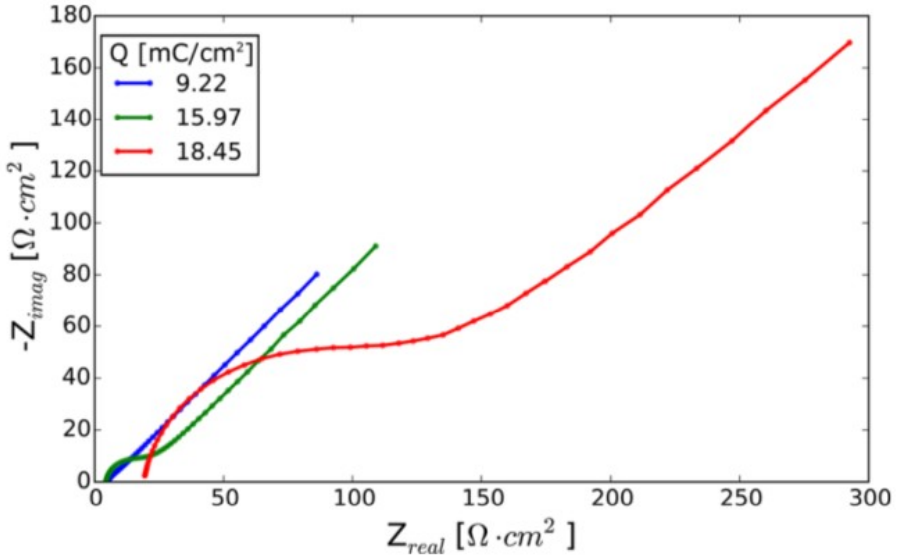


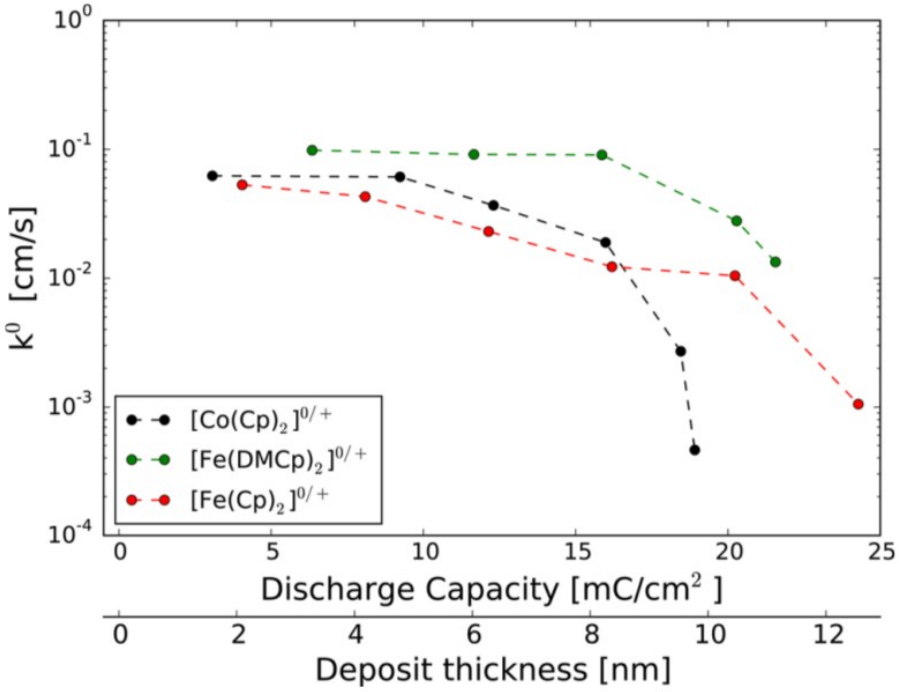


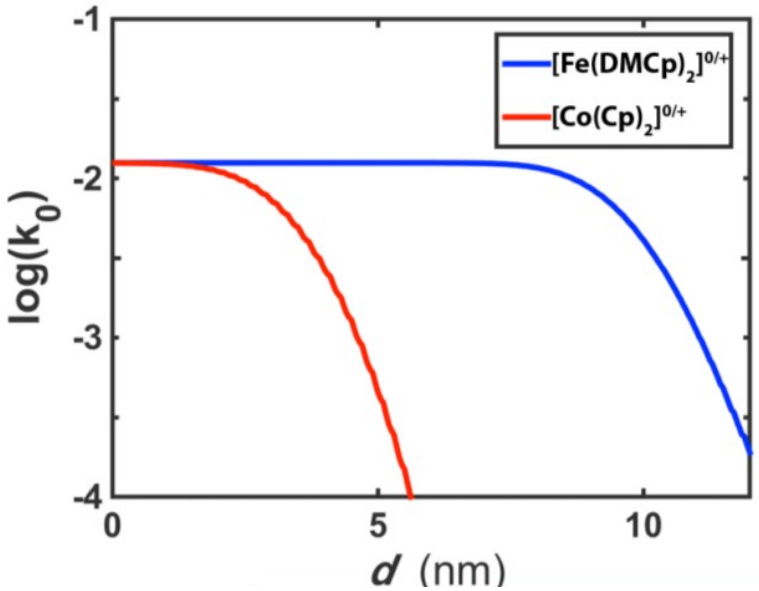
"Redox Probing Study of the Potential Dependence of Charge Transport Through Li_2O_2", Kristian B. Knudsen, Alan C. Luntz, Søren H. Jensen, Tejs Vegge, Johan Hjelm Journal of Physical Chemistry C, http://dx.doi.org/10.1021/acs.jpcc.5b08757

Charge Transfer across thin Li_2O_2 films

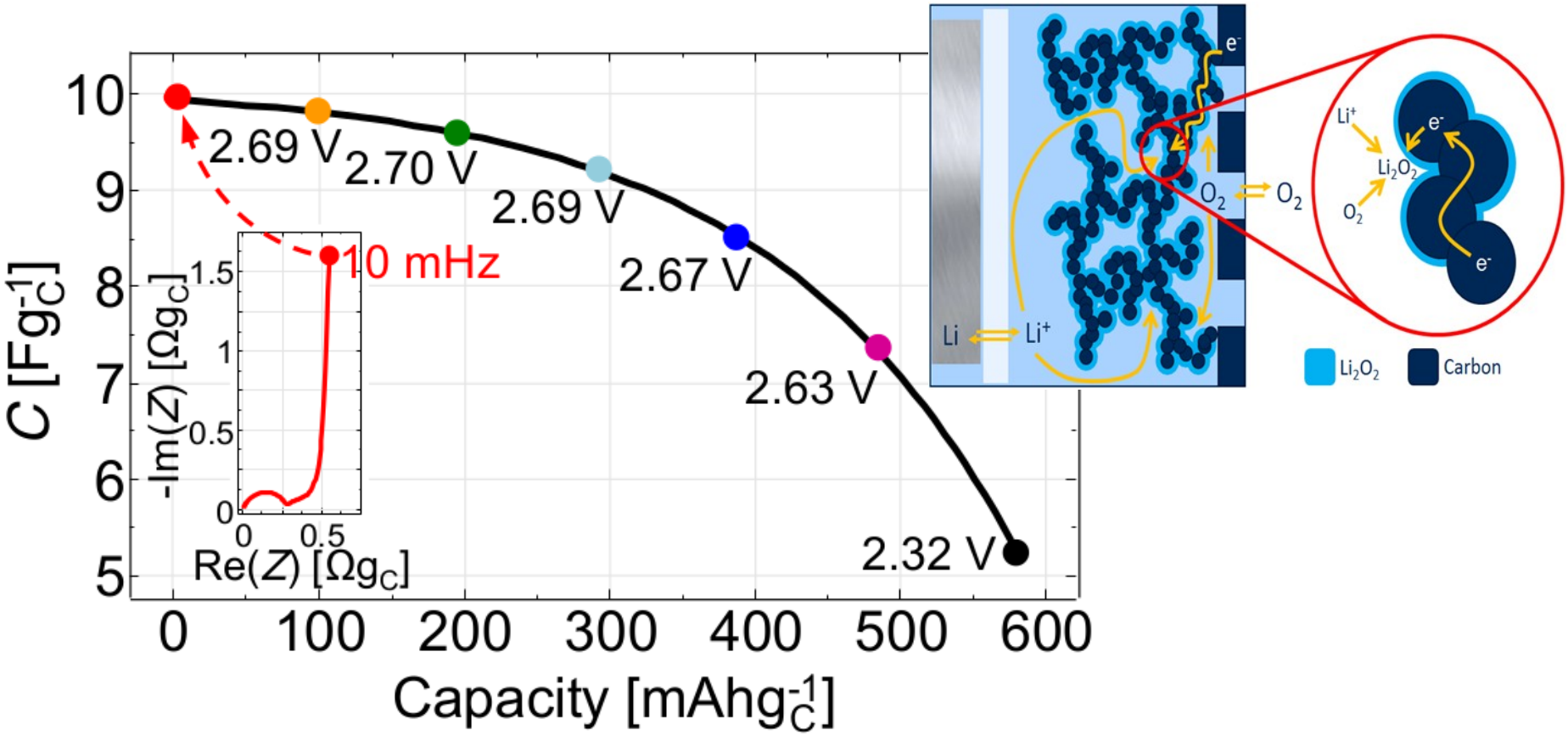
- Charge transfer resistance measured using impedance spectroscopy
- Heterogeneous charge transfer exchange rate vs Li_2O_2 thickness
- Results point to hole tunnelling as the dominant mechanism





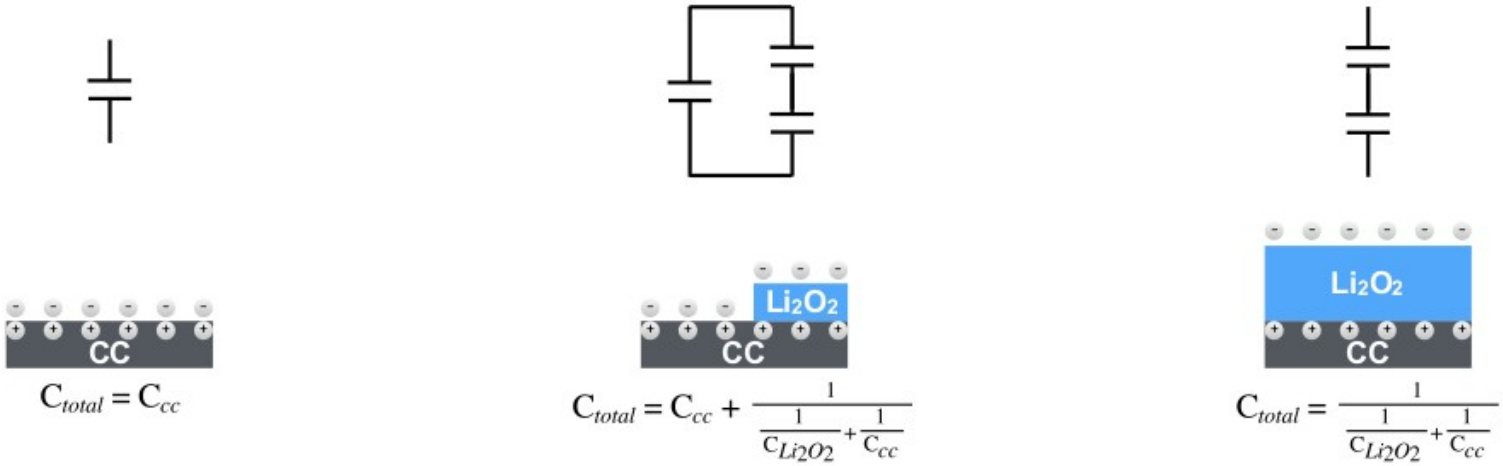


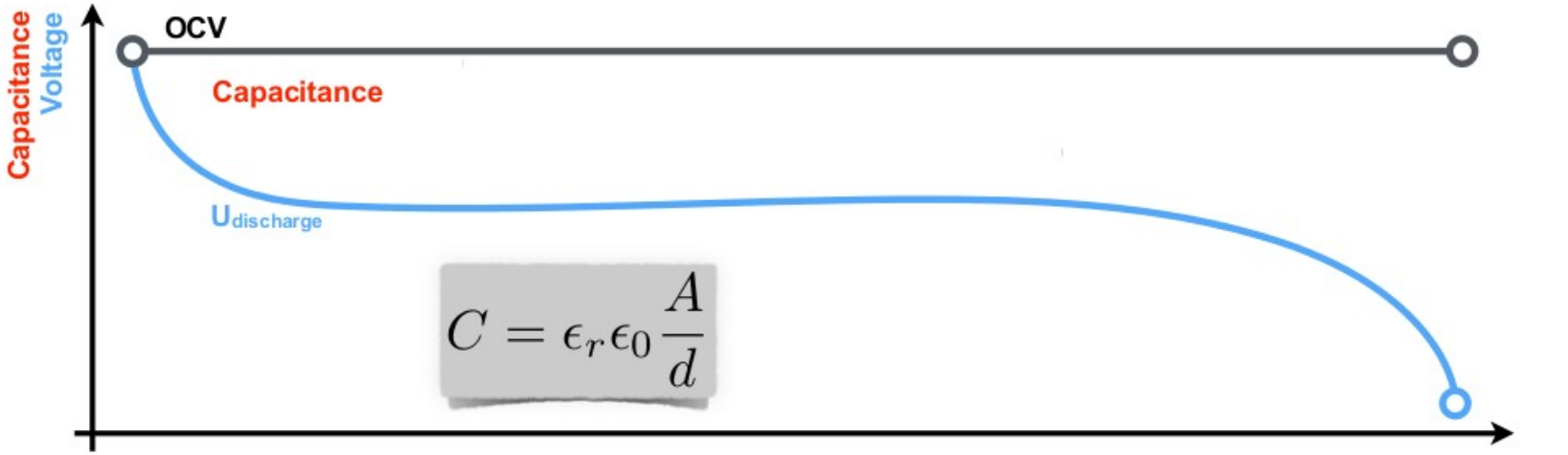
Battery Management for Metal-O_2 Systems



"Impedance-Based Battery Management for Metal-O_2 Systems."
Christensen, A. E.; Højberg, J.; Norby, P.; Vegge, T., J. Electrochem. Soc. 2015, 162 (10), A2075–A2079.

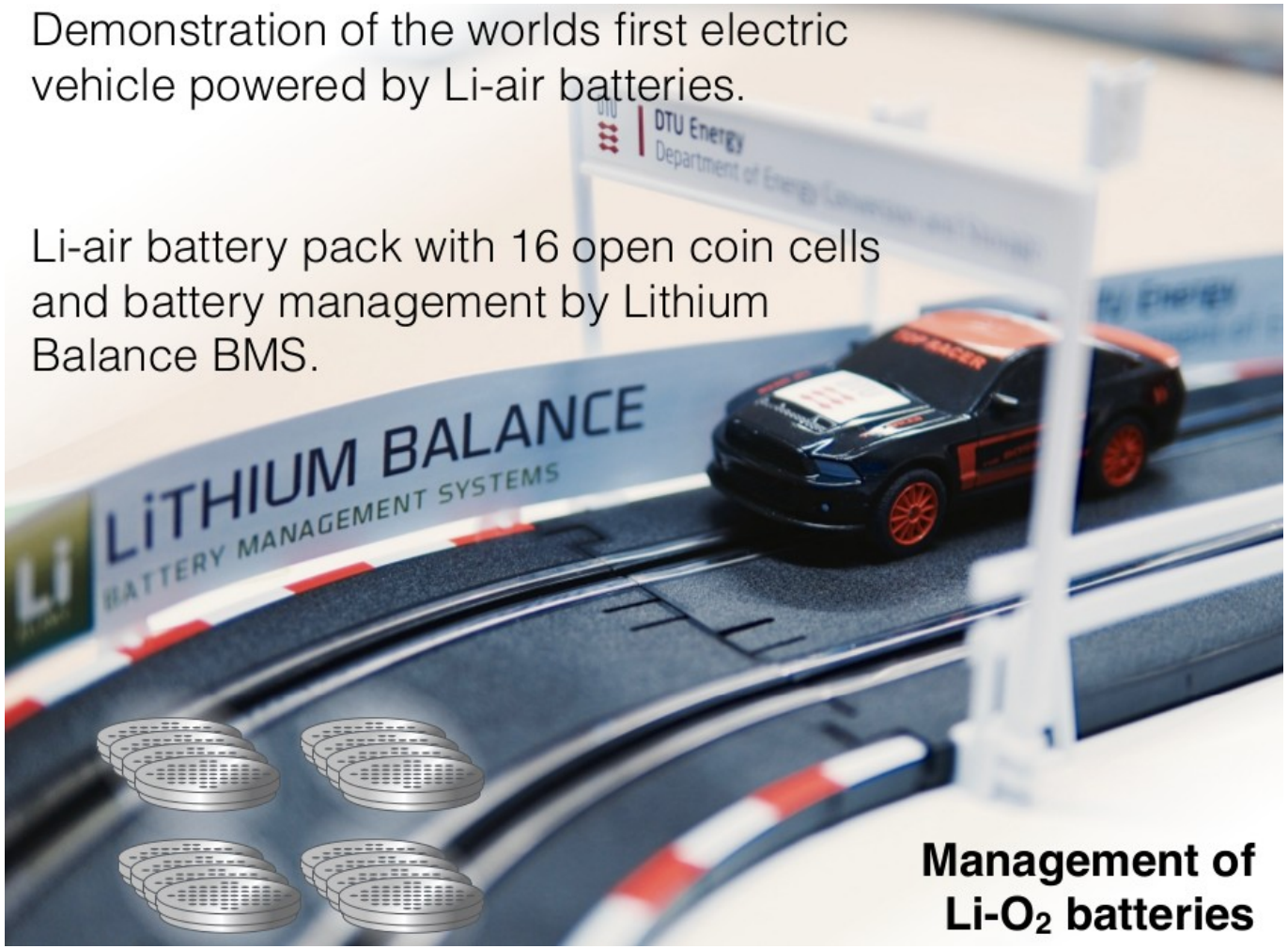
Battery Management / SOC Monitoring





"Impedance-Based Battery Management for Metal-O_2 Systems."
Christensen, A. E.; Højberg, J.; Norby, P.; Vegge, T., J. Electrochem. Soc. 2015, 162 (10), A2075–A2079.

First EV powered by Li-Air cells...



work by Andreas Elkjær Christensen

Posters

- "Zn near surface alloys for improved stability of zinc-air battery anodes"

Steen Lysgaard, Vladimir Tripkovic, Heine A. Hansen, Tejs Vegge

- "In situ X-ray diffraction studies of capillary based Li-air batteries"
  Mie Møller Storm, Rune Johnsen, Reza Younesi, Poul Norby

- "Oxide air-electrodes for Zn-Air Batteries"
  Vladimir Tripkovic, Steen Lysgaard, Heine A. Hansen, Tejs Vegge

- "Computational investigations of transport mechanisms across battery interfaces"
  Simon Loftager, Juan María García Lastra, Tejs Vegge

- "Analysis of the Interphase on Carbon Black Formed in High Voltage Batteries"
  Reza Younesi, Ane Sælland Christiansen, Scipioni, R.; Ngo, D.-T.; Simonsen, S. B.; Edström, K.; Hjelm, J.; Norby, P.

Acknowledgements

Battery group at DTU Energy

Funding bodies...

InnovationsFonden

HORIZON 2020

VILLUM FONDEN

SEVENTH FRAMEWORK PROGRAMME

...and thanks for your attention!