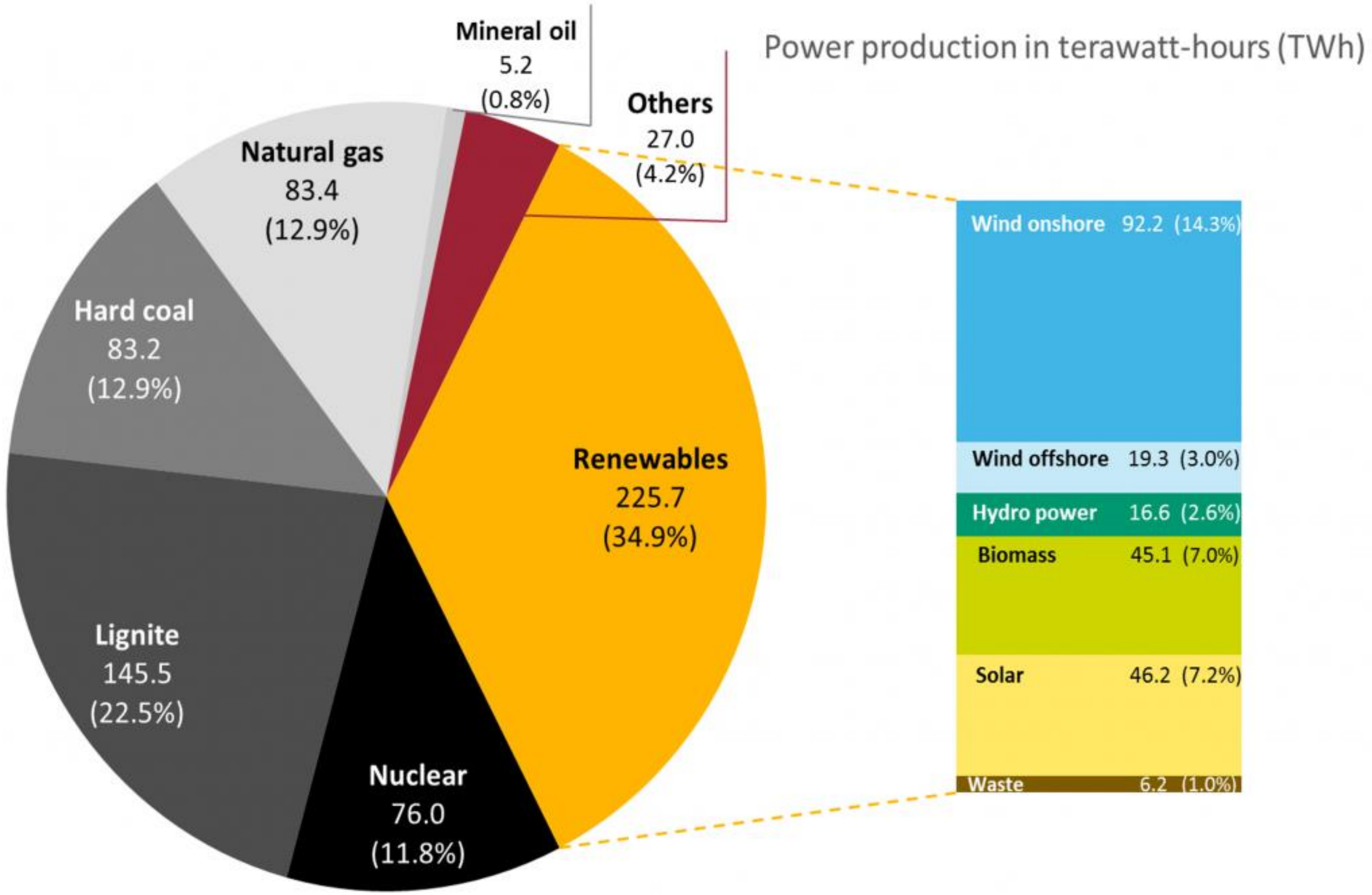
SONAR

L02-1954 - Modelling for the search for new active materials for redox flow batteries

Jens Noack
Fraunhofer ICT

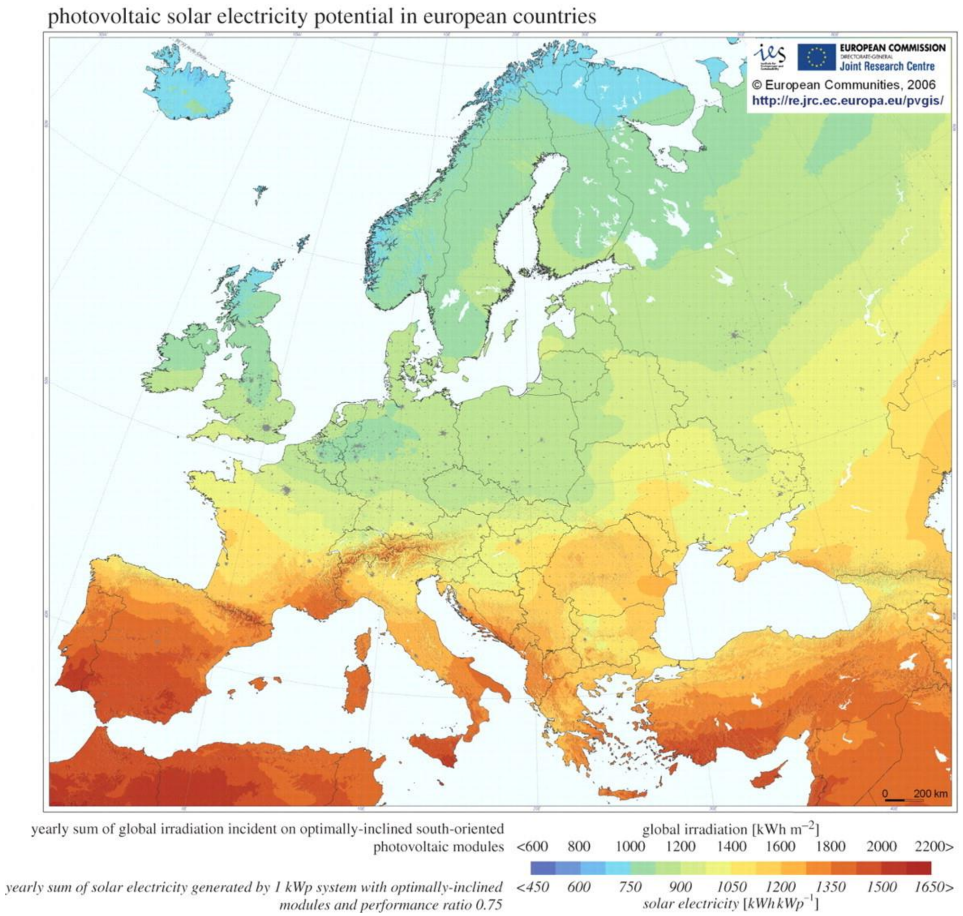
241st ECS Meeting
Vancouver/Canada 2022
June 1st 2022

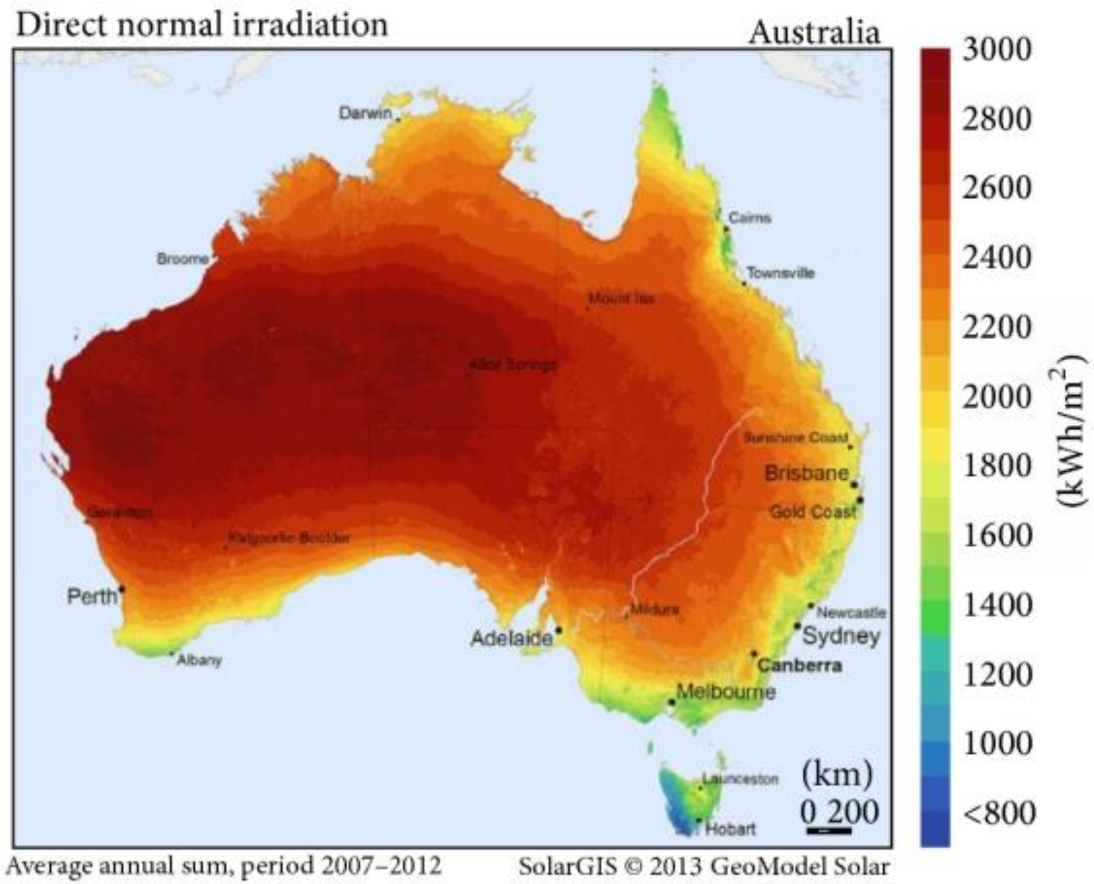
Share of energy sources in gross German power production in 2018



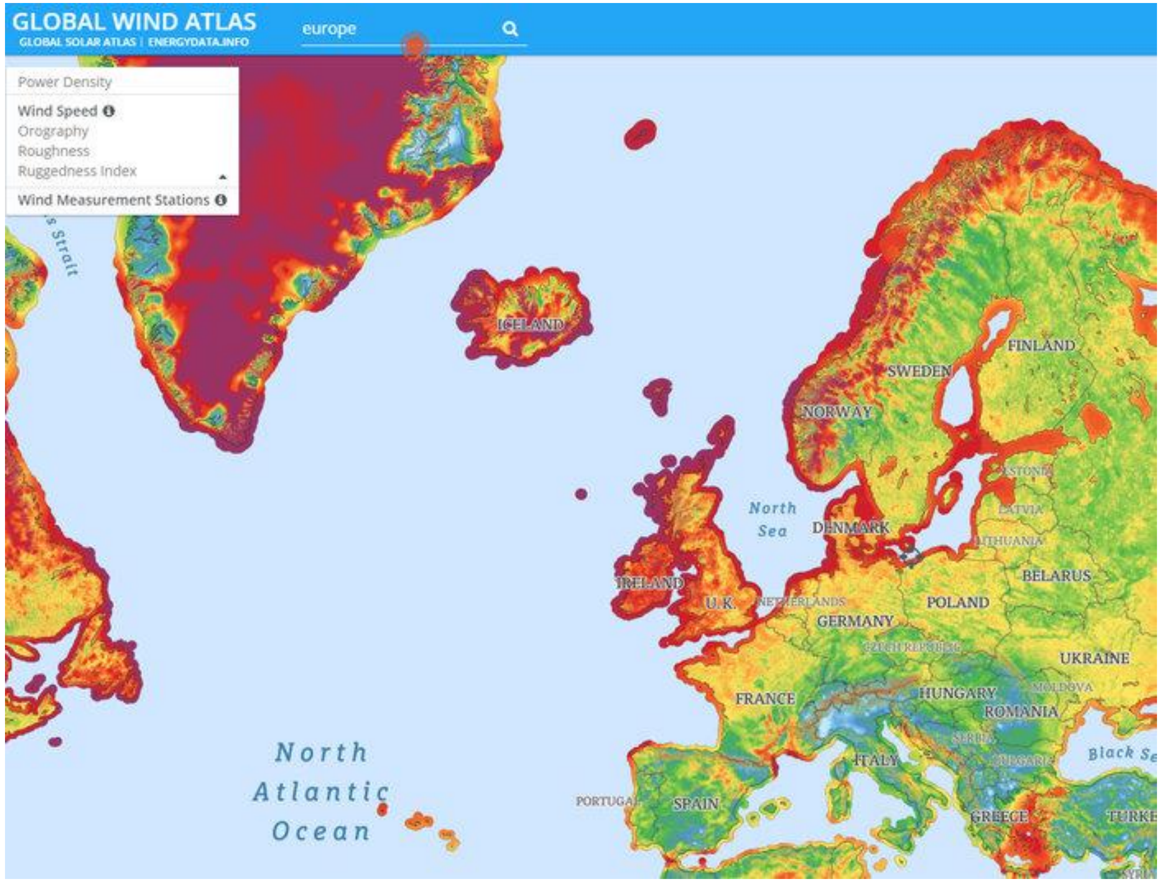
Source: Clean Energy Wire

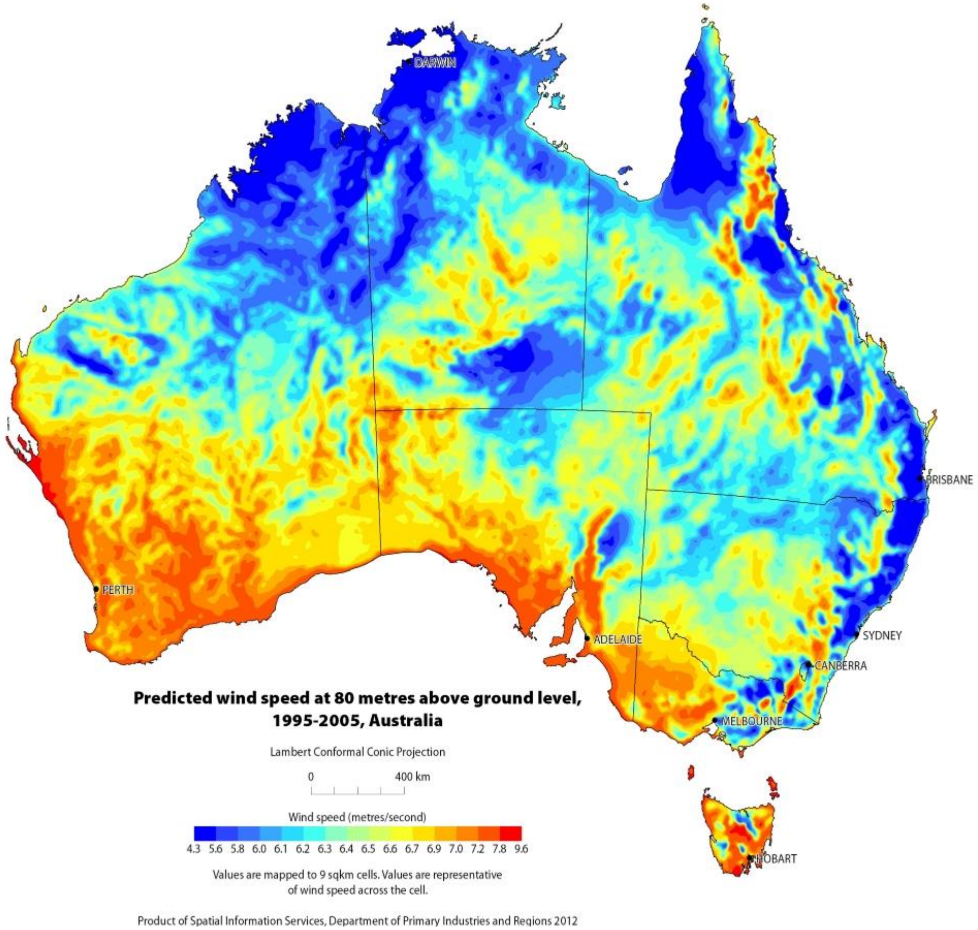
Renewable energy potential - Sun





Renewable energy potential - Wind





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The need for energy storage

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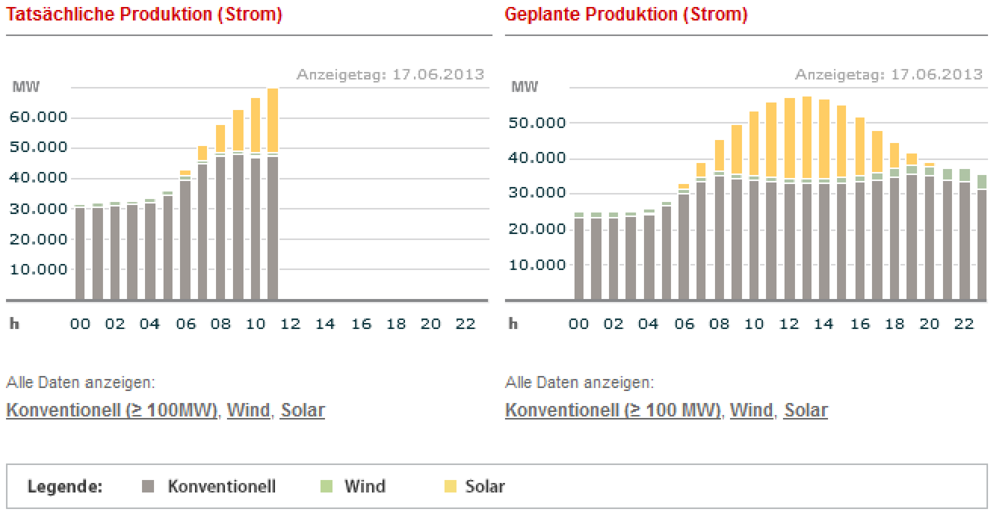
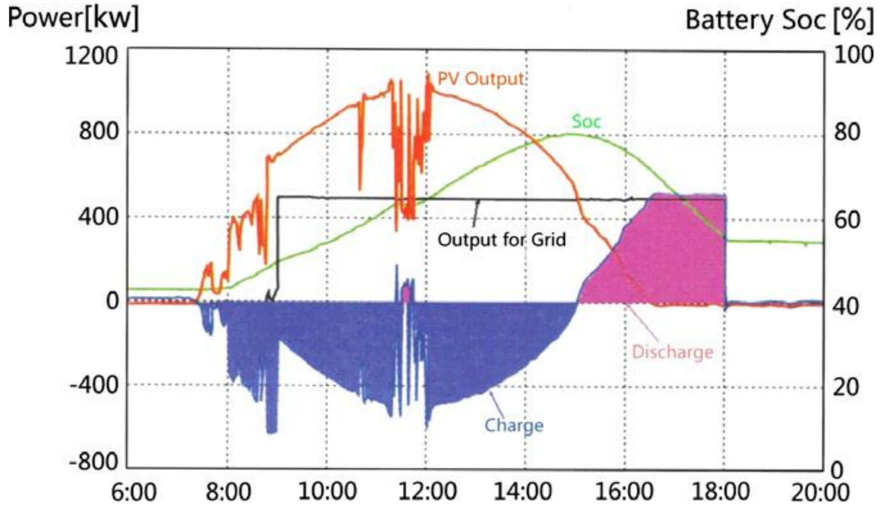


Production of electricity in Germany at 2013-06-17 Source: EEX



- Photovoltaic and wind power are decentralized fluctuating generators
- Fluctuations put a strain on electricity grids - Expensive grid extensions
- *(Re-adjustment through fast gas power plants)*
- With a high proportion of renewable energy, storage facilities are necessary for times when no sun is shineing and no wind is blowing.

-> Decentralised energy storage

Storage possibilities

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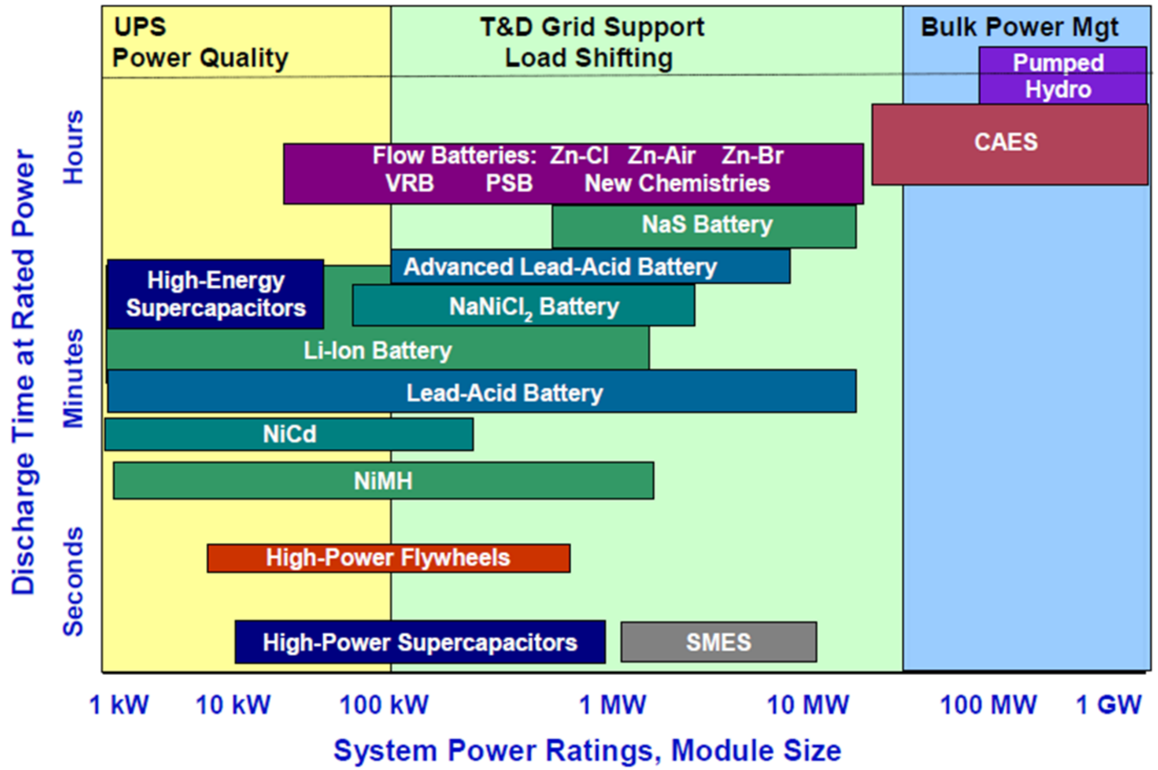
Storage

Mechanical
- Compressed Air (CAES)
- Pumped Hydro
- Flywheel
- Gravitational Potential

Electrical
- Supercaps
- Capacitors
- Superconducting magnetic (SMES)

Electrochemical
- Flow-Batteries
- Lithium-Ion-Batteries
- Sodium-Sulfur-Batteries
- Fuel Cells

Thermal
- Molten salt
- Phase-change material



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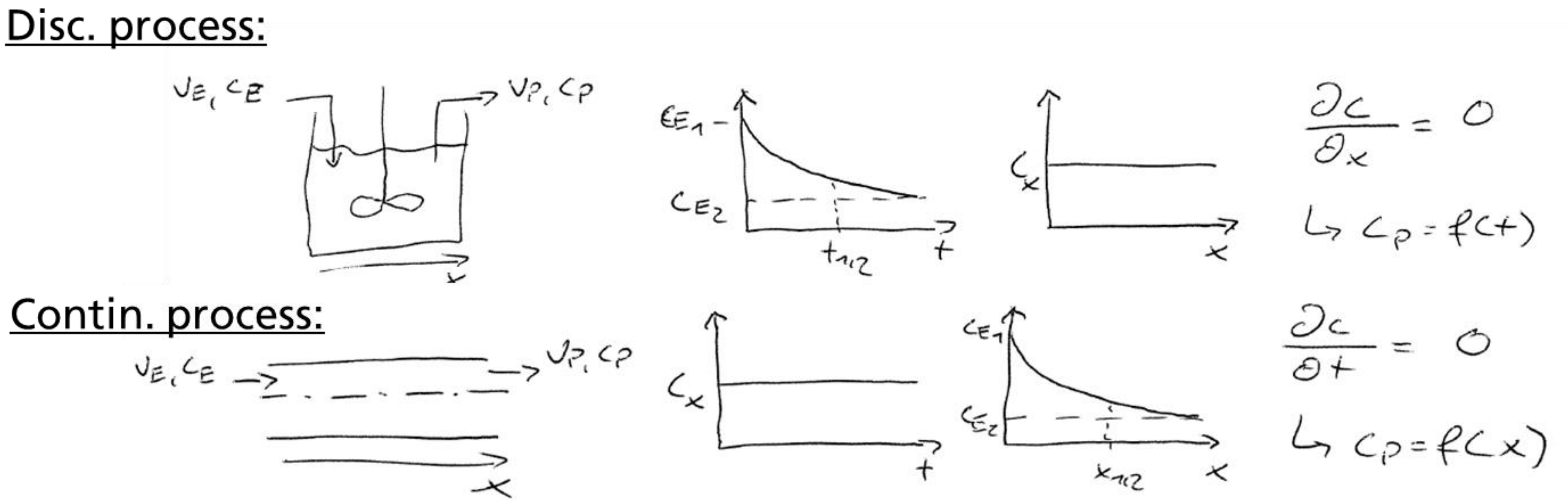
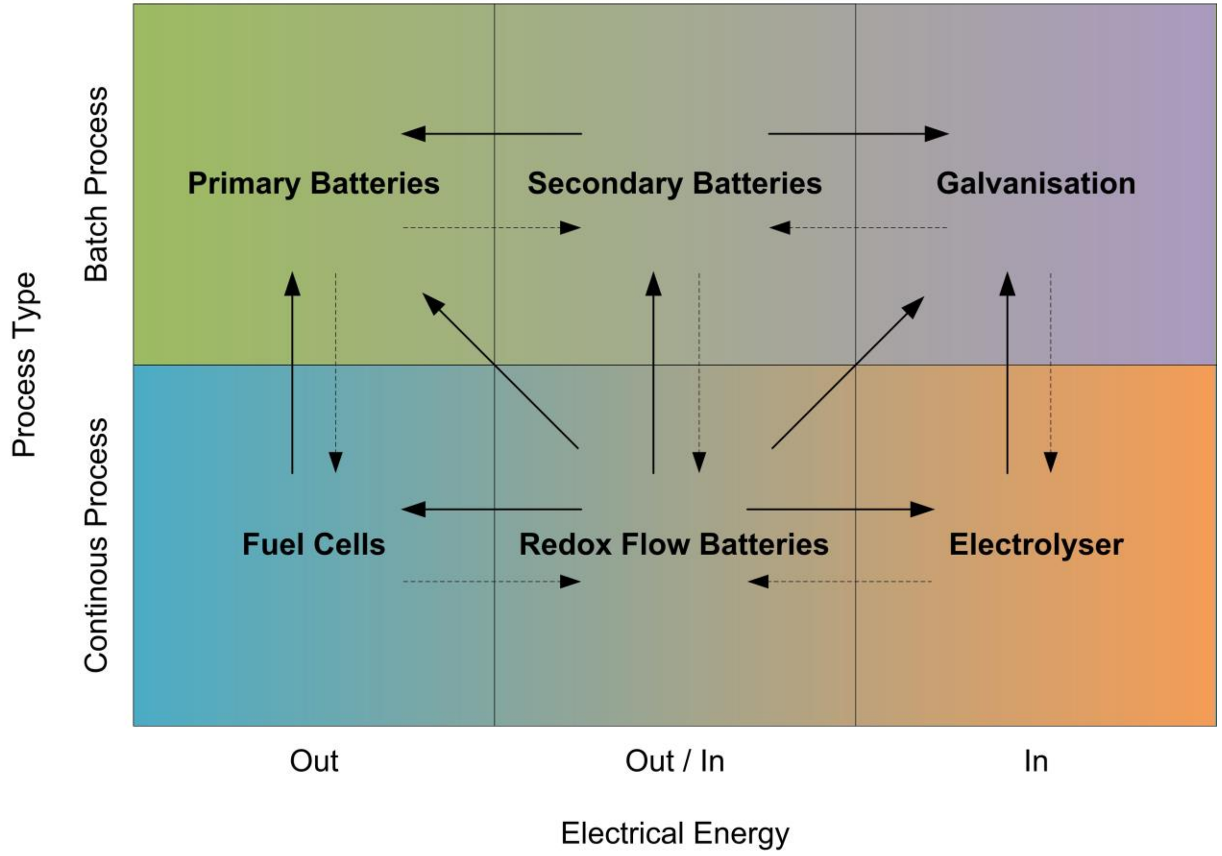
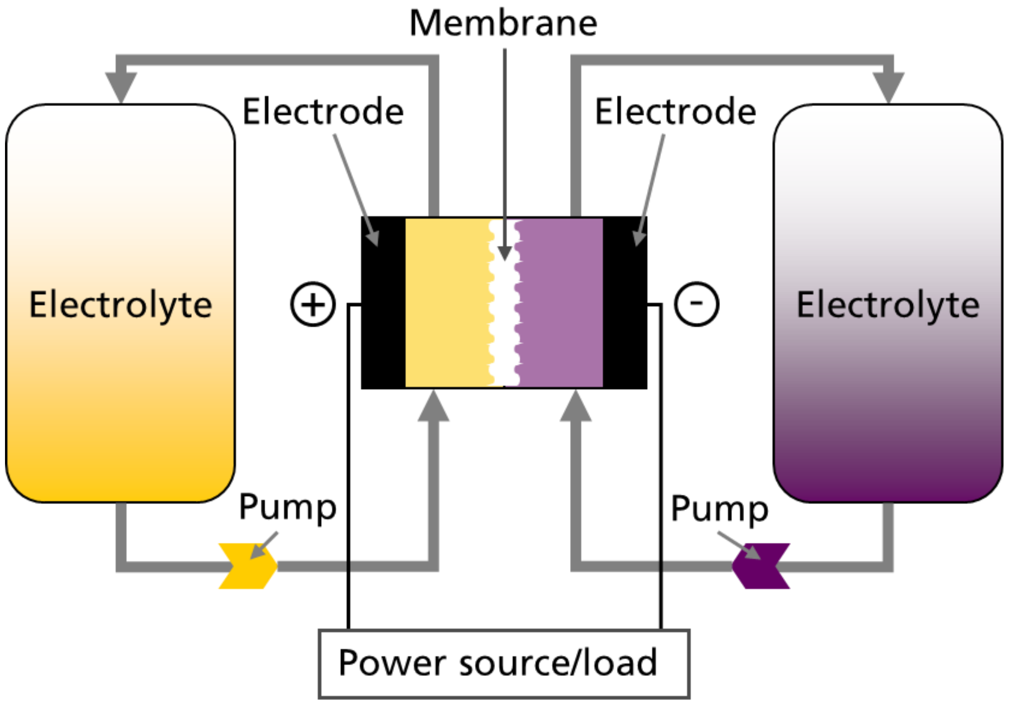
What is a flow battery?

- Arbitrary classical classification
  - Primary batteries
  - Secondary batteries
  - Fuel cells
- Flowing suspensions, reversible fuel cell, changing aggregate state, reversibility, Electrolysis, Galvanisation,... ?
- Better: Classification by basic process types



What is a flow battery?





Flow Batteries are electrochemical energy converter, which use flowing active materials!

15 years flow battery research at Fraunhofer ICT

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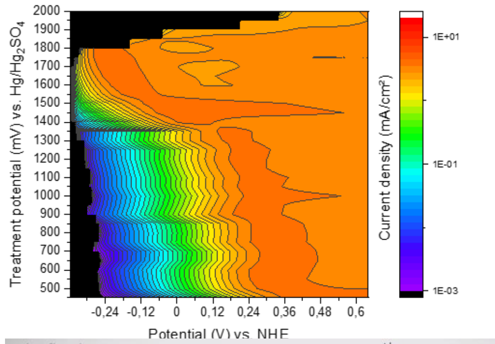

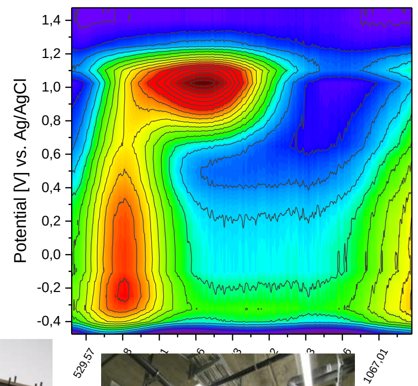

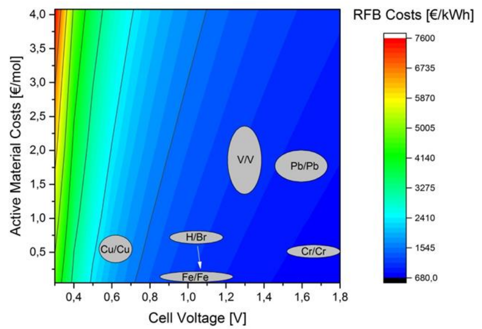

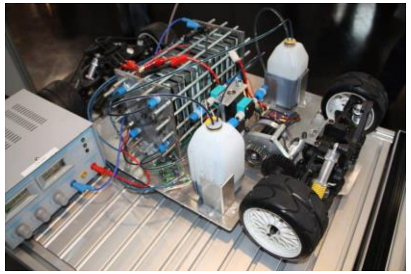

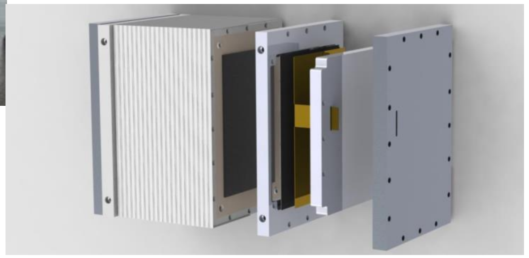

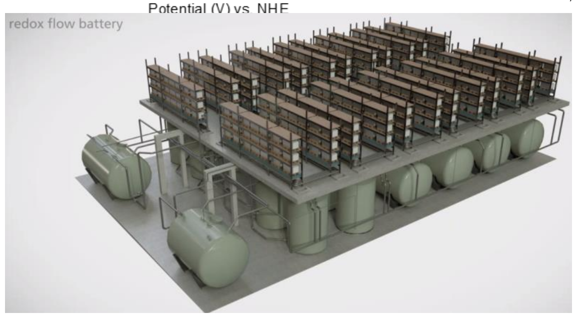

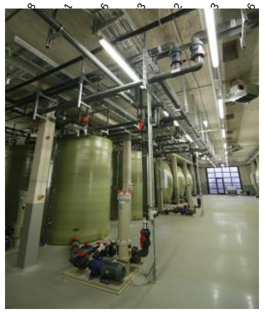

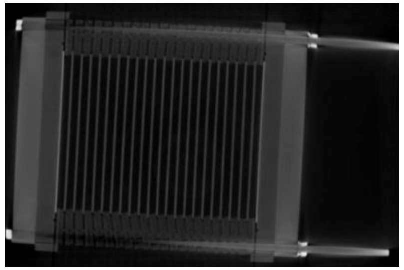

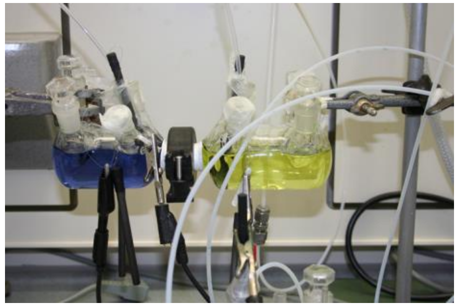

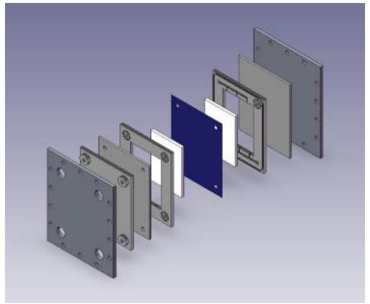

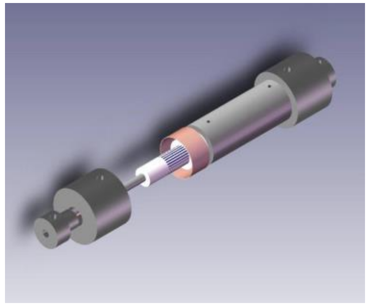

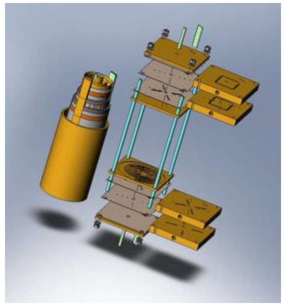

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Introduction - H2020 context

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H2020-EU.3.3.4. - A single, smart European electricity grid

LC-BAT-4-2019 - Advanced Redox Flow Batteries for stationary energy storage

LC-BAT-3-2019 - Modelling and simulation for Redox Flow Battery development

Project information

HIGREEW

Grant agreement ID: 875613

Status
Ongoing project

Start date: 1 November 2019
End date: 28 February 2023

Funded under:
H2020-EU.3.3.4.

Overall budget:
€ 3 786 747,50

EU contribution
€ 3 786 747,50

Coordinated by:
CENTRO DE INVESTIGACION COOPERATIVA DE ENERGIAS ALTERNATIVAS FUNDACION, CIC ENERGIGUNE FUNDAZIOA
Spain

Project information

CUBER

Grant agreement ID: 875605

Status
Ongoing project

Start date: 1 January 2020
End date: 31 December 2023

Funded under:
H2020-EU.3.3.4.

Overall budget:
€ 3 999 823,75

EU contribution
€ 3 999 823,75

Coordinated by:
AARHUS UNIVERSITET
Denmark

Project information

MELODY

Grant agreement ID: 875524

Status
Ongoing project

Start date: 1 January 2020
End date: 31 December 2023

Funded under:
H2020-EU.3.3.4.

Overall budget:
€ 3 996 612,50

EU contribution
€ 3 987 612,50

Coordinated by:
TECHNISCHE UNIVERSITEIT DELFT
Netherlands

Project information

BALIHT

Grant agreement ID: 875637

Status
Ongoing project

Start date: 1 December 2019
End date: 30 November 2022

Funded under:
H2020-EU.3.3.4.

Overall budget:
€ 4 098 552,50

EU contribution
€ 4 098 552,50

Coordinated by:
AIMPLAS - ASOCIACION DE INVESTIGACION DE MATERIALES PLASTICOS Y CONEXAS
Spain

Project information

SONAR

Grant agreement ID: 875489

Status
Ongoing project

Start date: 1 January 2020
End date: 31 December 2023

Funded under:
H2020-EU.3.3.4.

Overall budget:
€ 2 820 535

EU contribution
€ 2 385 985

Coordinated by:
FRAUNHOFER GESELLSCHAFT ZUR FOERDERUNG DER ANGEWANDTEN FORSCHUNG E.V.
Germany

Project information

CompBat

Grant agreement ID: 875565

Status
Ongoing project

Start date: 1 February 2020
End date: 31 January 2023

Funded under:
H2020-EU.3.3.4.

Overall budget:
€ 1 751 485

EU contribution
€ 1 751 485

Coordinated by:
AALTO KORKEAKOULUSAATIO SR
Finland

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Motivation

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- **Huge number of possibilities of organic active materials for redox flow batteries**

- Laboratory testing is time consuming and costly
  - Chemical tests (e.g. solubility, stability)
  - Electrochemical half-cell tests (e.g. potentials, kinetics)
  - Cell & system tests (performance)

- Techno-economics ? -> CAPEX ?
- Behavior in the grid ? -> Levelised cost of storage?

- Only LCOS (levelised cost of storage (lifetime cost / lifetime energy throughput)) gives compareable values!

Table 1: Selected examples of inorganic redox pairs for RFBs. The cell color corresponds to the pH value of the electrolyte: red - acidic, blue - alkaline, orange - neutral, green - acidic and alkaline. Development stage: A - half cell studies, B - prototype tested, C - technology is commercialized, * or Br_3^-/Br^- 1.06 V (based on polybromide formation), the potentials may vary depending on the electrolyte composition due to complex formation

Cathode		Mn_2O_3/MnO_2	$Fe(CN)_6^{4-}/Fe(CN)_6^{3-}$	Cu/Cu^+	I^-/I_3^-	Fe^{2+}/Fe^{3+}	VO^{2+}/VO_2^+	$Br^-/ClBr_2^-$	Br^-/Br_2^*	NpO_2^{2+}/NpO_2^+	I_2/IO_3^-	O^{2-}/O_2	$Cr^{3+}/HCrO_4^-$	Cl^-/Cl_2	Pb^{2+}/PbO_2	Mn^{2+}/Mn^{3+}	Ce^{3+}/Ce^{4+}	Co^{2+}/Co^{3+}
Anode	**E^0,V**	0.15	0.36	0.52	0.54	0.77	0.99	1.04	1.09	1.14	1.2	1.23	1.35	1.36	1.46	1.54	1.72	1.82
$Al/Al(OH)_4^-$	-2.31											B						
$Zn/Zn(OH)_4^{-2}$	-1.22	B	B															
Zn/Zn^{2+}	-0.76				B	B	B	B	C					B			B	
Fe/Fe^{2+}	-0.45					B												
S_2^{2-}/S	-0.43		B						C			B						
Cr^{2+}/Cr^{3+}	-0.41					C			A				B					
Cd/Cd^{2+}	-0.40					B												
V^{2+}/V^{3+}	-0.26					B	C	B				B				B	B	B
Pb/Pb^{2+}	-0.13														B			
Sn/Sn^{2+}	-0.14								B									
H_2/H^+	0.00					B	B		B					B				
Ti^{3+}/TiO^{2+}	0.04					A		A						A		B		
Cu^+/Cu^{2+}	0.15			B											B			
Np^{3+}/Np^{4+}	0.15									B								
Sn^{2+}/Sn^{4+}	0.15					B			B									
Cu/Cu^{2+}	0.34														B			
I^-/I_2	0.54										A							
Fe^{2+}/Fe^{3+}	0.77															B		

Development of a model-based high-throughput screening method

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Introduction

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Modelling for the search for new active materials for redox flow batteries

H2020-LC-BAT-3-2019

Project start: January 2020
Project end: December 2023 (4 Years)

7 Institutions, 4 Universities, 2 Research Organisations, 6 Companies (IEB)

Coordinator: Fraunhofer Gesellschaft (Germany)

5 Countries, 3 EU Countries, 1 H2020 Associated country (Switzerland), 1 External (Australia)

Project funding: 2.8 M€ 2.4 M€ (EU), 430 k€ (UNSW Australia)

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H2020-LC-BAT-2019-2020 (LC-BAT-3-2019) –

Modelling and Simulation for Redox Flow Batteries

The objective is to develop ***mathematical models for numerical simulation*** *and* ***high-volume pre-selection*** *of multi-species electrolyte flow and electrochemistry. Models should allow the characterisation of* ***new chemicals and designs****, the* ***related charge, mass and heat transport mechanisms, identifying cell-limiting mechanisms, forecasting cell performance and optimising the design and scale-up****. Of particular interest are performances in terms of* ***cell voltage, energy and power density, reliability and cost****.*

The simulation models should be ***validated with experimental examples*** *from known chemistries and representative prototypes, and show how new chemistries can be explored.*

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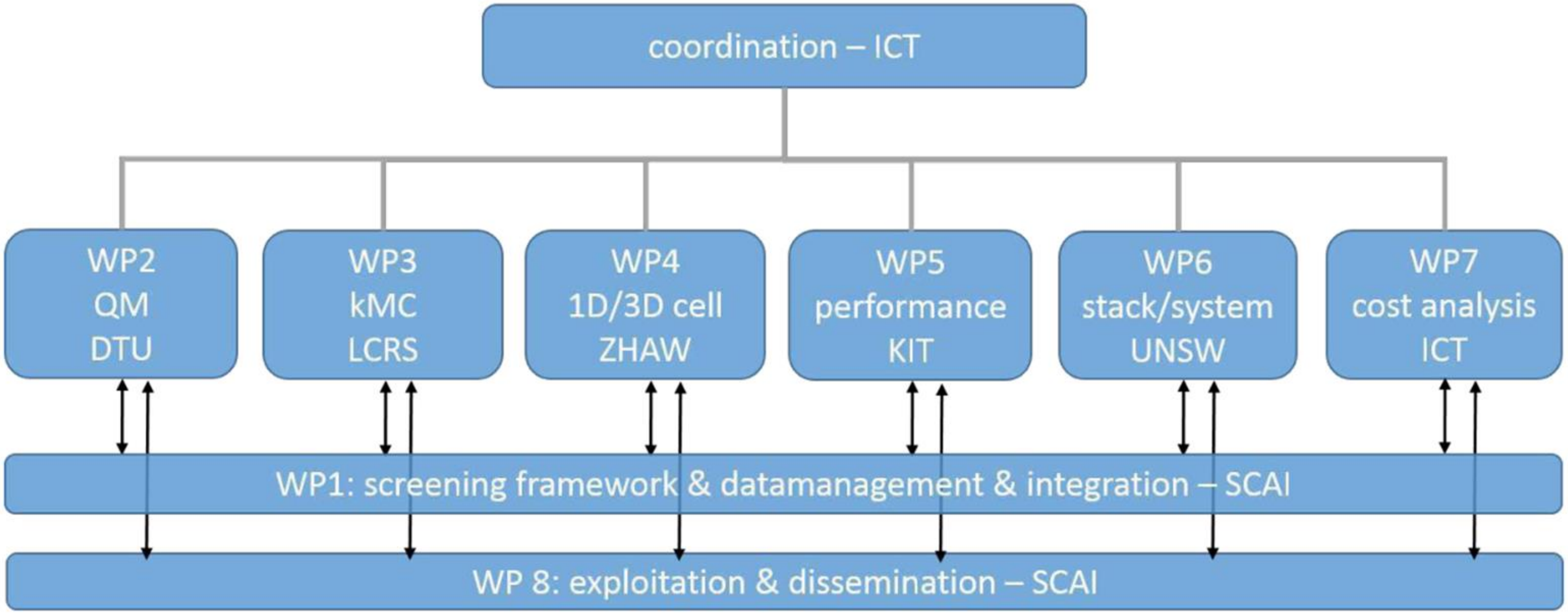
Introduction - Approach



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Introduction - Partner

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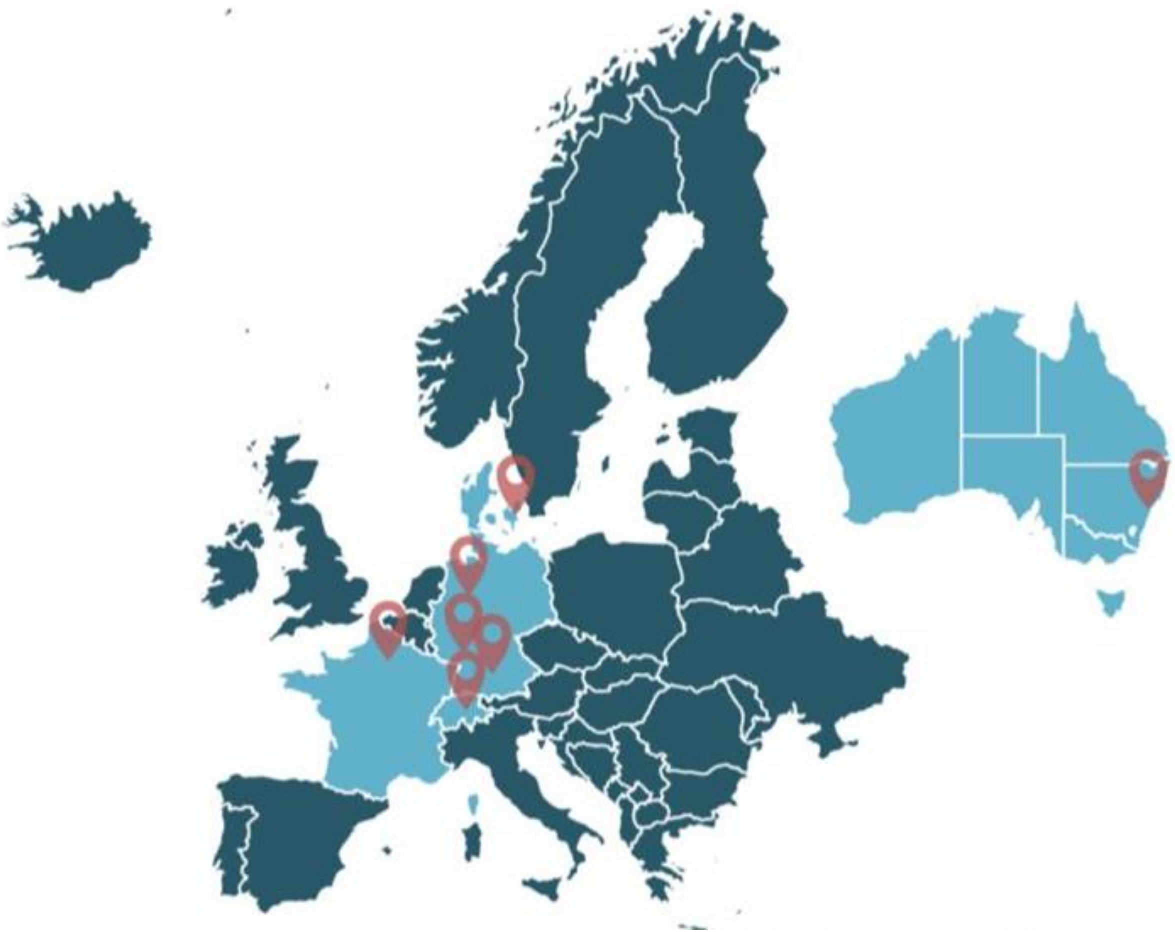

Fraunhofer-Institute for Chemical Technology (ICT)
GERMANY

Fraunhofer-Institute for Algorithms and Scientific Computing (SCAI)
GERMANY

Technical University of Denmark (DTU)
DENMARK

CNRS-Laboratoire de Réactivité et Chimie des Solides (LRCS)
FRANCE

Zurich University of Applied Science (ZHAW)
SWITZERLAND

Karlsruhe-Institute for Technology (KIT)
GERMANY

University of New South Wales (UNSW)
AUSTRALIA

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Introduction - Industrial Exploitation Board

Enerox
BUILDING ENERGY STORAGE INFRASTRUCTURE

volterion

JENA BATTERIES

sgl carbon

SCHMALZ

SCM
Software for Chemistry & Materials

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Introduction - Approach

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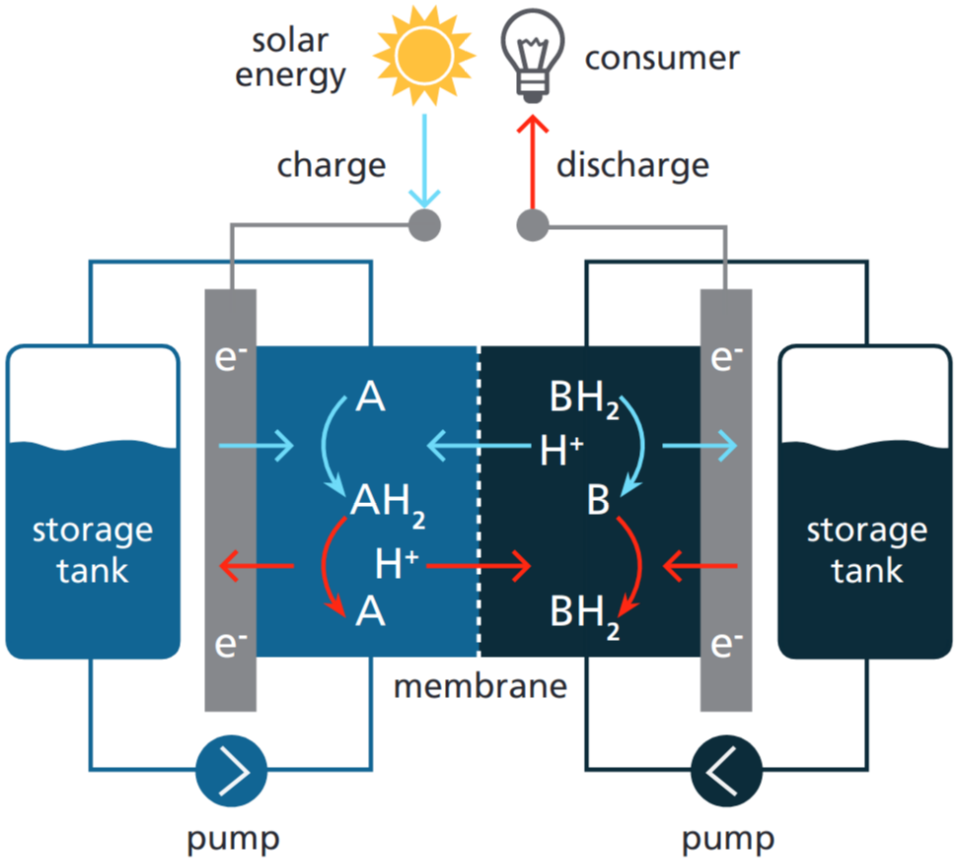

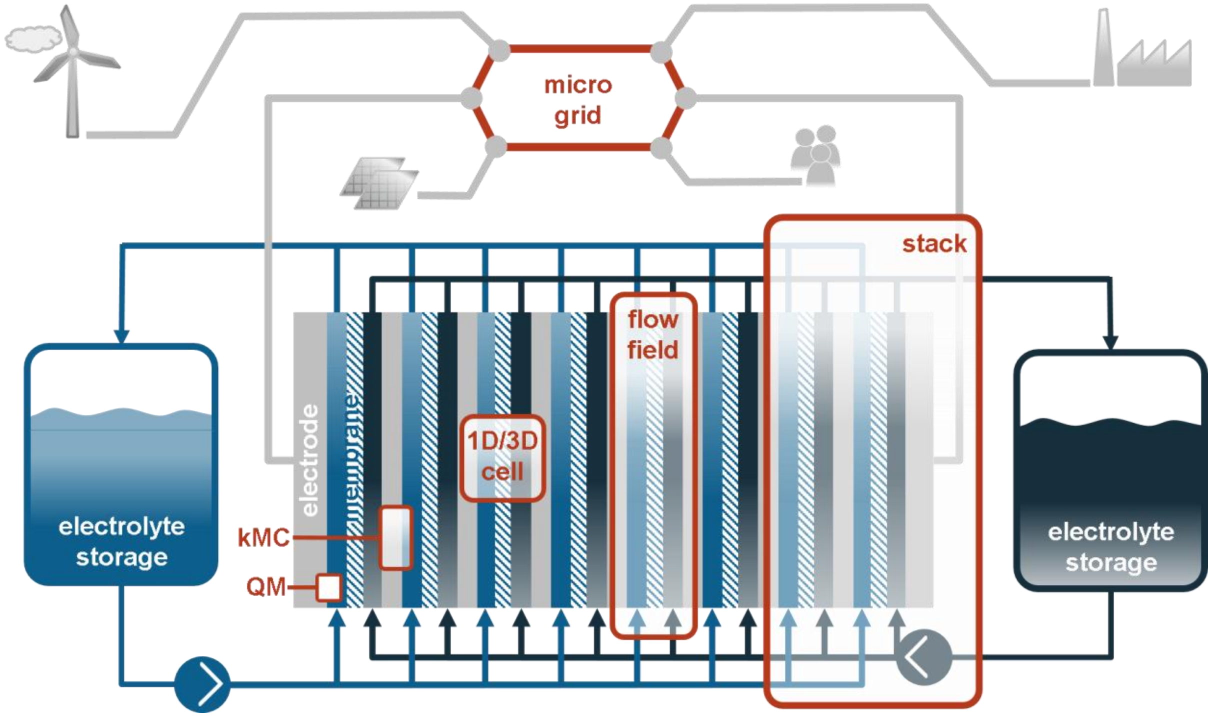

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Introduction - Approach

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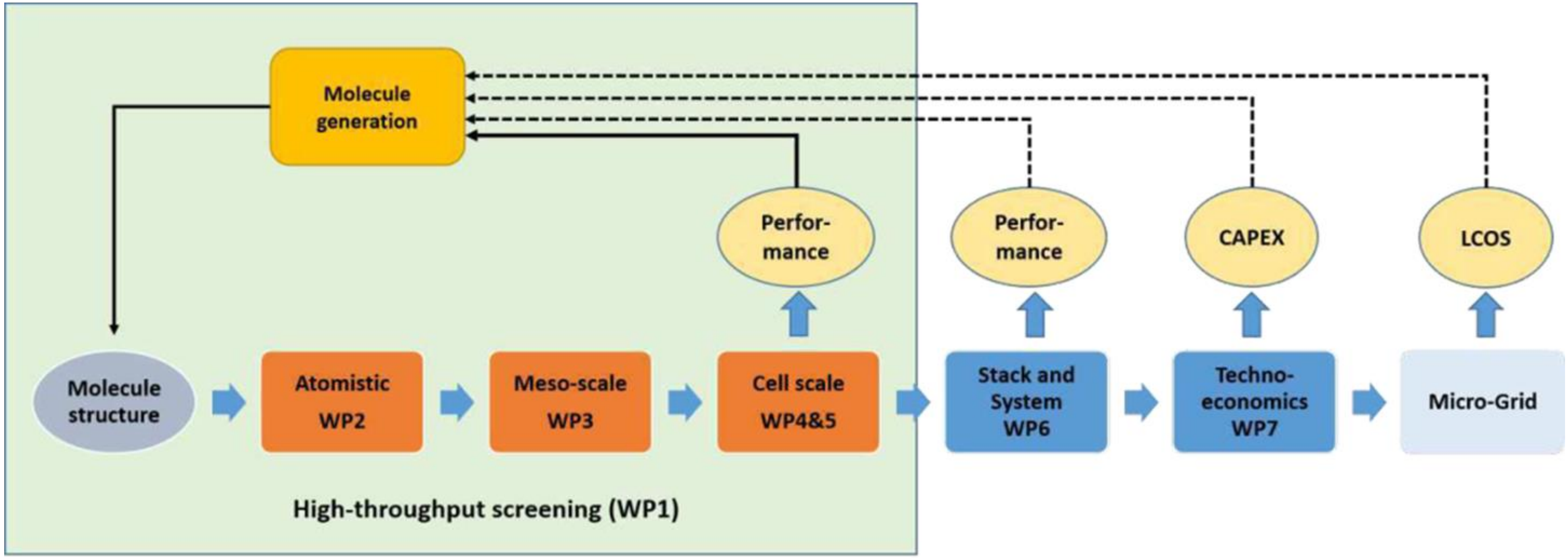
Atoms / Molecules → Electrode reactions / Half cell → 1D Cell → 3D Cell → System → Techno-economics → Gríd behavior



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Electronic structure modelling of electroactive molecules

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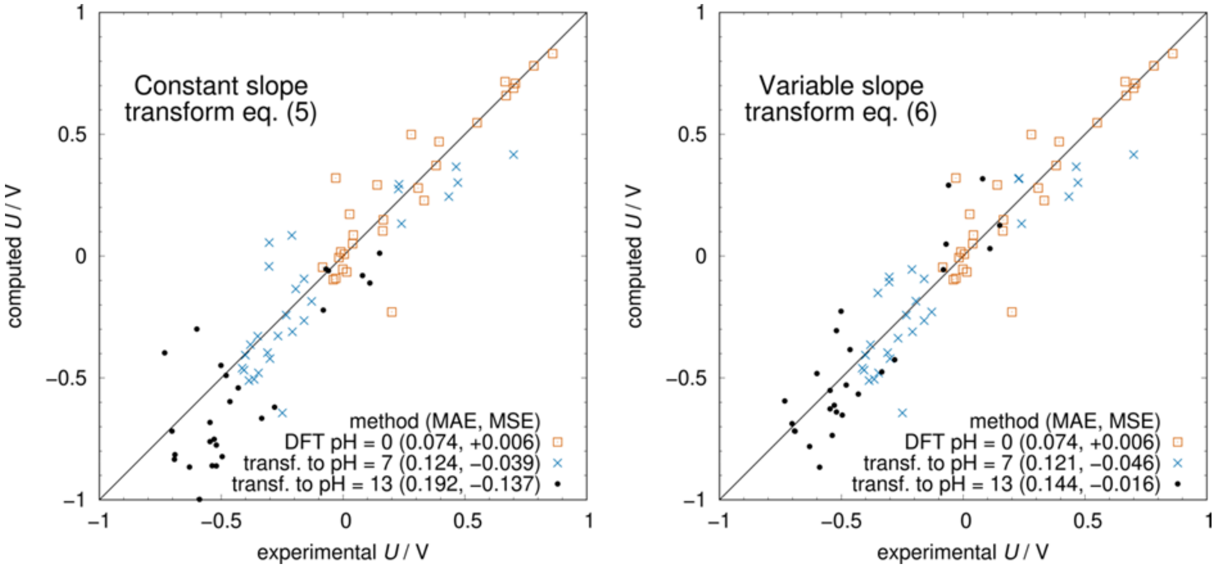
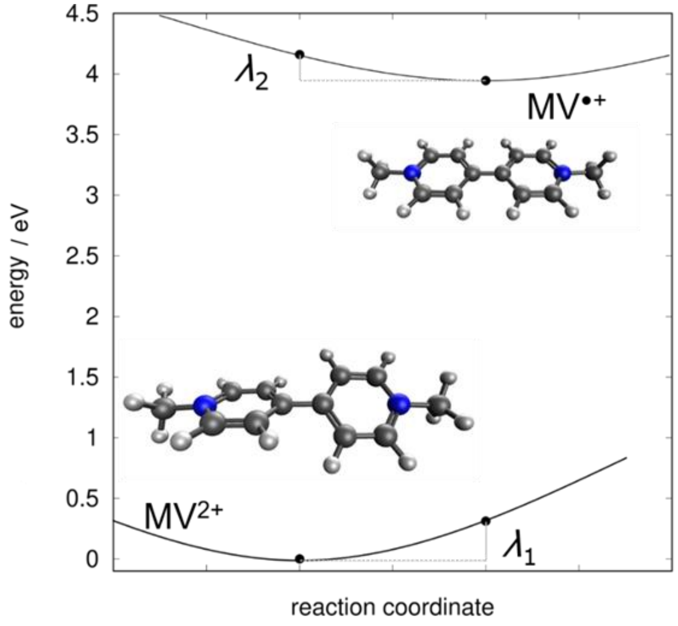
Computed vs. experimental redox potentials



Left, transformation from pH 0 to 7 and 13 is done using the number of protons at pH=0. Right, the slope of the Pourbaix diagram is updated at every pK_a.

Calculation of re-organisation energies



molecule	λ_1 / eV	λ_2 / eV	λ_i / eV
MV	0.228	0.297	0.263
EV	0.226	0.320	0.273
4-OH-TEMPO	0.496	0.462	0.479
AQS	1.485	1.601	1.543
BQDS	1.967	2.030	1.999

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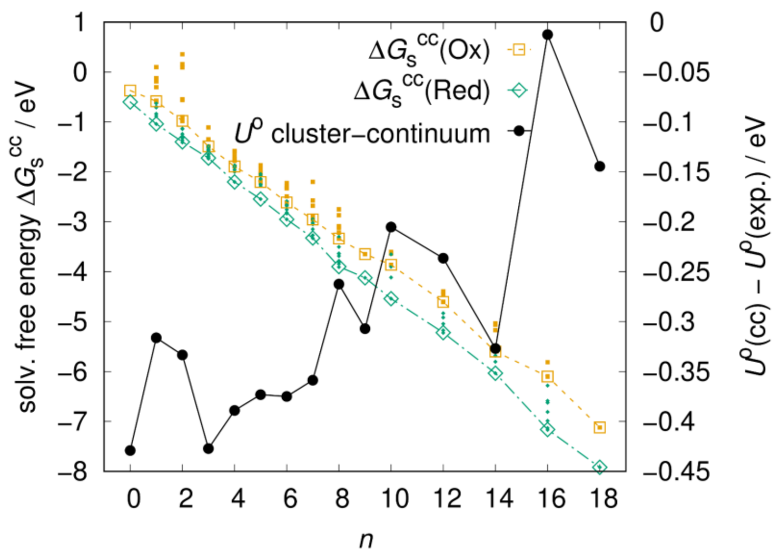
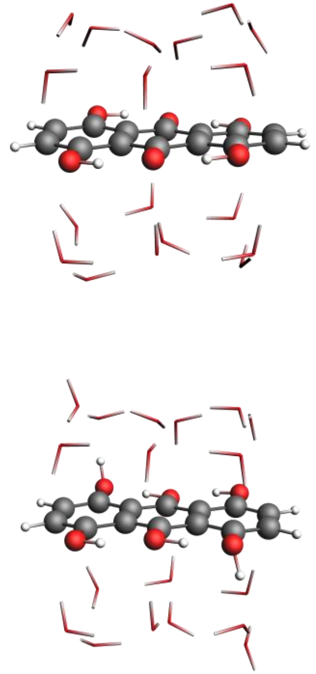
Electronic structure modelling of electroactive molecules

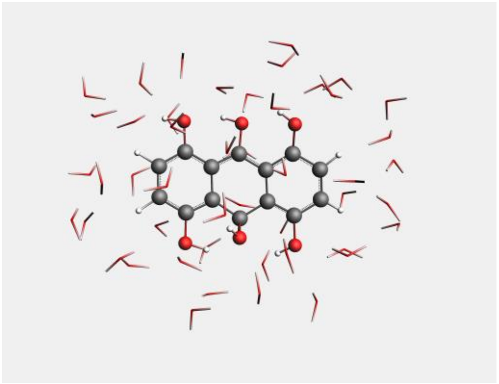
Beyond implicit solvation

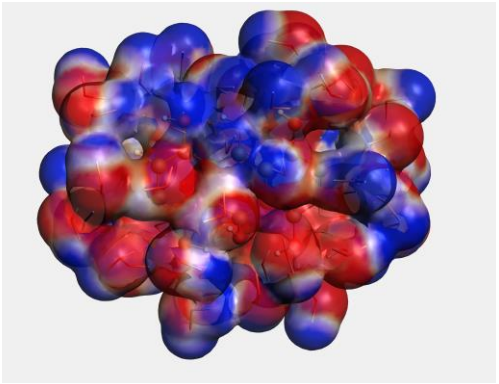




- each data point is averaged over multiple cluster conformations
- *n* = 18: we haven't reached a plateau in $\Delta G_{sol.}$.
- Potential getting closer to experiment
- Manual cluster construction has reached limit

Solute + cluster of 47 waters embedded in COSMO implicit solvation





1. MD simulation in a water droplet (using GFN-xTB semi-empirical method)
2. Extract uncorrelated snapshots
3. Extract cluster of water molecules corresponding to first solvation shell
4. Use cluster-continuum method to obtain solvation free energy and redox potential

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Meso-scale modelling of the electrochemical interface

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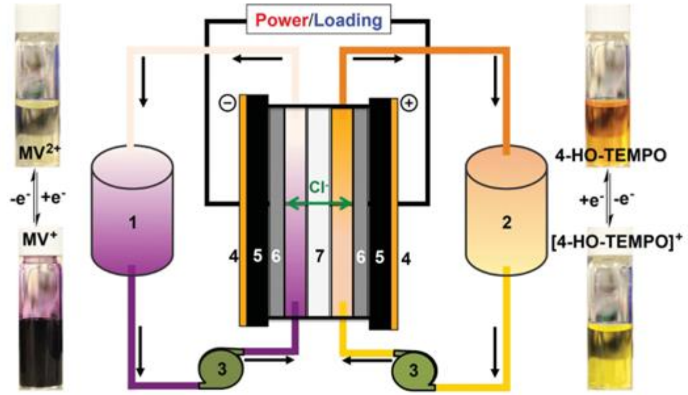
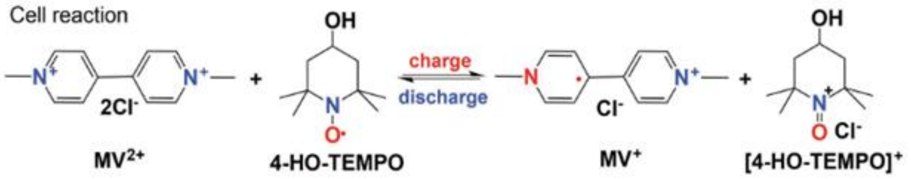
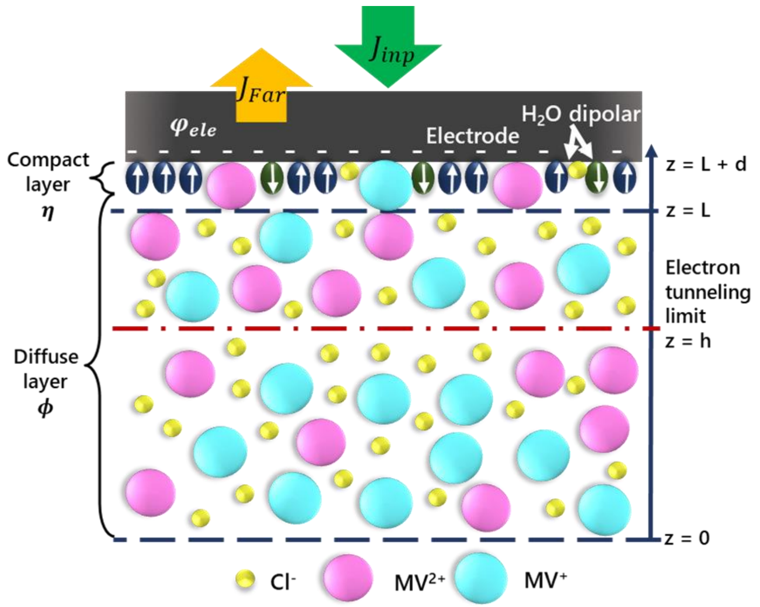


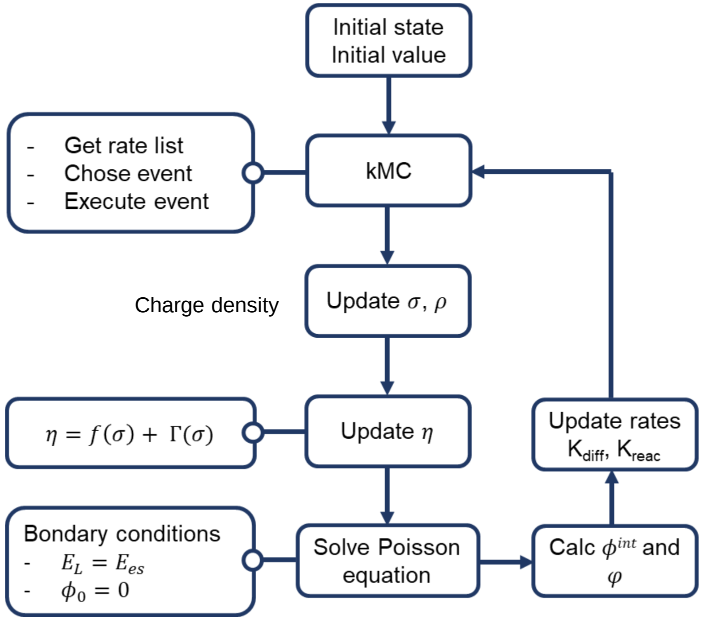


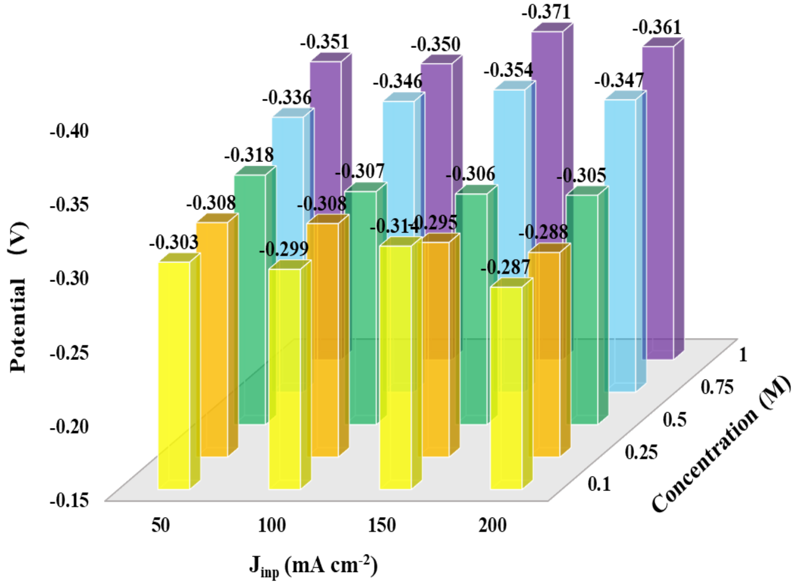
Consideration of

- Motion
- Electron transfer
- Adsorption/ desorption
- Dimerisation

-> Calculation of electrode potential φ







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Meso-scale modelling of the electrochemical interface

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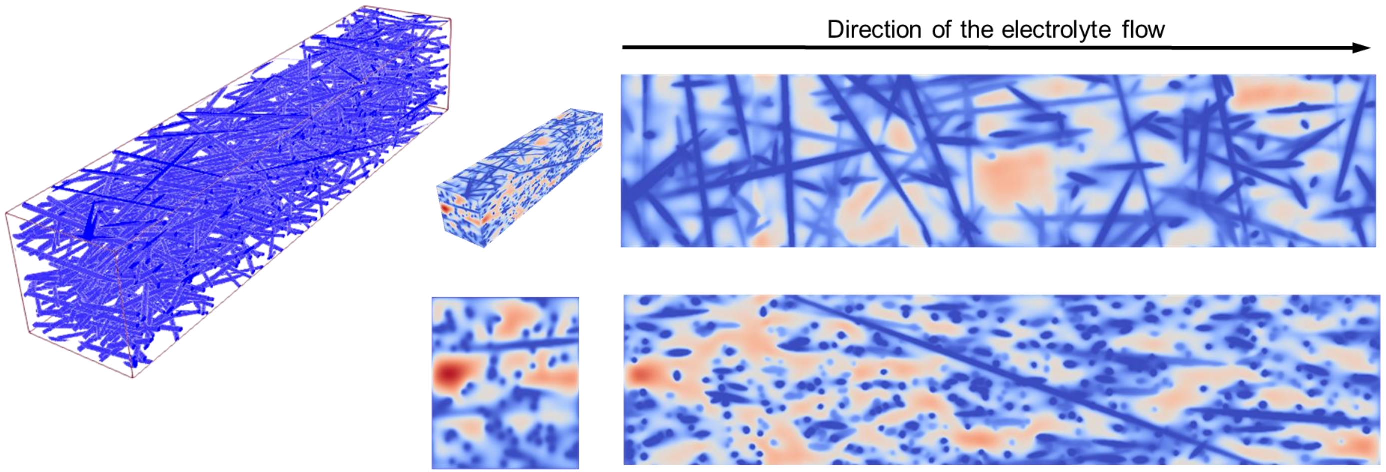
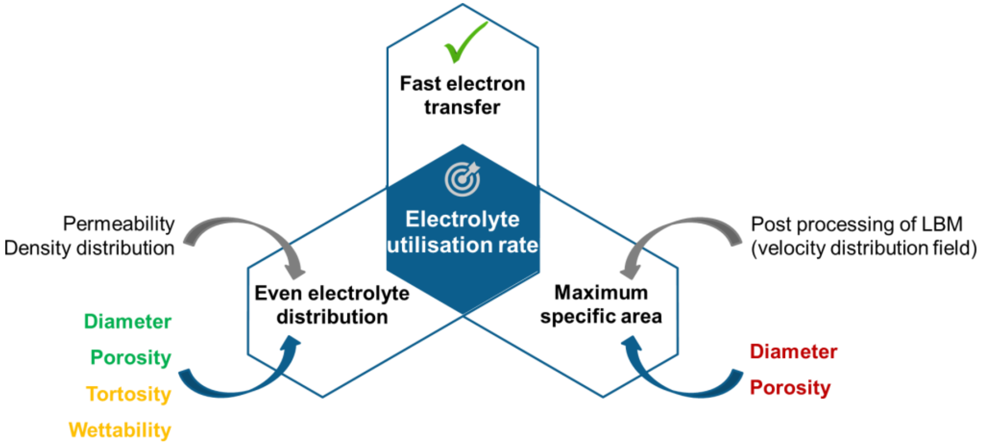
Lattice-Boltzman modelling

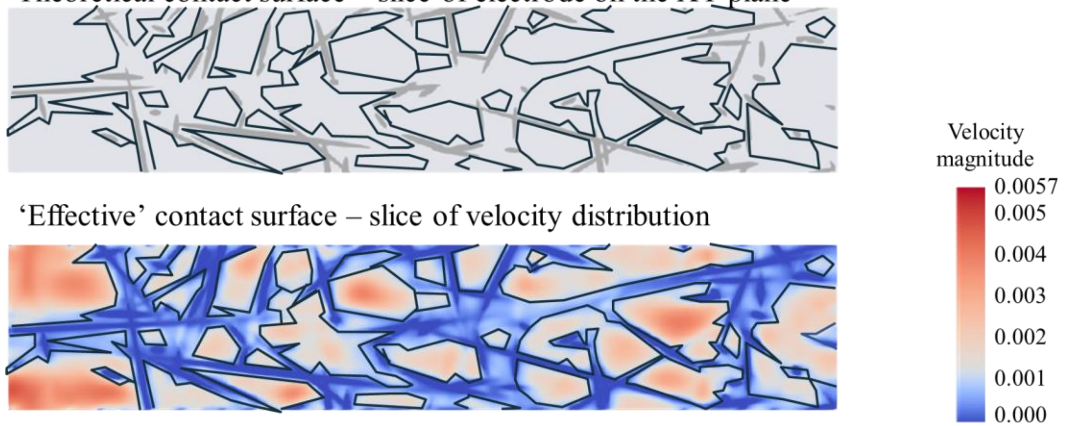
Velocity distribution of the electrolyte flow





GeoDict/ Tomography	• Carbon fiber; carbon foam; carbon paper electrode structure
LBM	• Simulate permeability of flow and the velocity distribution field
Post-processing	• Calculate theoretical/effective contact surface area (optimisation criteria)
Machine learning	• Optimise parameters of electrode structure (**diameter, shape orientation/tortuosity, porosity, wettability/contact angle…**)

Theoretical contact surface – slice of electrode on the XY plane



'Effective' contact surface – slice of velocity distribution

[Ref] R. Jervis, M. D. R. Kok, J. Montagut, J. T. Gostick, D. J. L. Brett, P. R. Shearing, Energy Technol. ***2018****, 6, 2488.*

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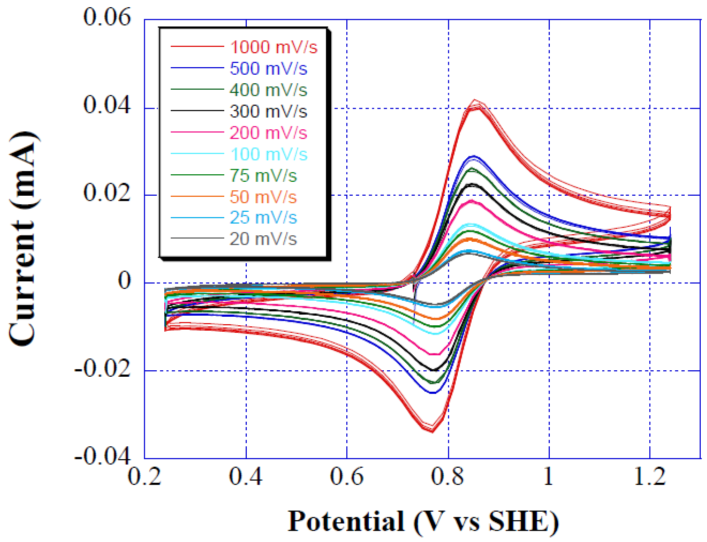
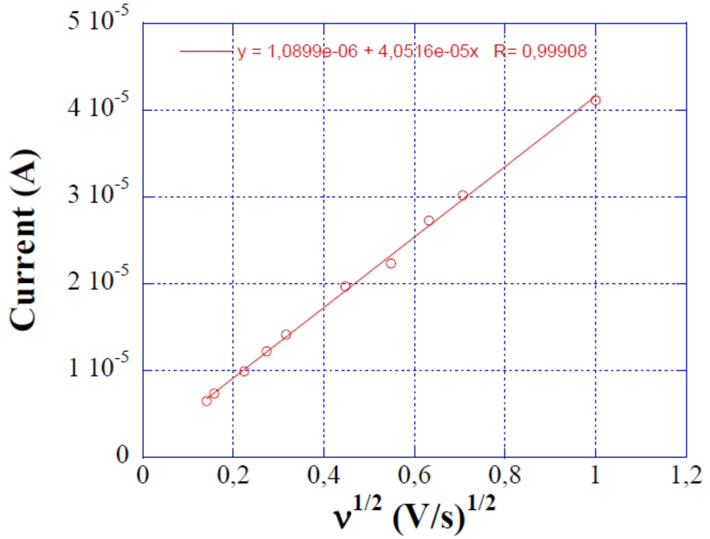
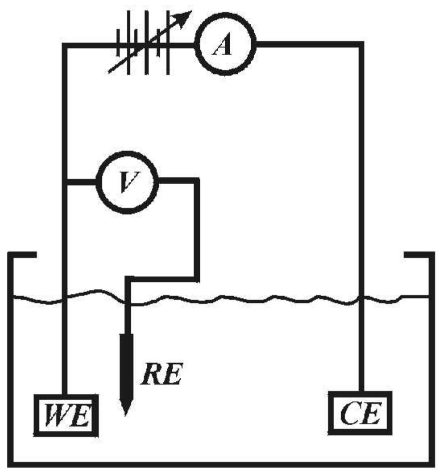
Experimental validation

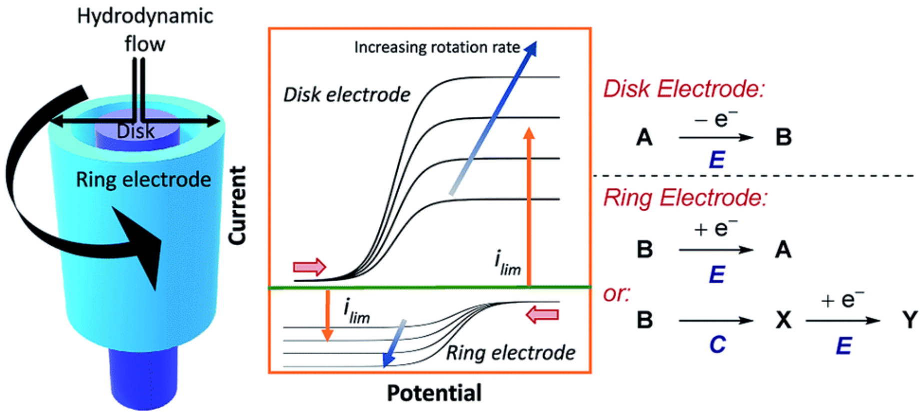
Example: Electrochemical properties of TEMPO-OH







Concentration (M)	D Randles-Sevcik ($cm^2.s^{-1}$)	E° ' (V vs SHE)	k° ($cm.s^{-1}$)	α
10^{-1}	$2.83.10^{-6}$	0.815	$2.96.10^{-3}$	0.34
10^{-2}	$3.93.10^{-6}$	0.807	$1.93.10^{-2}$	0.41
10^{-3}	$4.52.10^{-6}$	0.806	$4.47.10^{-2}$	0.36
10^{-4}	$4.46.10^{-6}$	0.,808	$2.42.10^{-2}$	0.37



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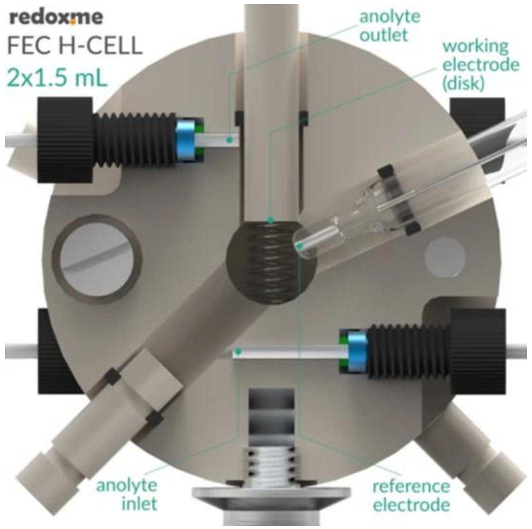
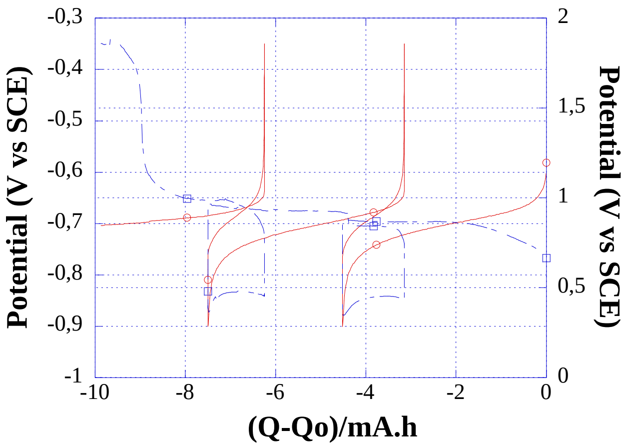
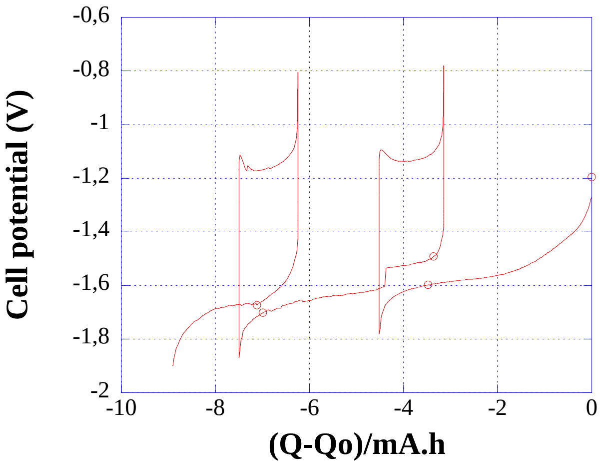
Experimental validation







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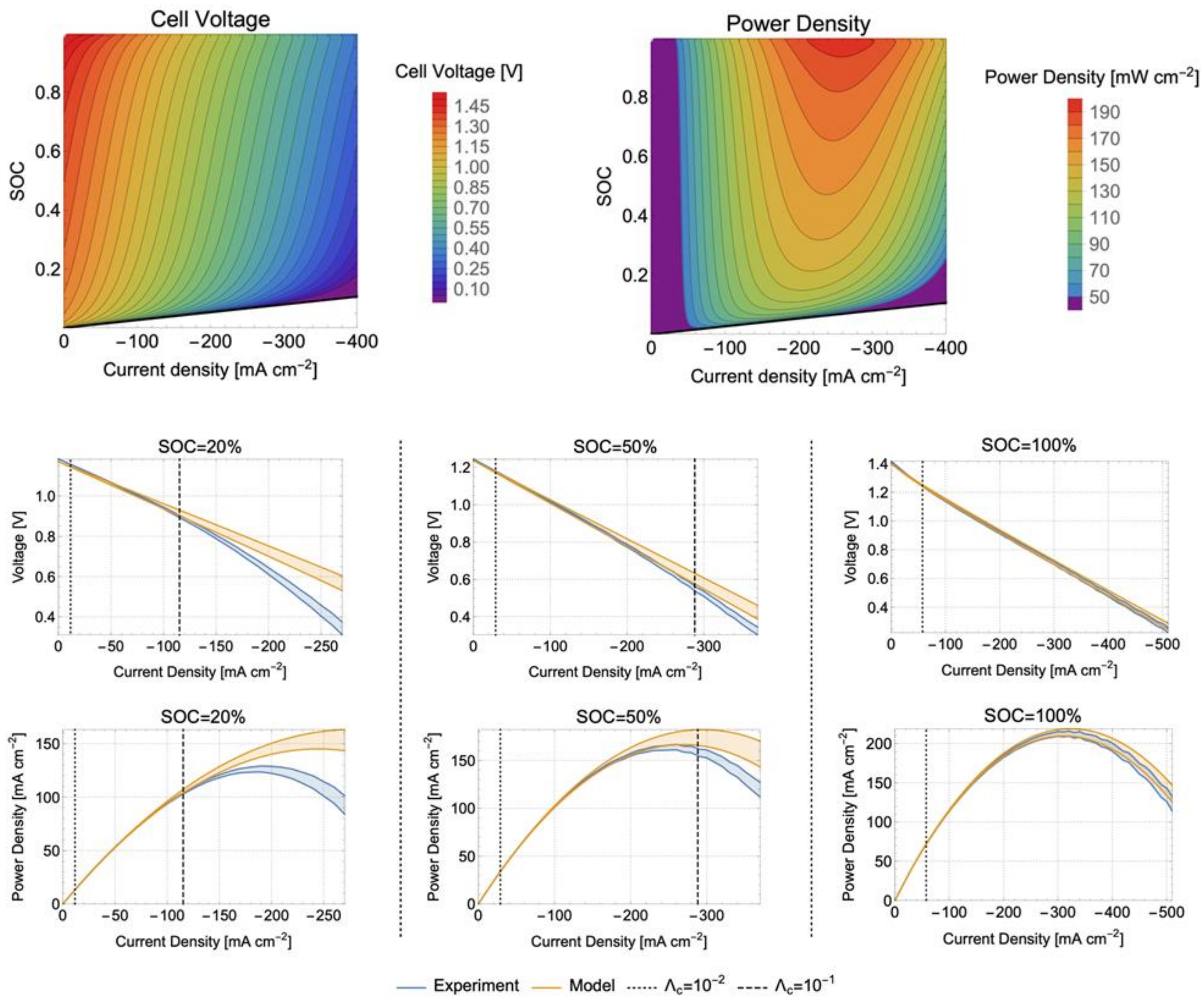
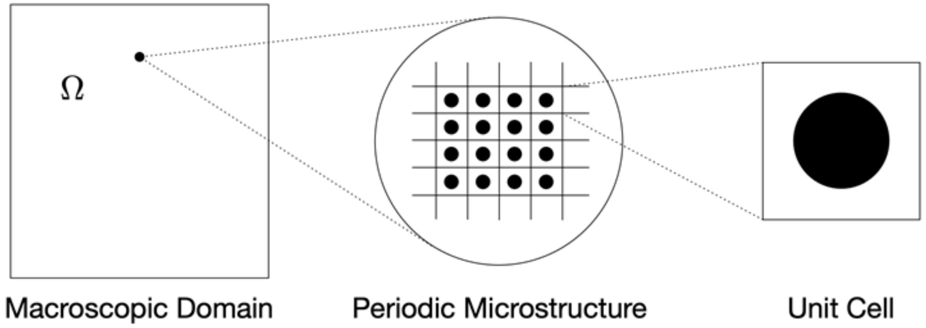
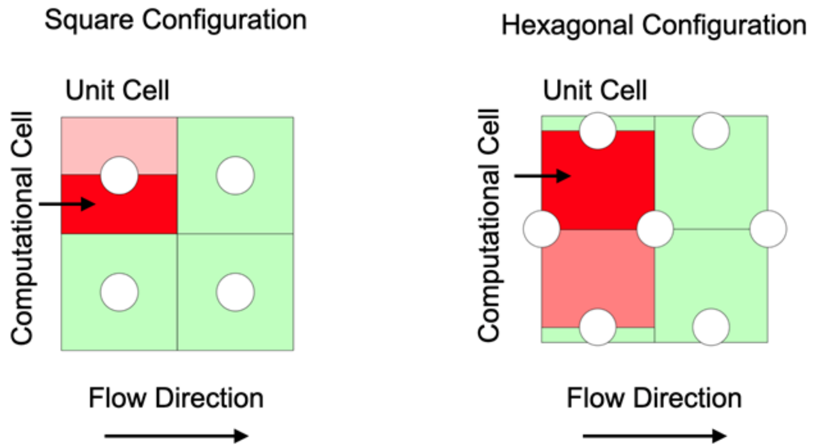
Bridging the scales: connection of electrochemical double layer properties, porous media flow and continuum modelling of RFBs

SONAR



- **Development of a 0D-U-I-SOC cell model**
- **Simulations based on MV/TMA-TEMPO**





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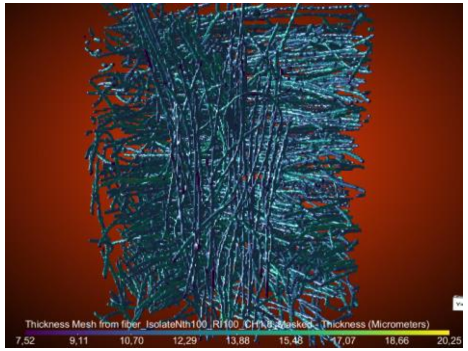
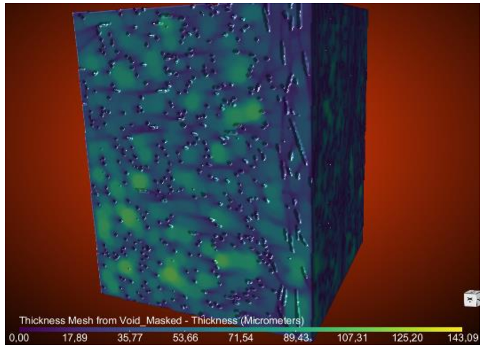
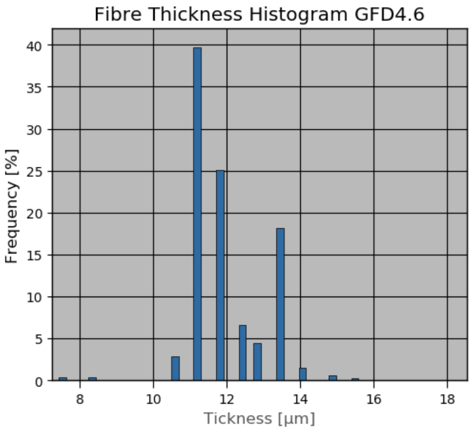
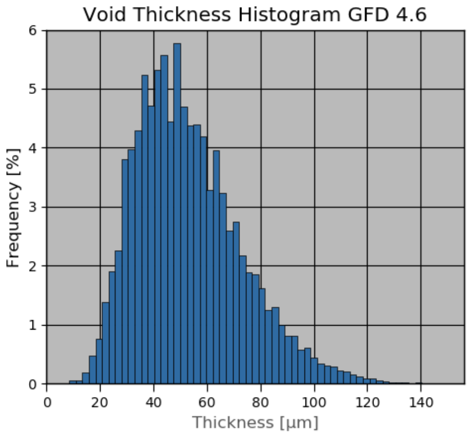
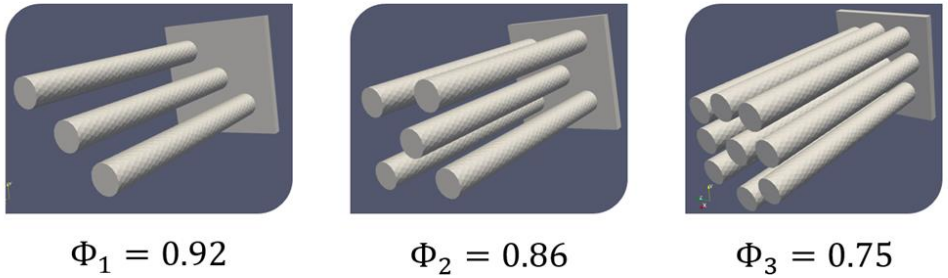
Micro Computer Tomography for electrode digitalisation

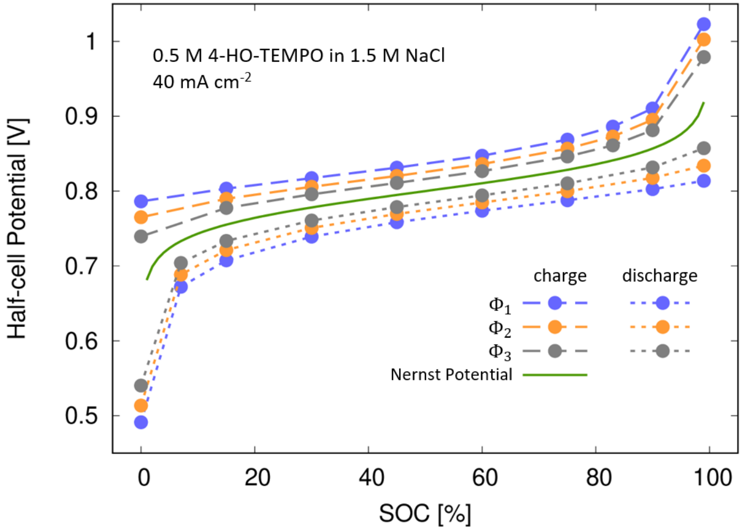












Different colours indicate different porosities. Dashed lines represent the charging process and dotted lines represent the discharging process. The simulations assume a constant supply of electrolyte

Cell performance simulation and cell design optimisation

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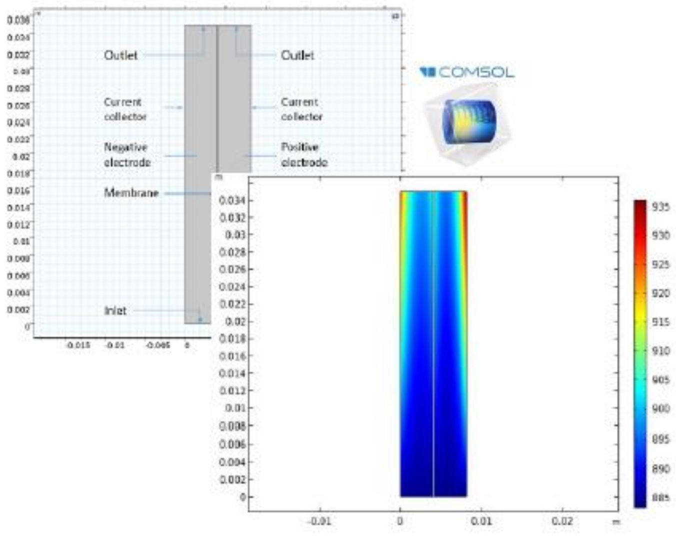
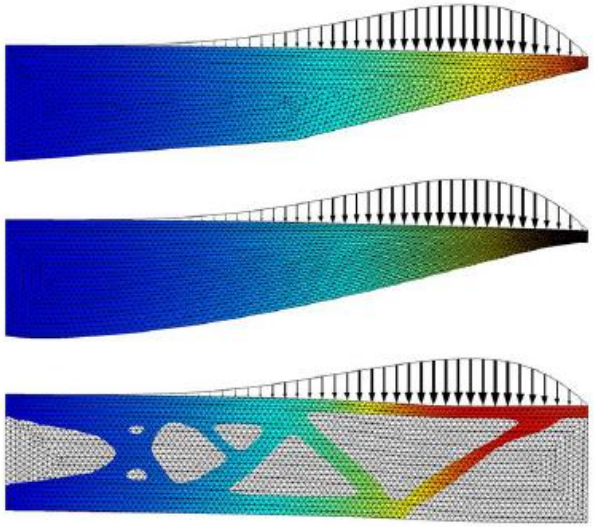
Flow cell design optimisation

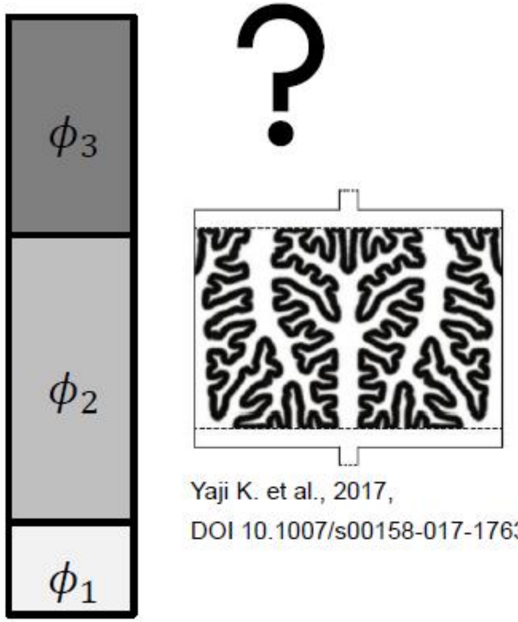
- **Aim: Finding an optimised cell design for intended operating conditions and desired redox pairs**



2D homogenized VRFB Model



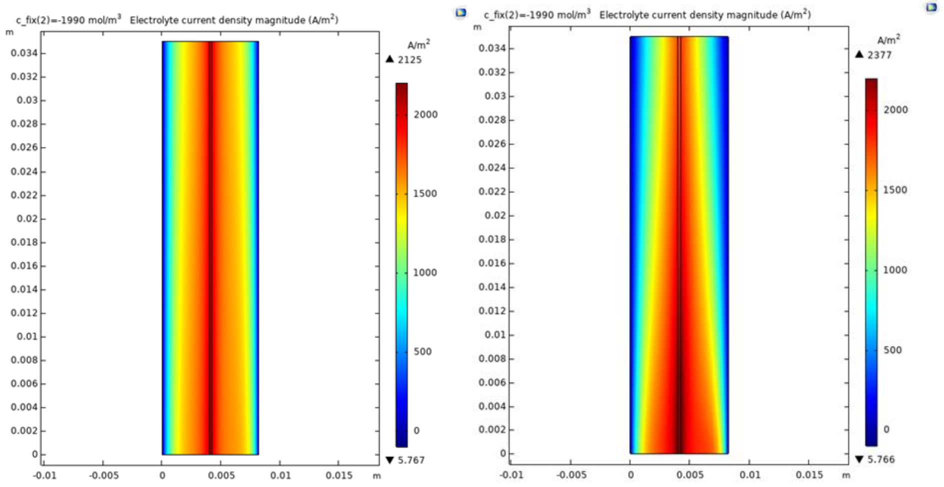
Optimisation Module



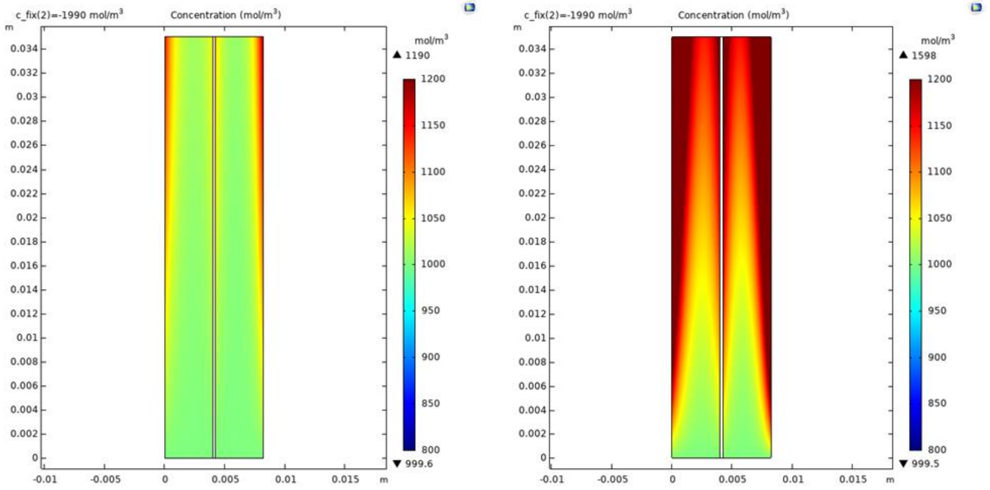
Yaji K. et al., 2017,
DOI 10.1007/s00158-017-1763-8

Optimal Design

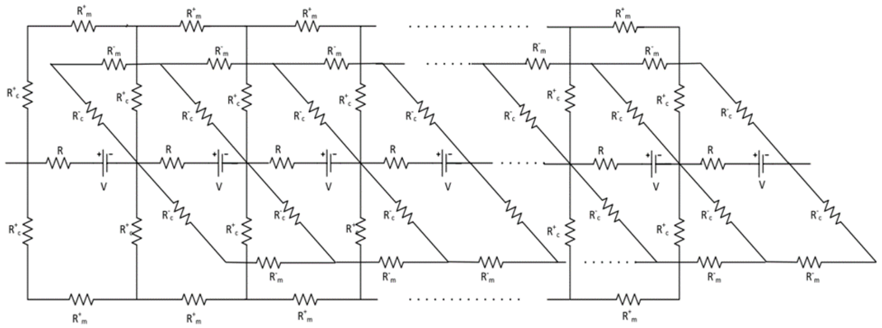
WP6 - Stack and system level modelling



Current distribution of the 2D single cell of VRFB under different constant flow rates, left) flowrate = 30 mL/min right) flow rate = 5 mL/min



V(III) / V(IV) Species concentration of the 2D single cell of VRFB under different flow rate left) flow rate = 30 mL/min right) flow rate = 5 mL/min



Shunt current resistor networks for stack modelling

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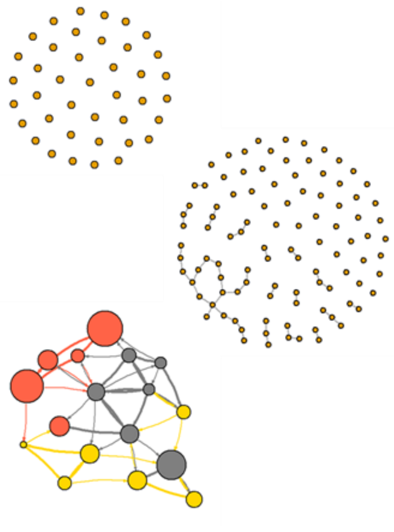
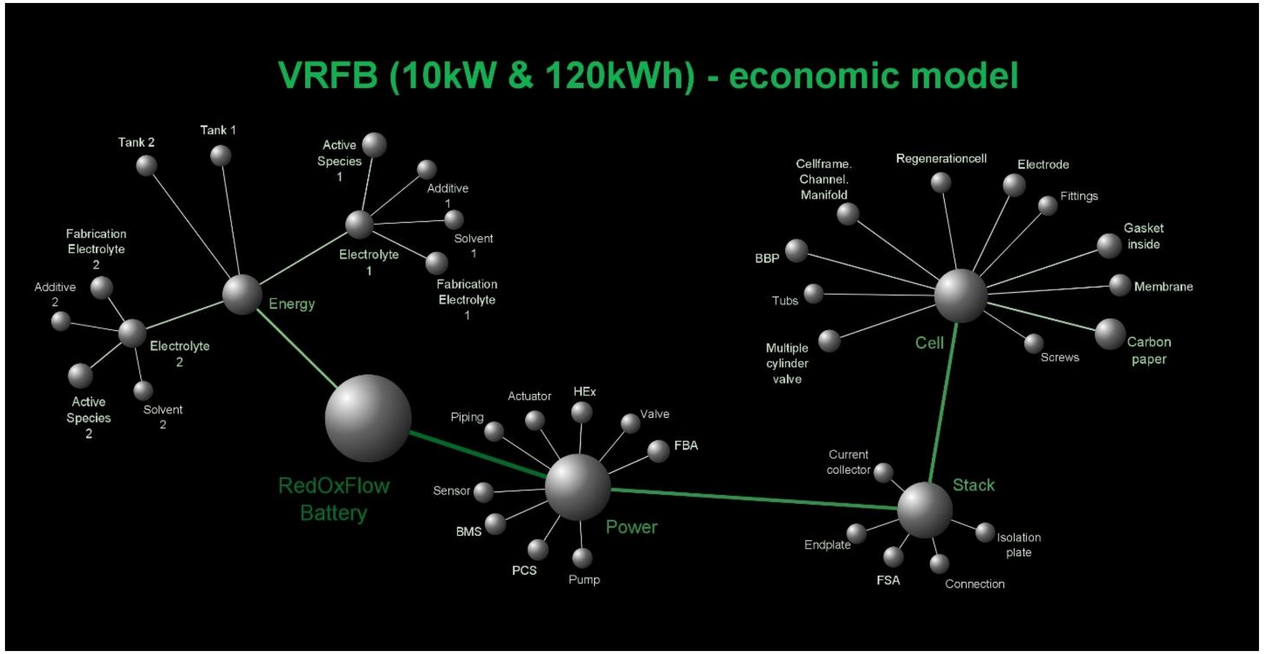
Cost analysis of redox flow batteries

- Step 1 knots: Identify components
- Step 2 edges: Find dependences (physico-chemical & economical ones)
- Step 3 weight: Measure the influences





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Cost analysis of redox flow batteries

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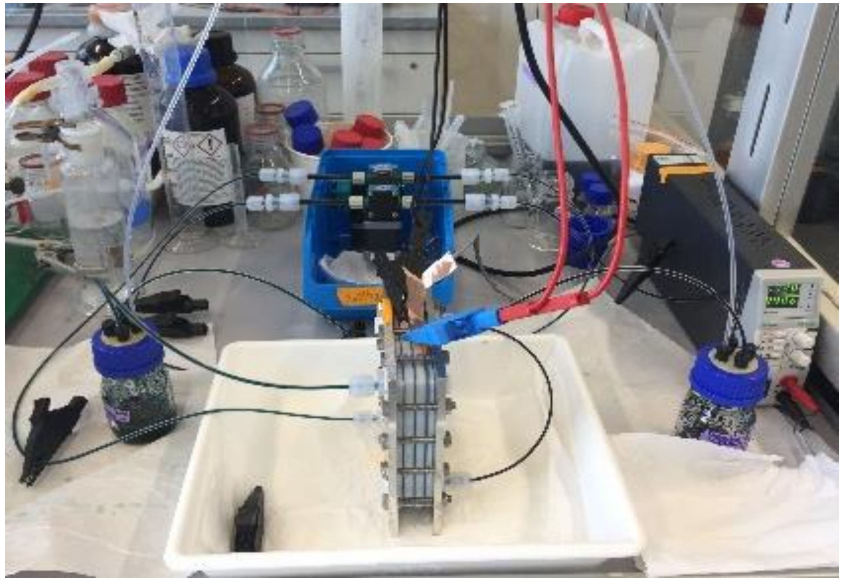
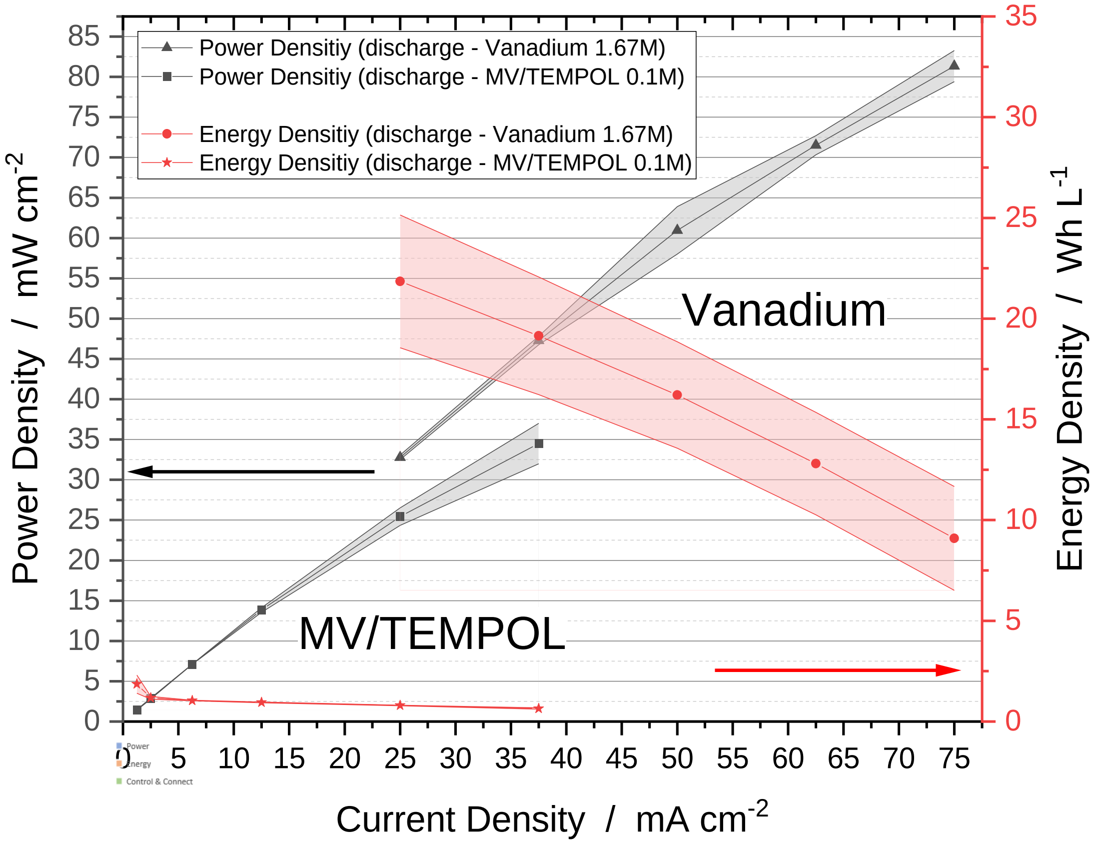
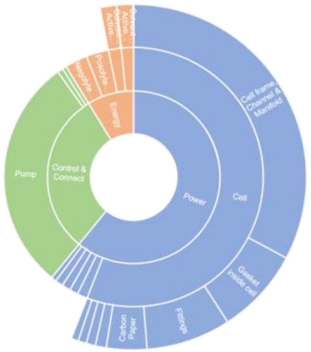


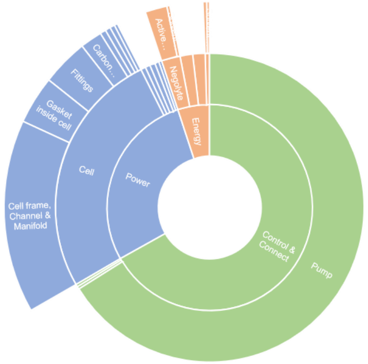
Values from laboratory scale flow batteries as starting point



Coststructure V 1,67 M



Coststructure MV/TEMPOL 0,1 M



Coststructure MV/TEMPOL 0,5 M

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Framework for high-volume pre-selection, data integration and design

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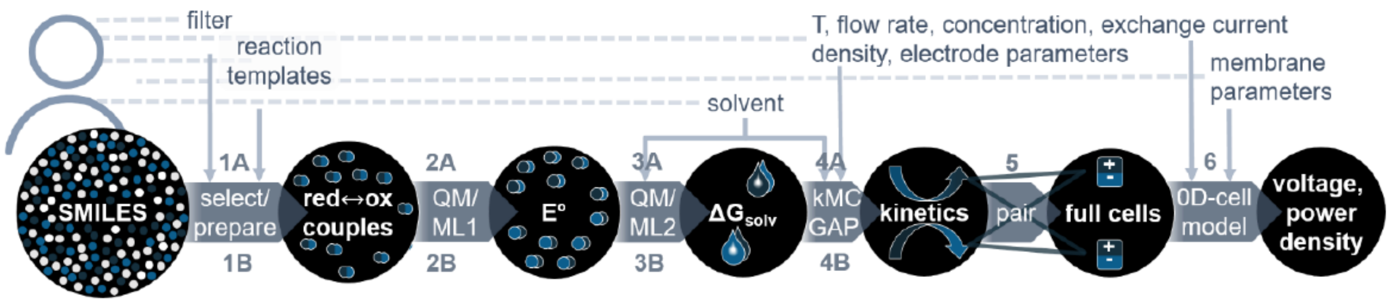
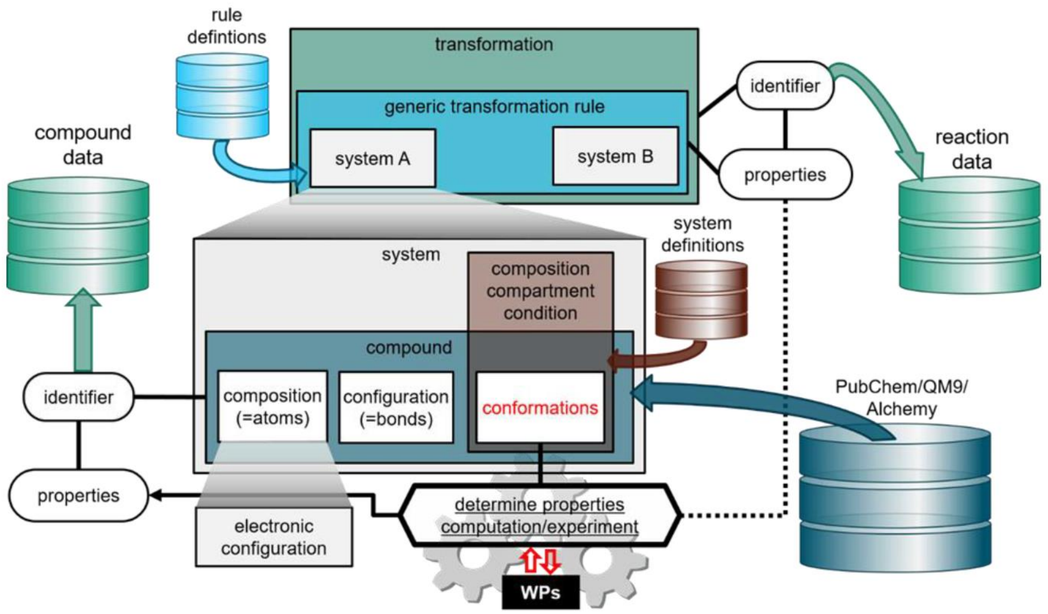
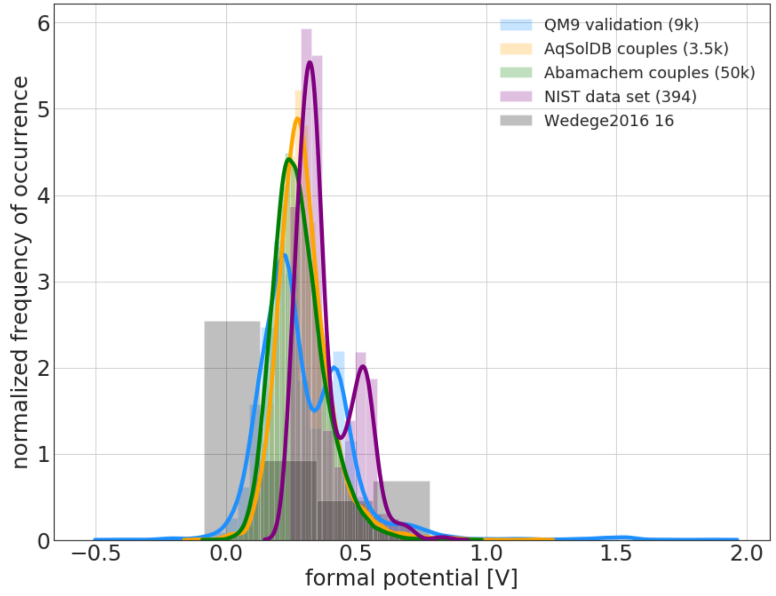
Data flow



Data management



Calculated formal potentials vs. databases



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REDOX FOX - Prototype!

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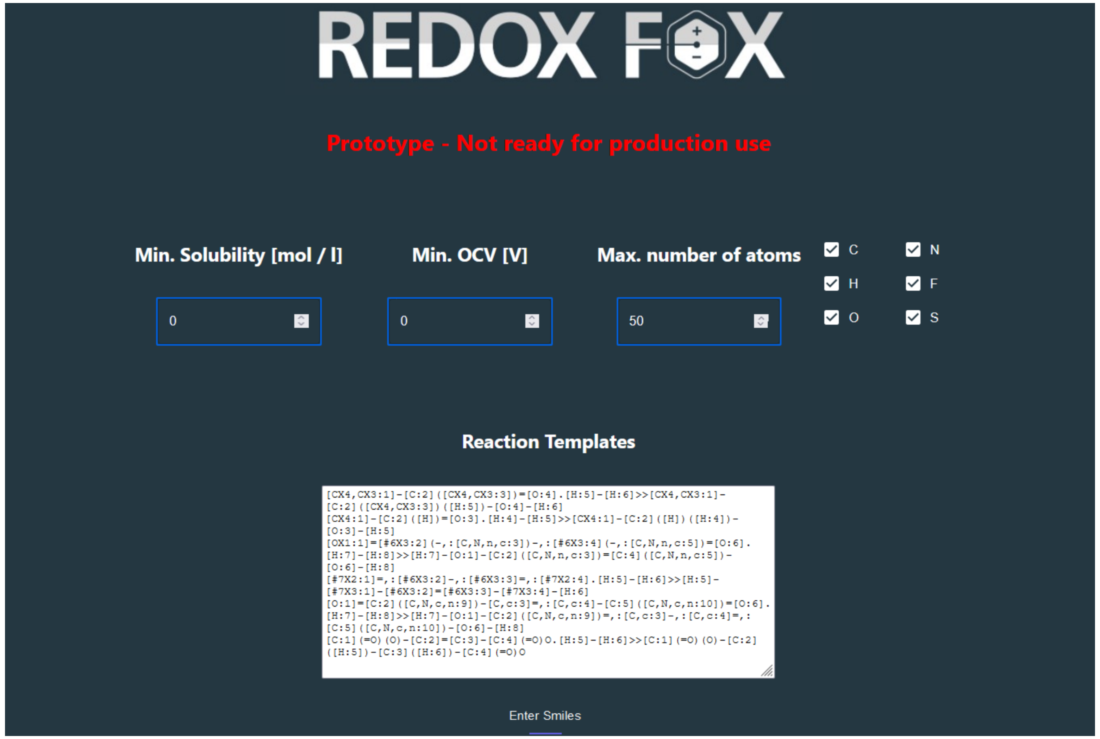

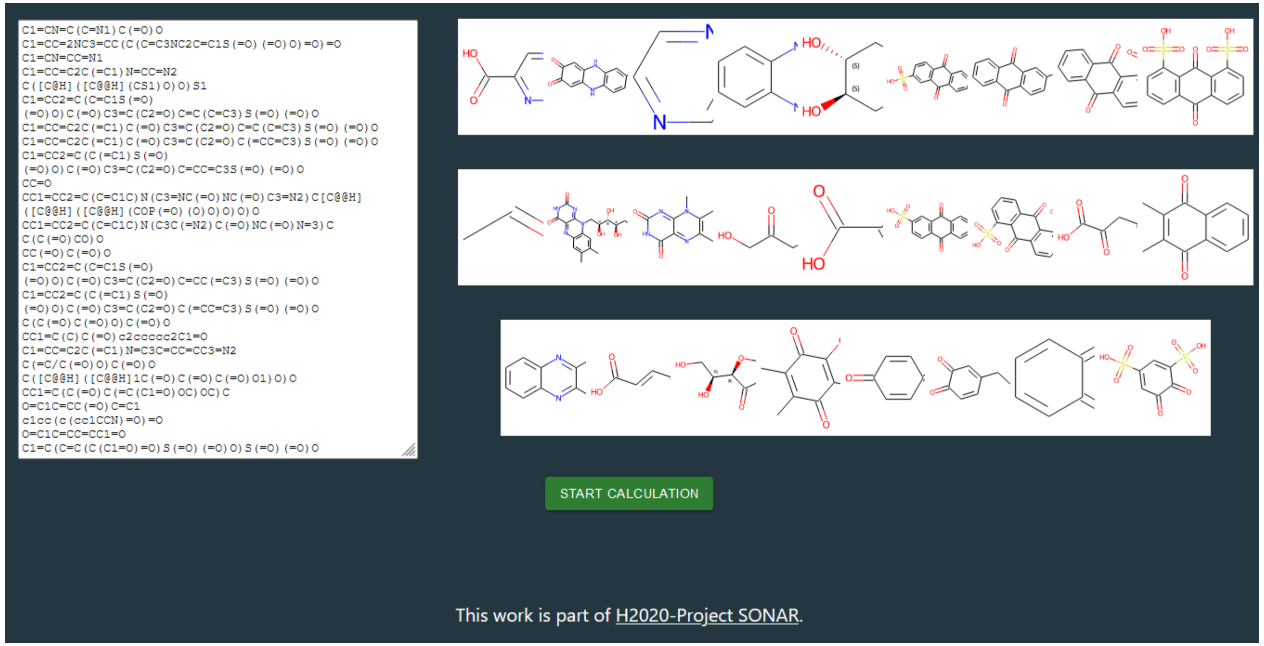

https://redoxfox.scai.fraunhofer.de/

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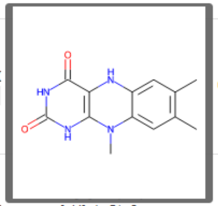
PREVIOUS PAGE 1/51 | NEXT PAGE 1/51

ID	Posolyt Smiles Ox	Posolyt Smiles Red	Negolyt Smiles Ox	Negolyt Smiles Red	OCV	Posolyt SAScore Ox	Posolyt Solubility Ox	Posolyt SAScore Red	Posolyt Solubility Red	Posolyt Std. Reduction Potential	Negolyt SAScore Ox	Negolyt Solubility Ox	Negolyt SAScore Red	Negolyt Solubility Red	Negolyt Std. Reduction Potential
0	O=C1C=CC=CC1=O	[H]OC1=C(O[H])C=CC=(	CC1=CC2=C(C=C1C)N((	[H]n1c(=O) [nH]c(=O)c2=c-1n(C)c1cc(C)c(C)cc1n-2[	0.6446	2.5009	0.1272	3.9228	1.1062	-0.0912	2.5356	0.0005	3.9033	0.0007	-0.7358
1	O=C1C=CC(=O)C=C1	[H]OC1([H])C=CC(=O)C:	CC1=CC2=C(C=C1C)N((		0.6301	2.3547	0.1263	4.9152	0.4608	-0.1058	2.5356	0.0005	3.9033	0.0007	-0.7358
2	C1=CN=C(C=N1)C(=O)C	[H]n1ccn([H])C(C(=O)O):	CC1=CC2=C(C=C1C)N((	[nH]c(=O)c2=c-1n(C)c1cc(C)c(C)cc1n-2[	0.6250	2.0292	0.2524	5.1466	0.2820	-0.1109	2.5356	0.0005	3.9033	0.0007	-0.7358
3	C(=C/C(=O)O)C(=O)O	[H]C(C(=O)O)C([H])C(=C	CC1=CC2=C(C=C1C)N((	[H]n1c(=O) [nH]c(=O)c2=c-1n(C)c1cc(C)c(C)cc1n-2[	0.6230	2.1106	0.1273	5.2846	0.4051	-0.1129	2.5356	0.0005	3.9033	0.0007	-0.7358
4	C1=CN=CC=N1	[H]n1ccn([H])C=C1	CC1=CC2=C(C=C1C)N((	[H]n1c(=O) [nH]c(=O)c2=c-1n(C)c1cc(C)c(C)cc1n-2[	0.5914	1.9736	3.5755	6.0525	3.9600	-0.1444	2.5356	0.0005	3.9033	0.0007	-0.7358
ID	Posolyt Smiles Ox	Posolyt Smiles Red	Negolyt Smiles Ox	Negolyt Smiles Red	OCV	Posolyt SAScore Ox	Posolyt Solubility Ox	Posolyt SAScore Red	Posolyt Solubility Red	Posolyt Std. Reduction Potential	Negolyt SAScore Ox	Negolyt SAScore Ox	Negolyt SAScore Red	Negolyt Solubility Red	Negolyt Std. Reduction Potential



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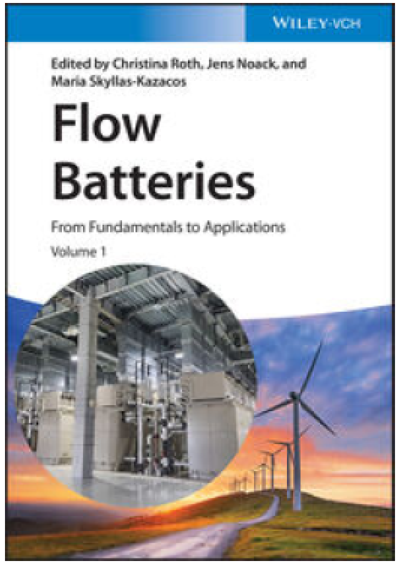

Flow Batteries: From Fundamentals to Applications, 2 Volume Set

Christina Roth (Editor), Jens Noack (Editor), Maria Skyllas-Kazacos (Editor)

Hardcover 978-3-527-34922-7 December 2022

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Thank you for your attention!

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