Computational screening of organic molecules for flow battery applications

Pekka Peljo, Materials Engineering, University of Turku

pekka.peljo@utu.fi

UNIVERSITY OF TURKU

compbat

Towards high-throughput screening in CompBat

Imre Papai

UPPSALA UNIVERSITET
UNIVERSITÀ DI PISA
Aalto University School of Chemical Engineering
JYVÄSKYLÄN YLIOPISTO UNIVERSITY OF JYVÄSKYLÄ
ttk
UNIVERSITY OF TURKU
Skoltech Skolkovo Institute of Science and Technology

Molecular database

Computational protocol

a) **initial 3D structures** – automatic generation of 2D (Lewis) structures (***CombiGlide***), conversion to 3D (***LigPrep***); protonation state at pH = 7 in water (***Epic***); preliminary conformational search with OPLS force field (***Macromodel***)

b) **conformational analysis** – geometry optimization at semiempirical GFN2-xTB level (***xtb*** package); finite temperature and implicit solvent effects (room temperature, water); CREST procedure for extensive conformational search (***crest*** utility) → most stable conformers

c) **electronic energies** – DFT calculations at M06-2X/6-311+G** level (***Gaussian***), single-point calculations in gas- and solution phase (SMD) → aqueous phase solvation free energies

d) **Gibbs free energies** – $G^{aq}_{red/oxid} = E^{DFT}_{red/oxid} + \Delta G^{xtb}$ → redox potentials according to the Nernst formula

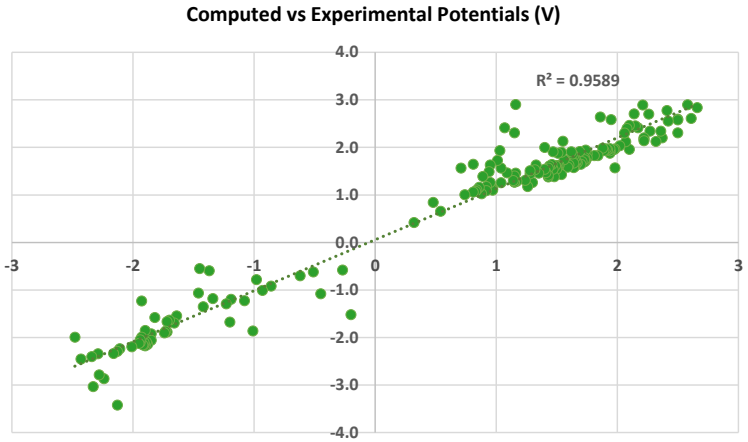
Composite method – force field + semiempirical + accurate QC

Software packages: *Schrödinger*, *xtb*, *Gaussian* + own scripts

Molecular database

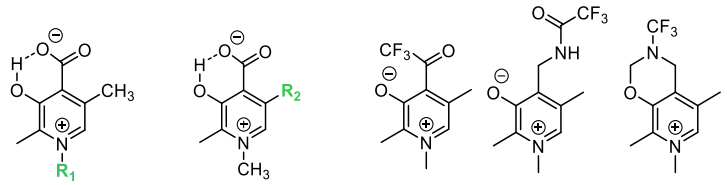
Test and benchmark calculations

a) Tests wrt experimental data



188 organic molecules (CV data), MAE = 0.24 V

b) Benchmarks wrt full DFT data



R_1 = P1-P8

R_2 = b1,c1,d1, e1,f1,g1,h1

MAE: 0.07 V 0.07 V 0.09 V

18 molecules from our database, MAE < 0.1 V

Molecular database

Pyridoxal databases

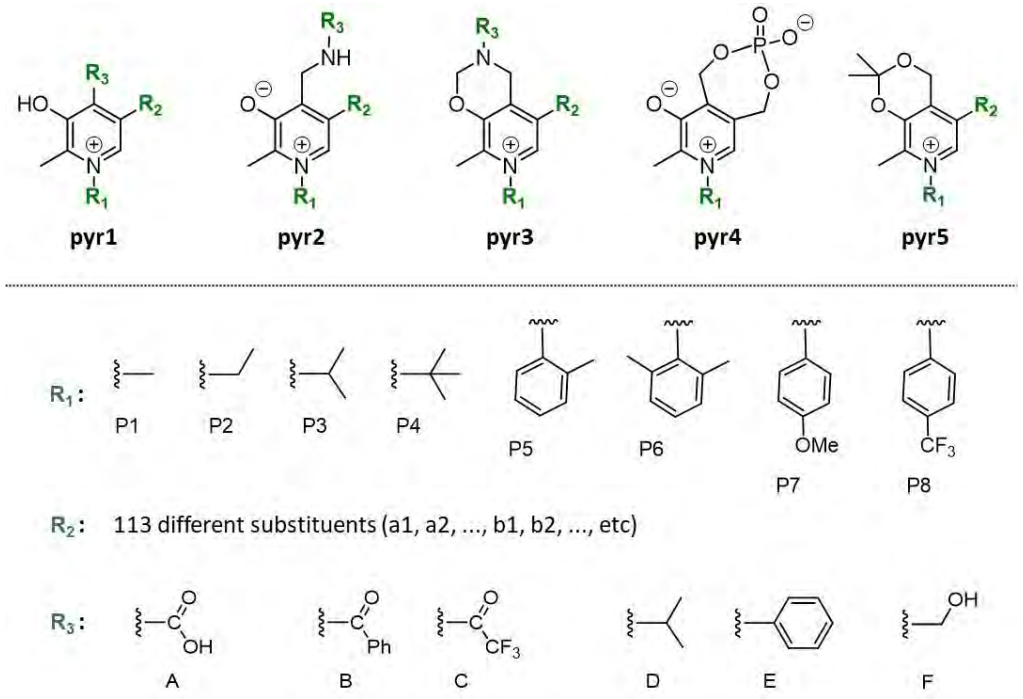
Latest database

- five pyridoxal frameworks (suggested by JYU)
- R_1, R_2 and R_3 substituents

↓

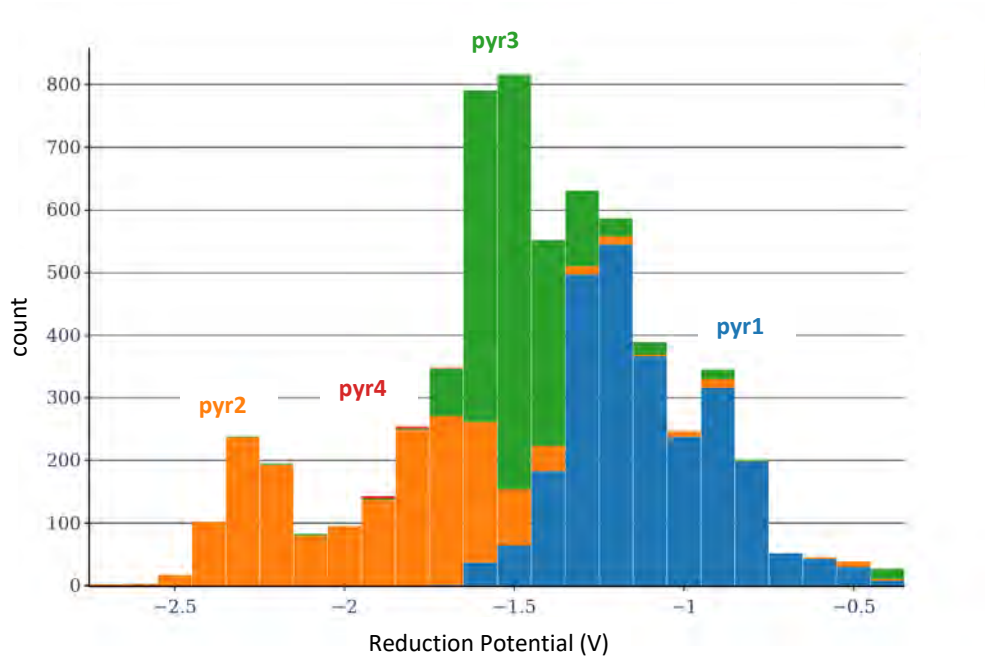
6712 molecules

(for some molecules "reactions" are observed)



Molecular database

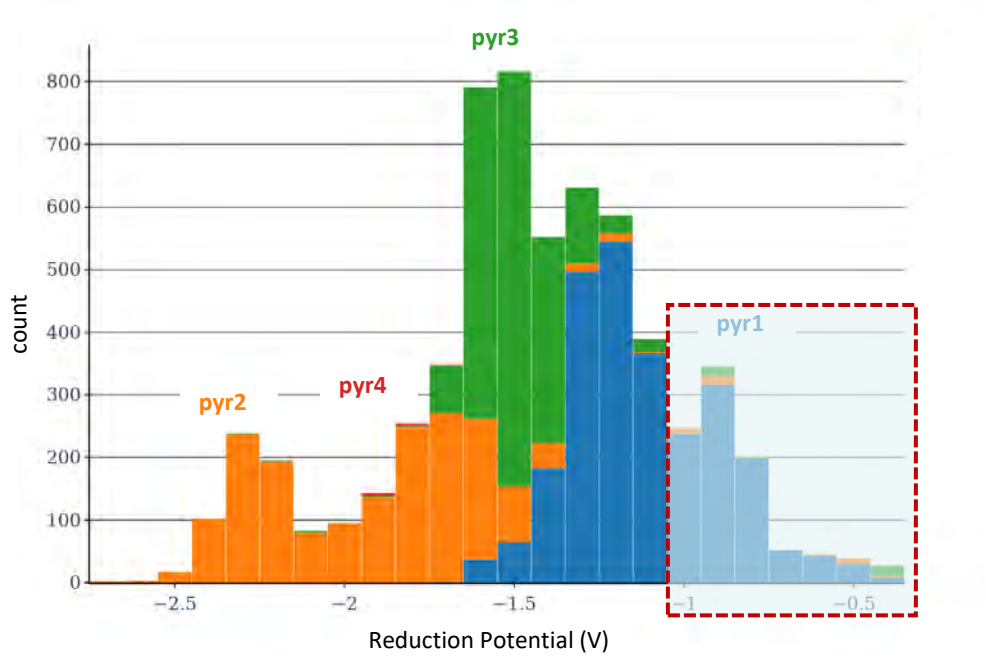
Reduction potentials

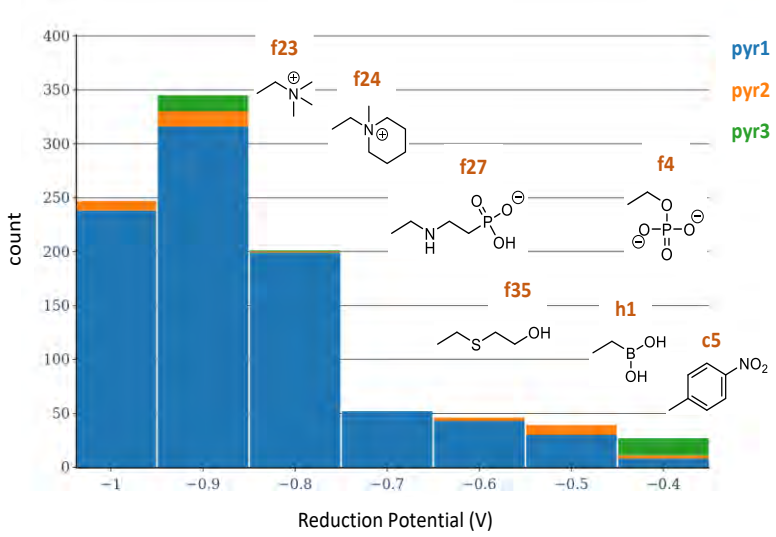


electrochemically relevant range (above -1 V) has lower populations, where almost all compounds are from pyr1

Molecular database

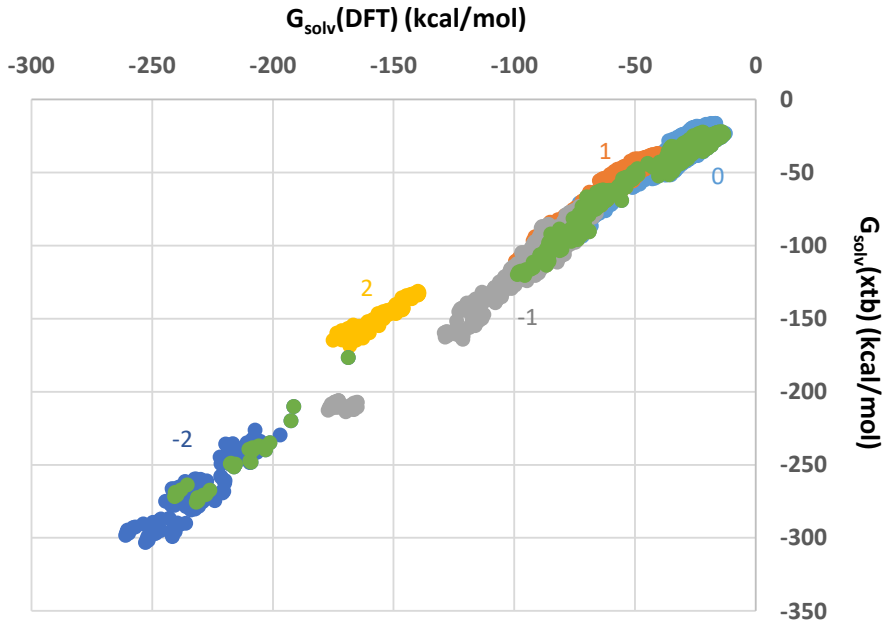
Reduction potentials





Molecular database

Solvation free energy in aqueous solutions

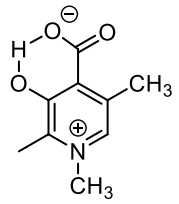


xTB versus DFT:
reasonably good correlation
→ xtb can be efficiently used

Machine learning

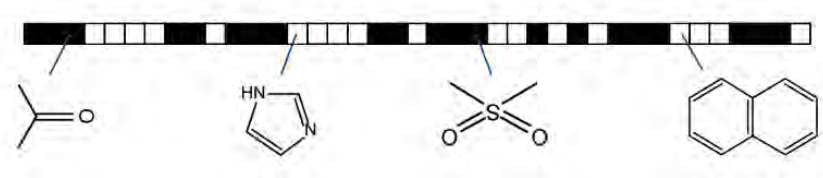
Molecular representations

a) SMILES strings



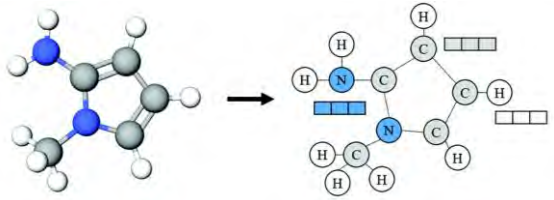
Cc1c[n+](C)c(C)c(O)c1C(=O)[O-] (atoms, bonds, connectivity)

b) Molecular fingerprints



(presence or absence of substructures)

c) Graph representation



(atoms = nodes, bonds = edges each node has a feature vector)

Machine learning

Machine learning methods

a) traditional ML methods (Aalto)

- Random Forest analysis followed by SHAP feature attribution analysis

b) deep-learning methods (TTK)

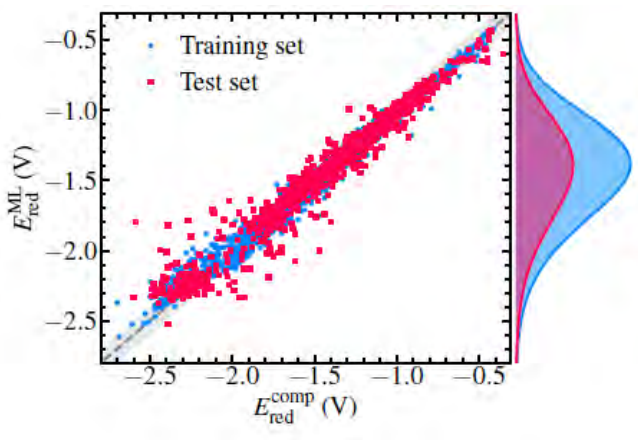
- convolutional neural networks (CNN)
- extract higher-level features from the raw input
- ***DeepChem*** ML library via the ***Google Colab*** service
- collaboration with **Budapest University of Technology**
- 3D graph convolutional network (3DGCN)

Machine learning

RF analysis

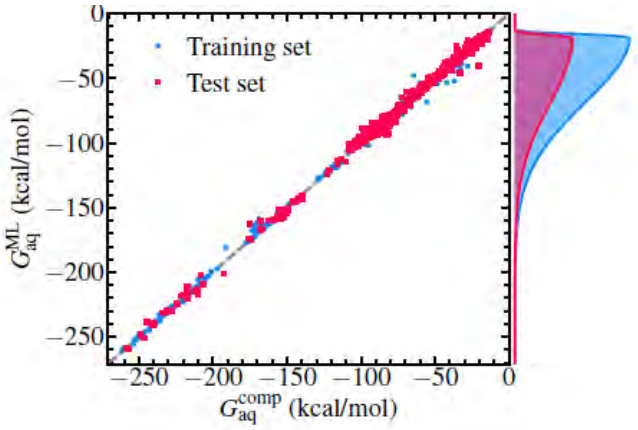
ECFP fingerprints
training/test 80% + 20%

potentials



MAE = 0.05±0.01 V
(smaller than the intrinsic error)

solubilities



MAE = 1.3±0.1 kcal/mol
(very accurate predictions)

Machine learning

Results

Performance of 3 different DeepChem methods for reduction potentials (DB-II)

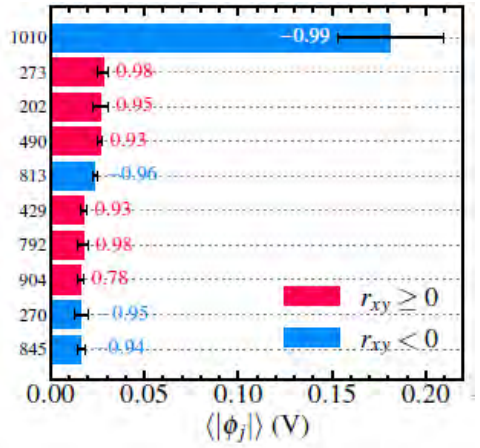
Model	MAE Te	RMSE Tr + V + Te	R^2 Tr + V + Te
TextCNNModel	0.065 ± 0.005	0.062 ± 0.005	0.984 ± 0.001
RobustMultiRegressor	0.061 ± 0.002	0.063 ± 0.006	0.981 ± 0.0003
AttentiveFPModel	0.065 ± 0.006	0.08 ± 0.006	0.972 ± 0.004

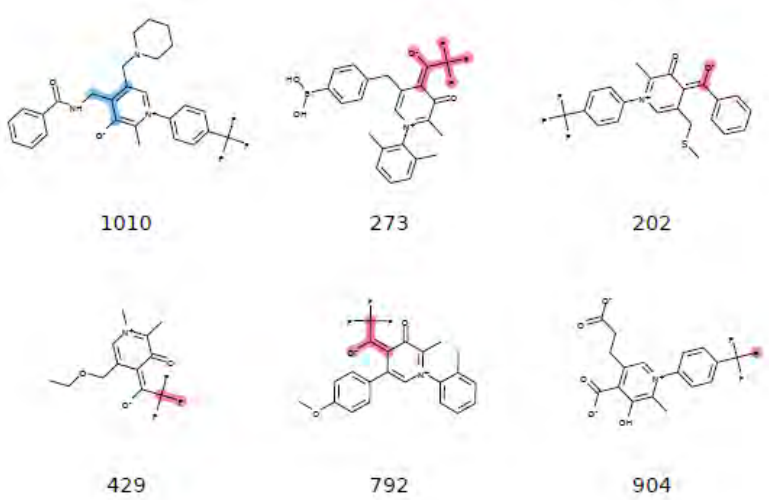
(**MAE**: mean absolute error; **RMSE**: root mean square error; **R^2**: coefficient of determination; **Tr**, **V** and **Te**: training, validation and test sets, 80% + 10% + 10%)

→ **good performance, but not better than RF (surprising!)**

Machine learning

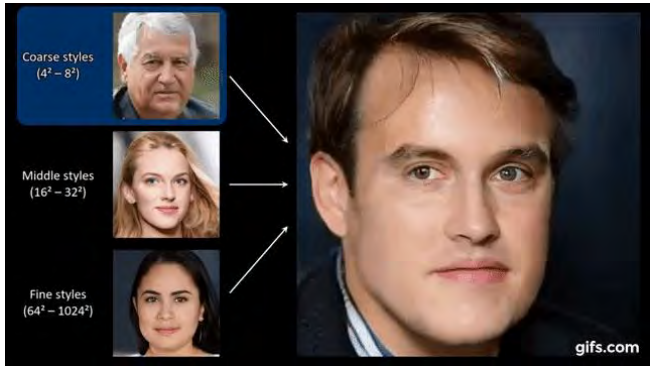
Feature attribution analysis





contribution of different molecular substructures: impact of electron-withdrawing and donating groups

Is ML a hype?



human faces generated by the GAN

Participants

Contributors:

Imre Pápai

Andrea Hamza

Ádám Madarász

Flóra Németh

Kari Laasonen

Arsalan Hashemi

Rasmus Kronberg

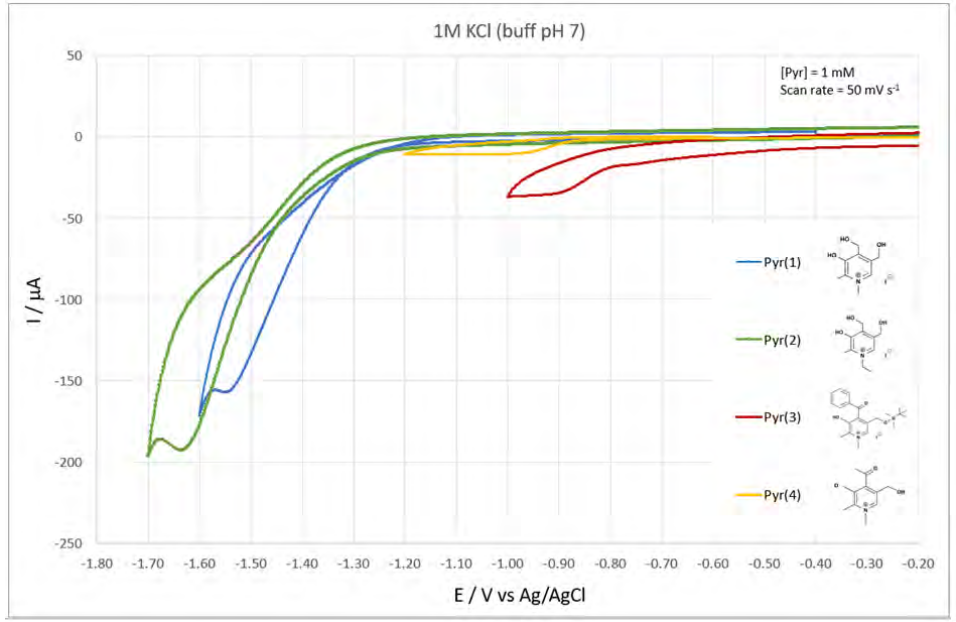
Organic aqueous batteries

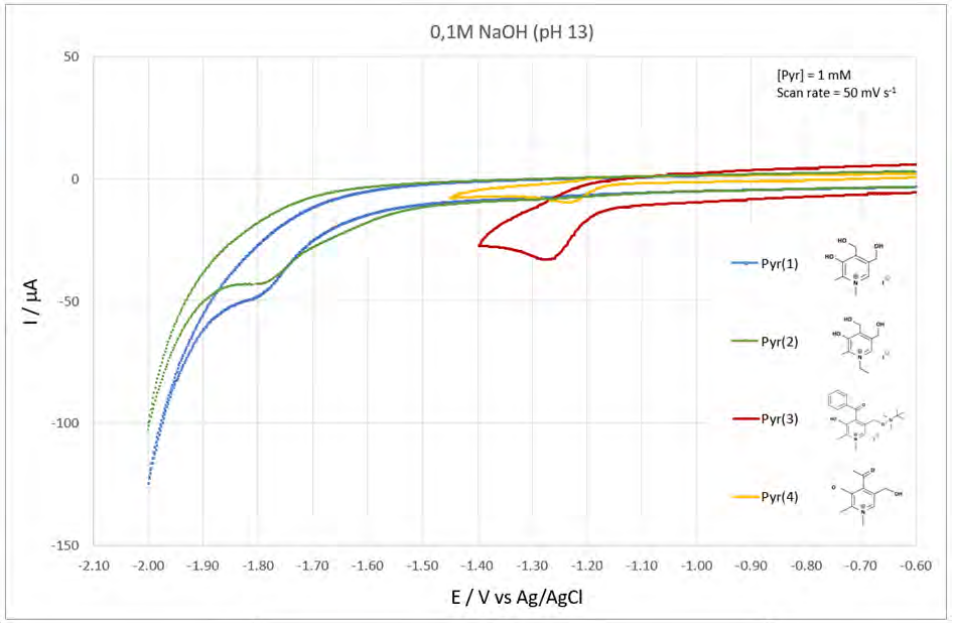
- High-throughput screening is coming

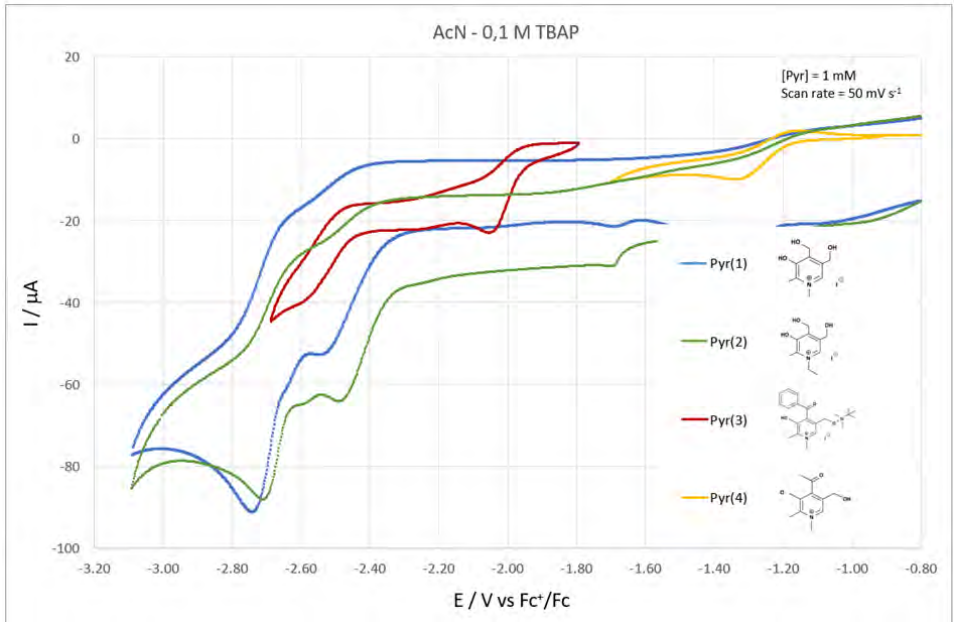
- Cost?
- Stability?

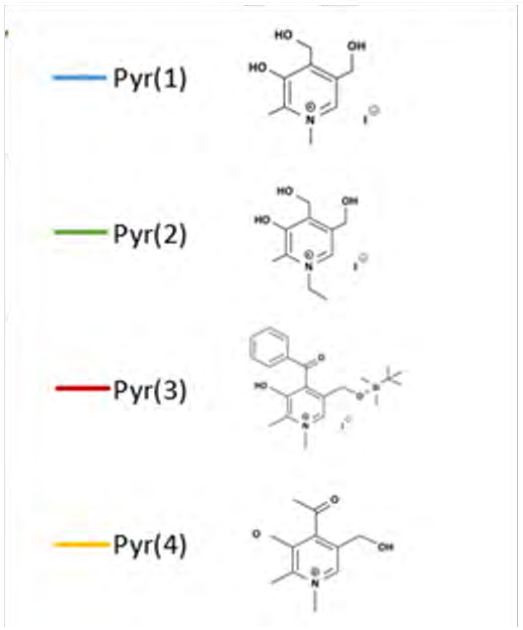
Issues

- Reproducibility problems
  - Oxygen
  - Cross-over
  - Side reactions, degradation
  - Are results comparable?
- **Stability**
  - Very difficult to evaluate computationally!
  - Reproducible experimental testing needed!
  - Voltammetry is not enough.

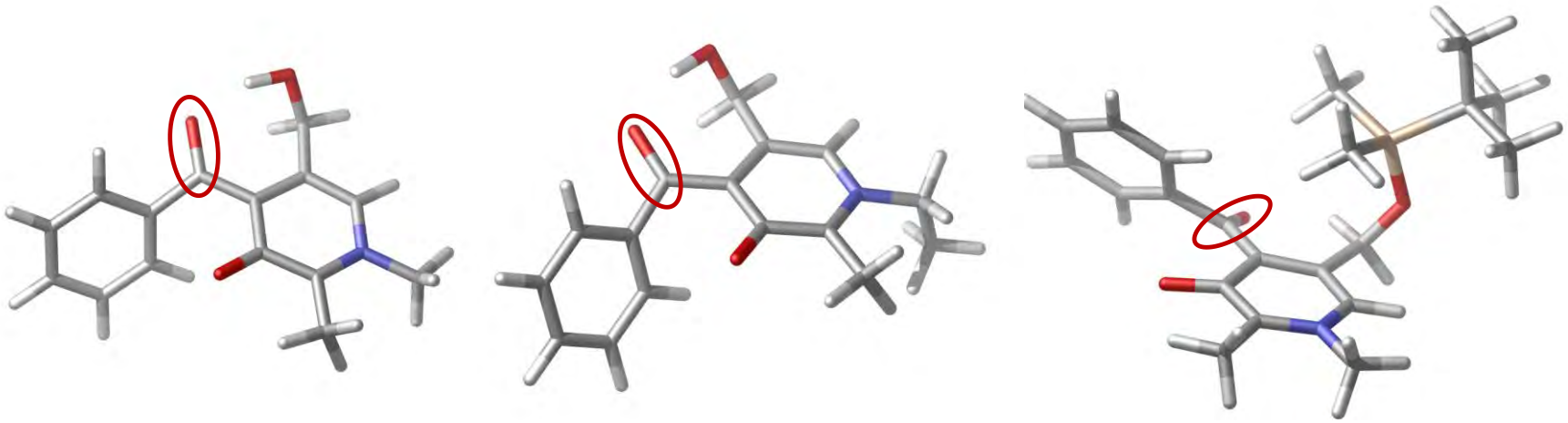

1M KCl (buff pH 7)
[Pyr] = 1 mM
Scan rate = 50 mV s-1
Pyr(1)
Pyr(2)
Pyr(3)
Pyr(4)
I / µA
E / V vs Ag/AgCl


0,1M NaOH (pH 13)
[Pyr] = 1 mM
Scan rate = 50 mV s-1
Pyr(1)
Pyr(2)
Pyr(3)
Pyr(4)
I / µA
E / V vs Ag/AgCl


AcN - 0,1 M TBAP
[Pyr] = 1 mM
Scan rate = 50 mV s-1
Pyr(1)
Pyr(2)
Pyr(3)
Pyr(4)
I / µA
E / V vs Fc+/Fc


Pyr(1)
Pyr(2)
Pyr(3)
Pyr(4)

Computational evaluation of stability



0.053 (0.060) 0.053 (0.060) 0.049 (0.061)

The bond elongations (in Å) for the synthesized molecules. Values obtained from DFT geometry optimizations are shown in parenthesis.

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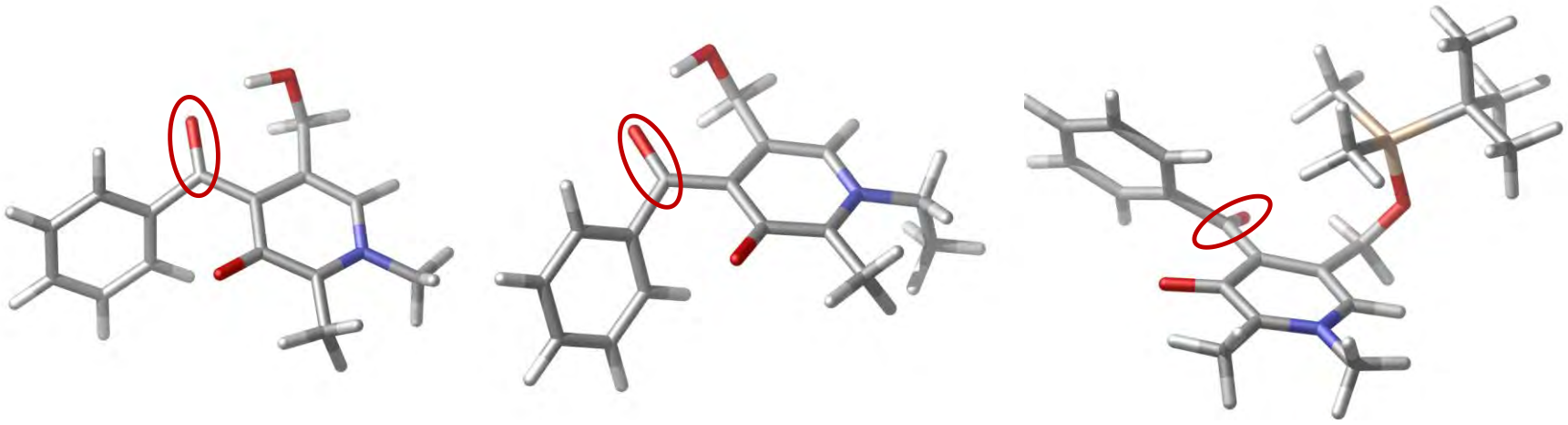
Computational evaluation of stability



0.053 (0.060) 0.053 (0.060) 0.049 (0.061)

The bond elongations (in Å) for the synthesized molecules. Values obtained from DFT geometry optimizations are shown in parenthesis.

Elongation unlikely to explain instability upon reduction.
Experimental analysis suggests dimerization.

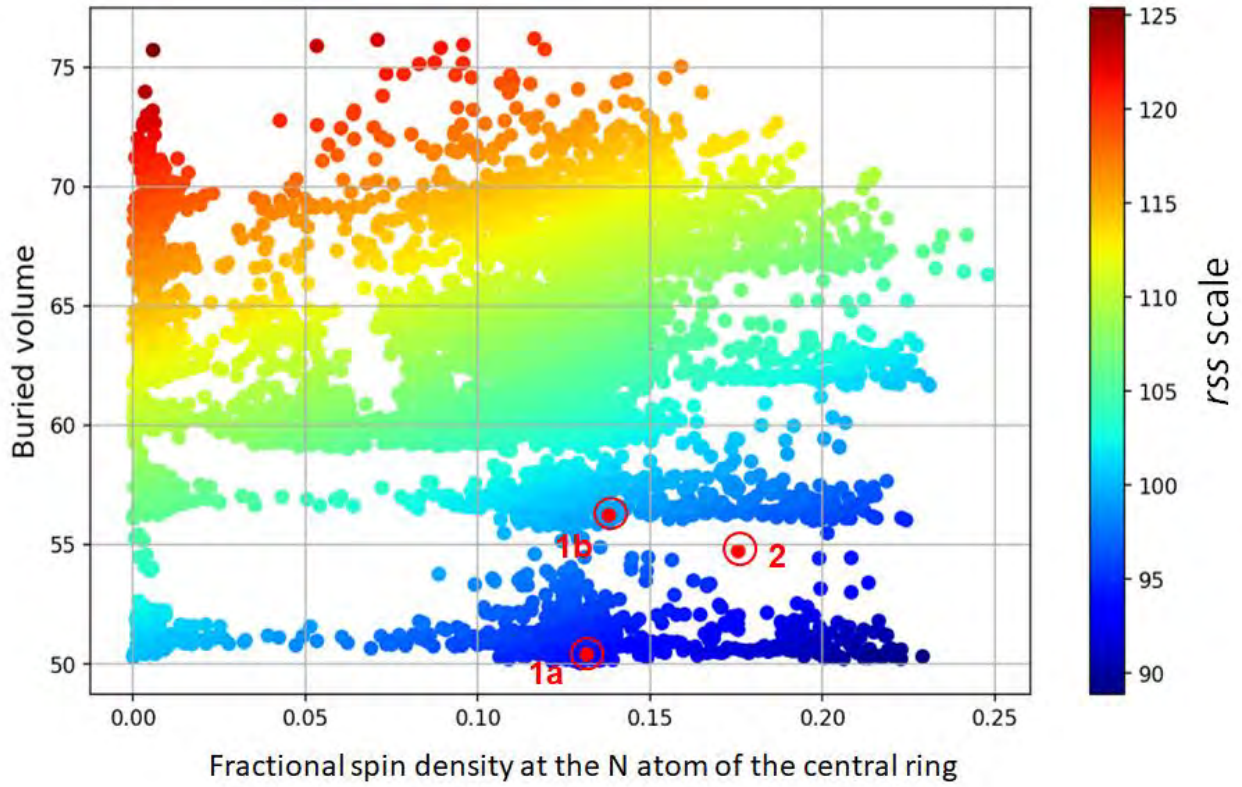
Computational evaluation of stability or radicals

- Thermodynamic stability closely related to the degree of electron spin delocalization (via π-conjugation)

- Kinetics influenced by steric effects

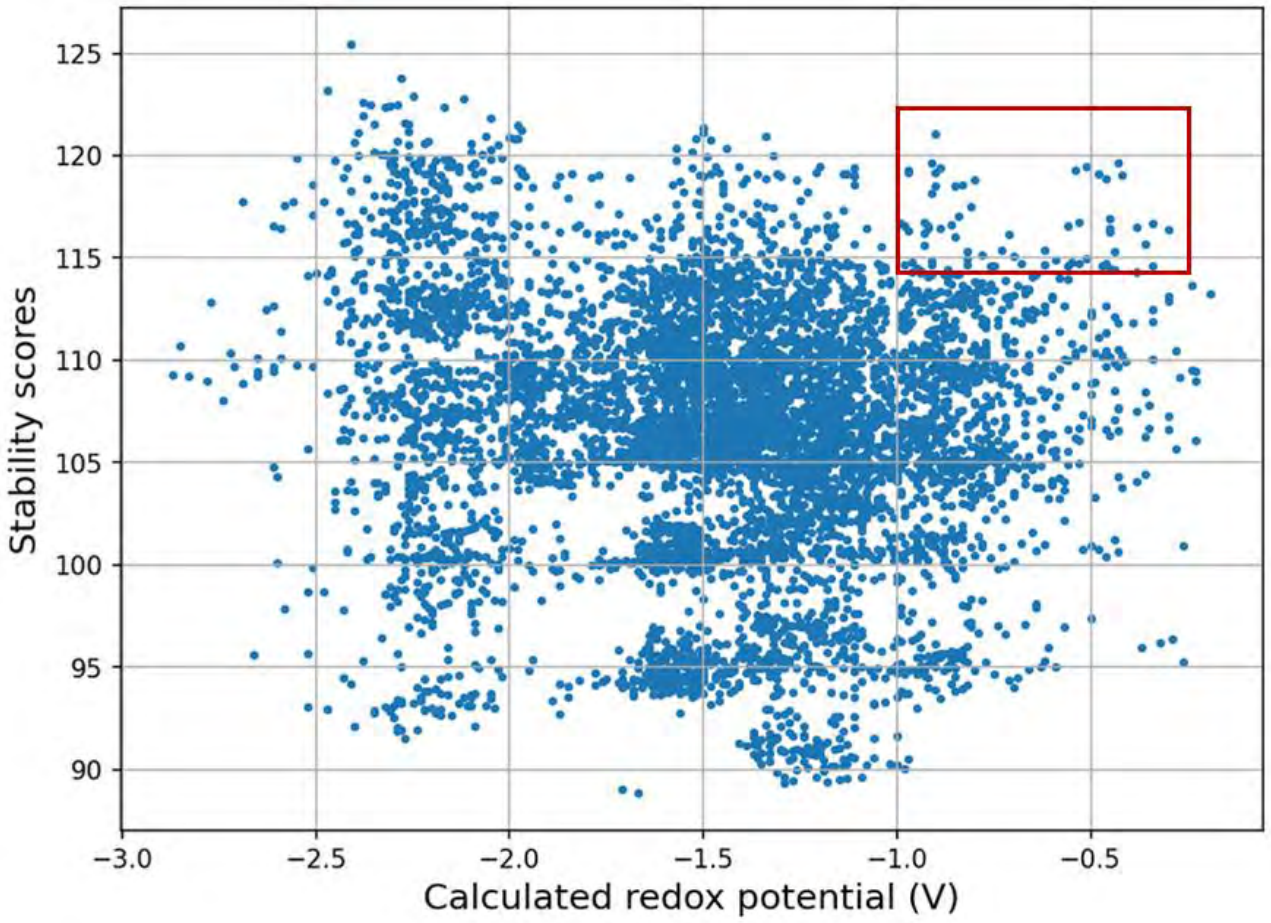
Sowndarya S. V., S.; St. John, P. C.; Paton, R. S. A quantitative metric for organic radical stability and persistence using thermodynamic and kinetic features, *Chem. Sci.*, 2021, *12*, 13158–13166.

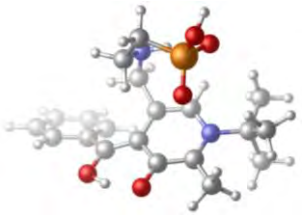
Radical stability score of CompBat molecules



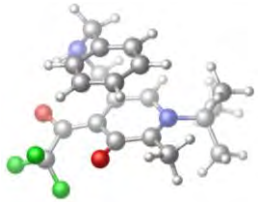
Computed radical stability data for the entire set of radicals included in the DB-II database. Color scale is used for *rss* data (blue – unstable, red – stable radicals).

Radical stability score of CompBat molecules

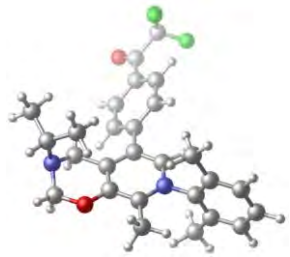




1335

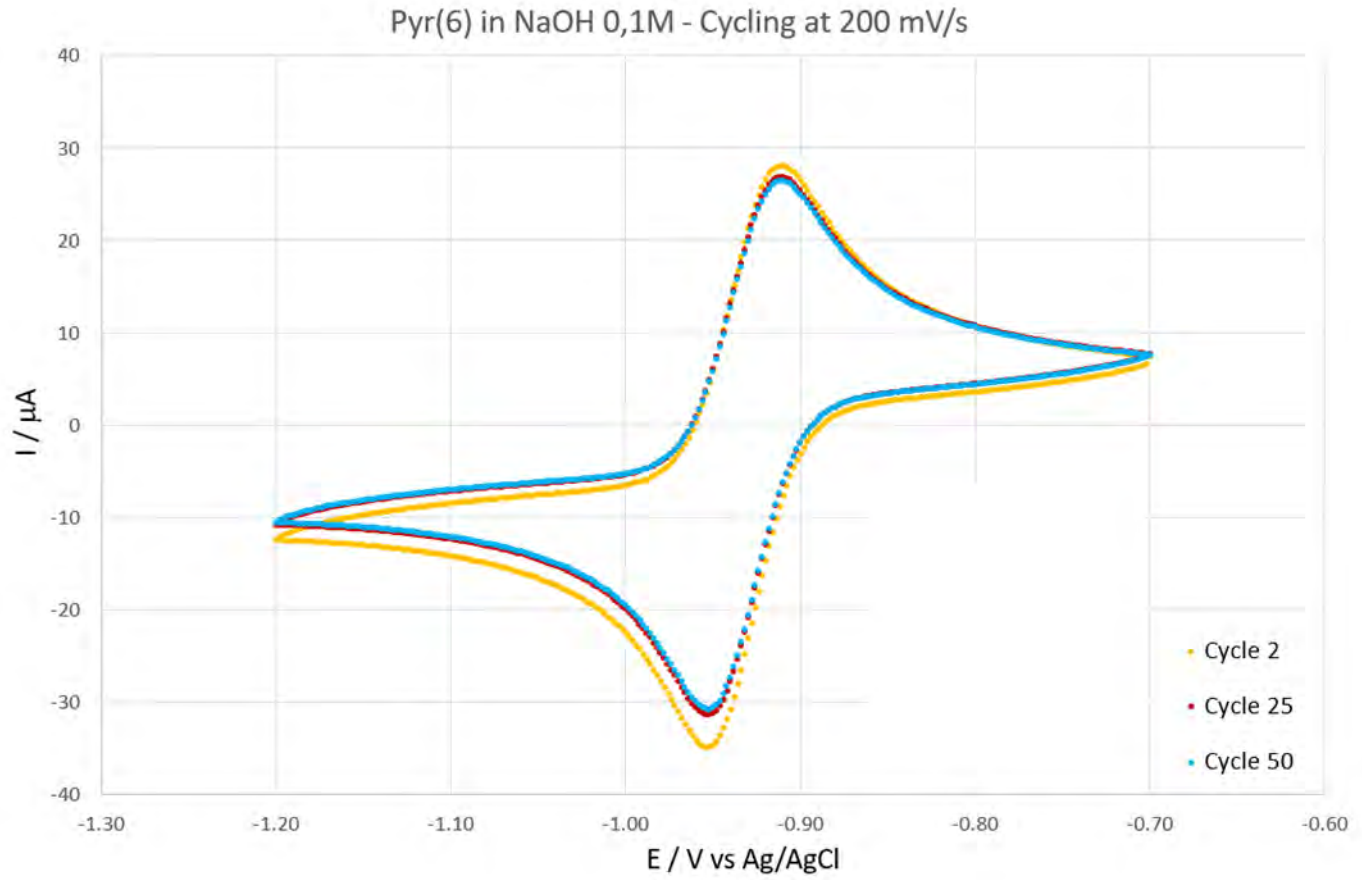


2227



6357

First stable vitamin B6 based molecule synthesized



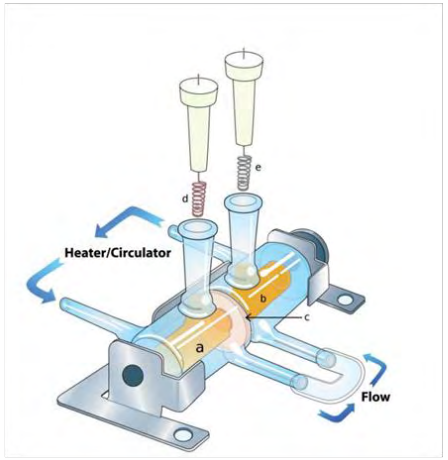
Open questions

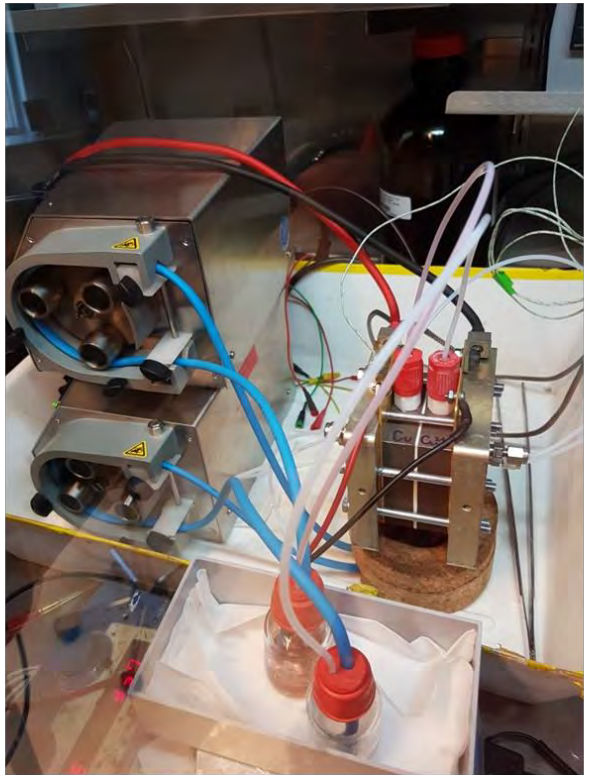
- Computational high-throughput screening
  - For redox potentials and solubility
  - Stability of radicals?
  - Stability of other molecules?
- How to get data for machine learning for design of new materials?
  - Reproducible data
  - High throughput synthesis
  - High throughput testing

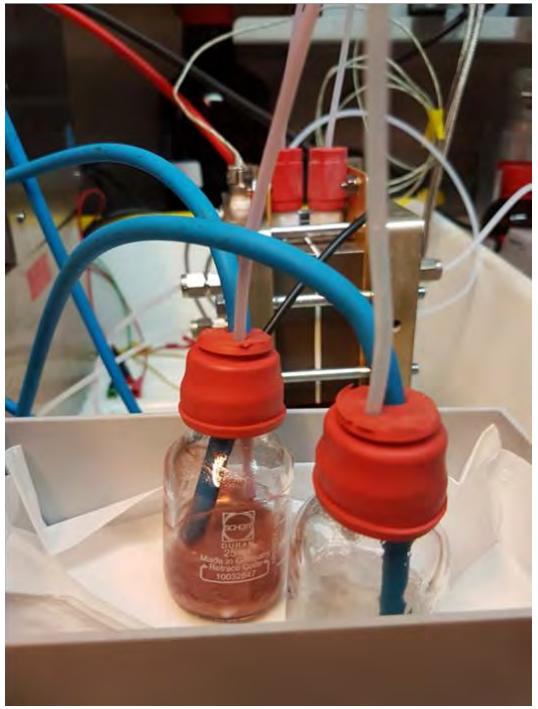
Open questions

- Computational high-throughput screening
  - For redox potentials and solubility
  - Stability?
- How to get data for machine learning for design of new materials?
  - **Reproducible data**
  - **High throughput synthesis**
  - **High throughput testing**

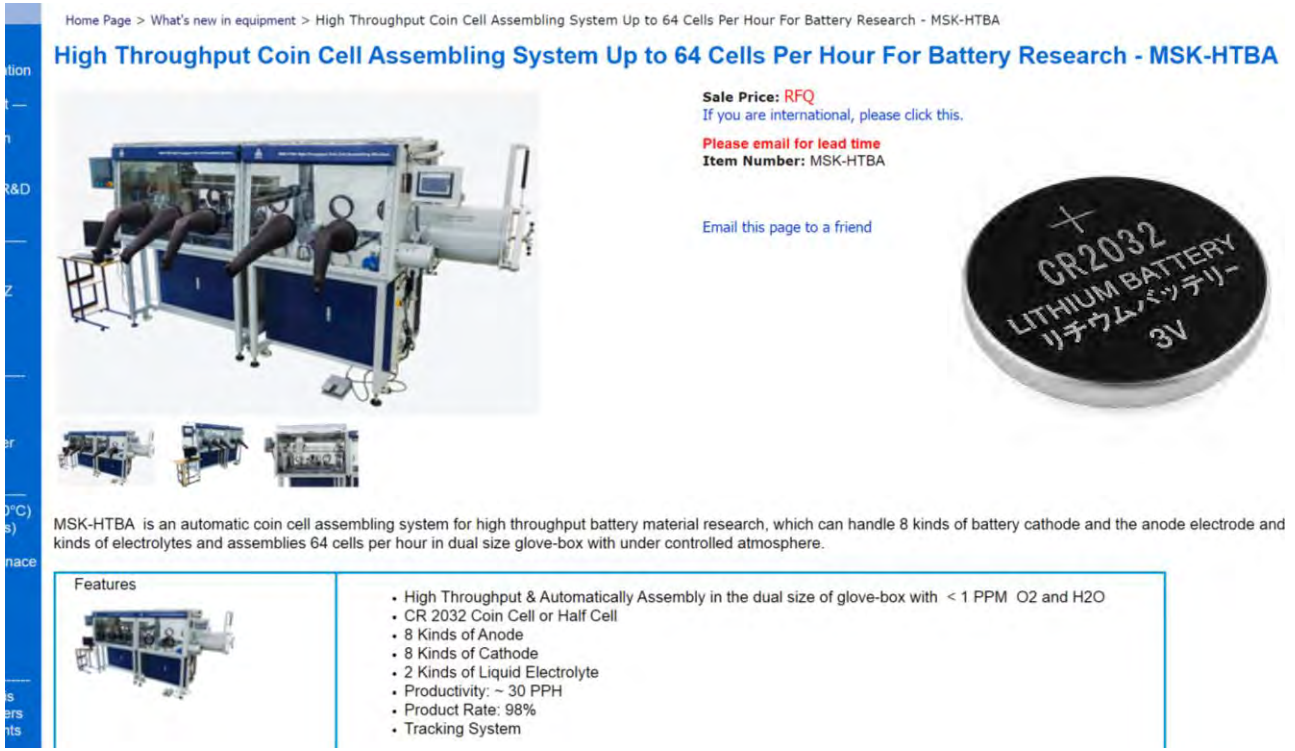
High-throguhput testing?







High throughput testing for Li-ion batteries



Home Page > What's new in equipment > High Throughput Coin Cell Assembling System Up to 64 Cells Per Hour For Battery Research - MSK-HTBA

High Throughput Coin Cell Assembling System Up to 64 Cells Per Hour For Battery Research - MSK-HTBA

Sale Price: RFQ
If you are international, please click this.

Please email for lead time
Item Number: MSK-HTBA

Email this page to a friend

MSK-HTBA is an automatic coin cell assembling system for high throughput battery material research, which can handle 8 kinds of battery cathode and the anode electrode and kinds of electrolytes and assemblies 64 cells per hour in dual size glove-box with under controlled atmosphere.

Features

- High Throughput & Automatically Assembly in the dual size of glove-box with < 1 PPM O2 and H2O
- CR 2032 Coin Cell or Half Cell
- 8 Kinds of Anode
- 8 Kinds of Cathode
- 2 Kinds of Liquid Electrolyte
- Productivity: ~ 30 PPH
- Product Rate: 98%
- Tracking System


Lan
Technics

https://www.chemistryworld.com/news/your-new-labmate-does-700-reactions-in-eight-days-and-its-a-robot/4012125.article

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UNIVERSITY OF TURKU

Gabriel Gonzalez

compbat

https://compbat.eu/

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