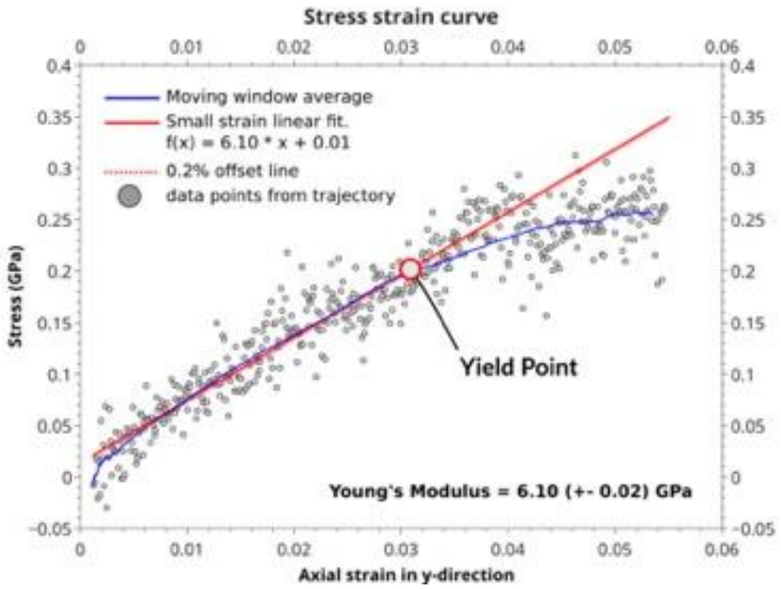
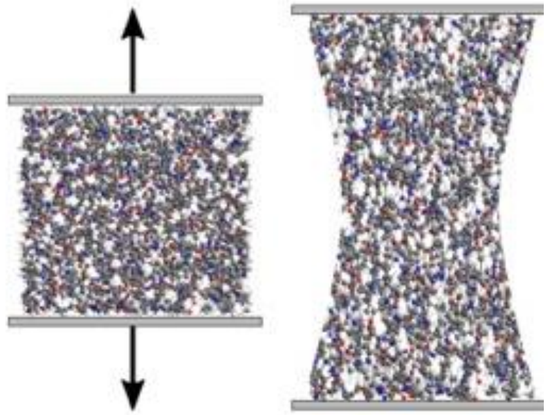
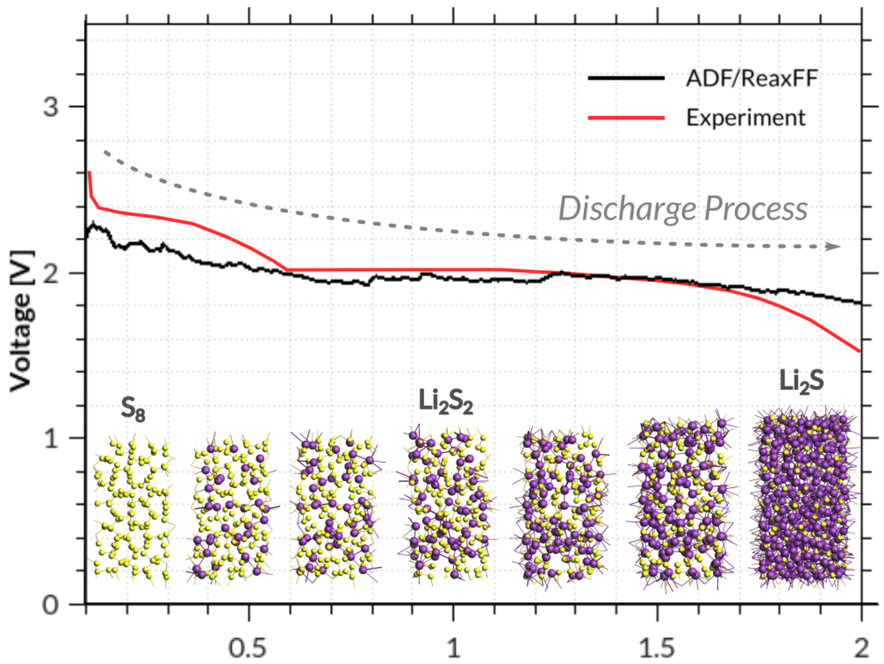
Computer-Accelerated Materials Discovery

Atomistic modeling to reduce R&D time & costs







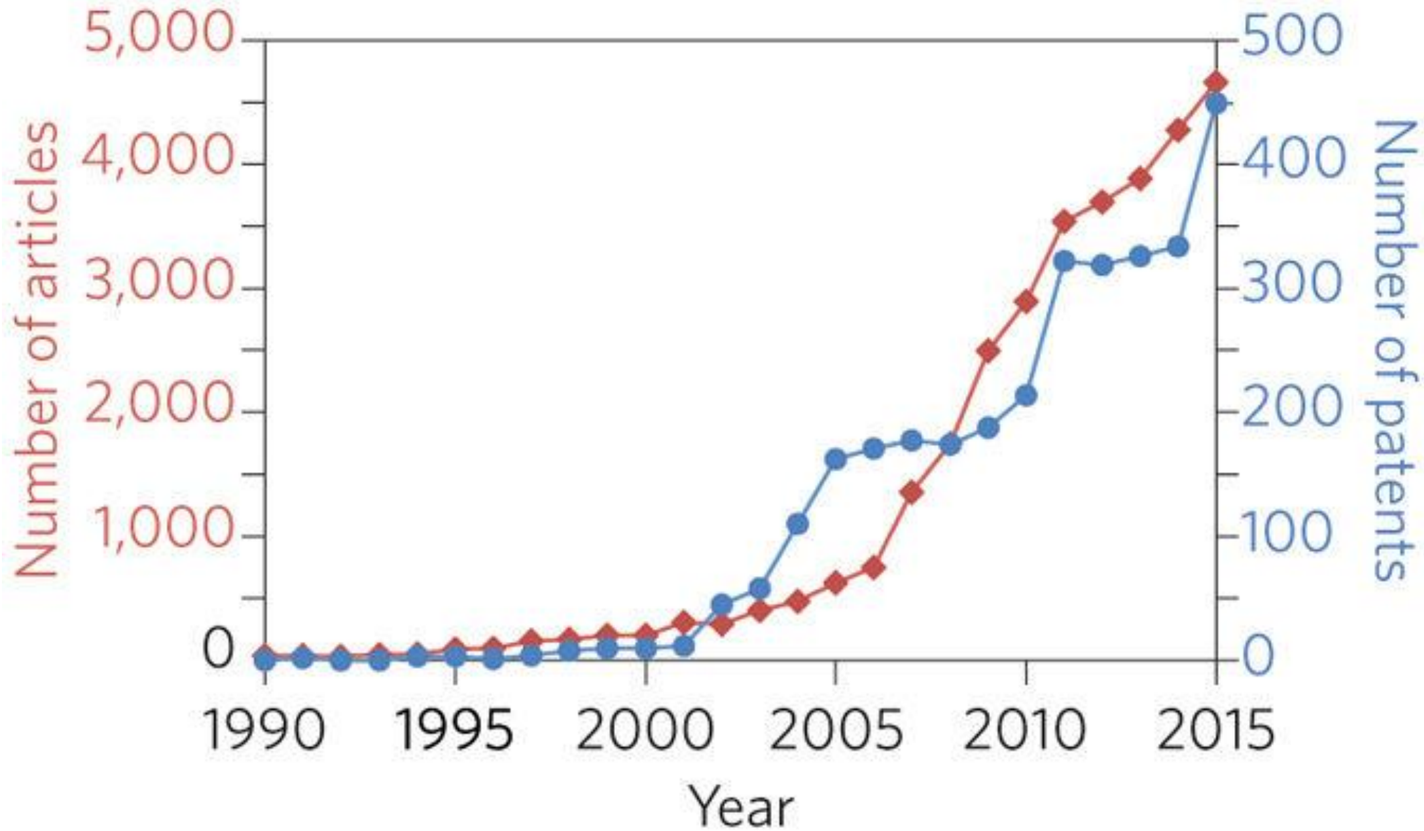
SCM
Software for Chemistry & Materials

Fedor Goumans, Chief Customer Officer

goumans@scm.com

Why use materials modeling in RD&I?

- **Reduce time & costs to invention**
  - Predict: screening potential candidates
  - Understand: insight in material -> property relationships



articles & patents in materials science with "density functional theory", Nat. Mat. 4619

Bottom-up Property Prediction

Properties are determined at the atomistic level => predict, understand & improve through modeling

Batteries:
fast recharge, high capacity, safety

Materials: light & durable
Paint/glass: optical properties, coating

Reduce friction

Solar cells: long lifetime & high efficiency

Background: SCM, ADF & AMS

- ADF: first DFT code to **understand chemistry** (1970s)
  Baerends@VU (>'73), Ziegler@Calgary(+) (>'75)

- 1980s: support from Mitsui, Shell, Akzo, Unilever

- SCM: Spin-off company 1995

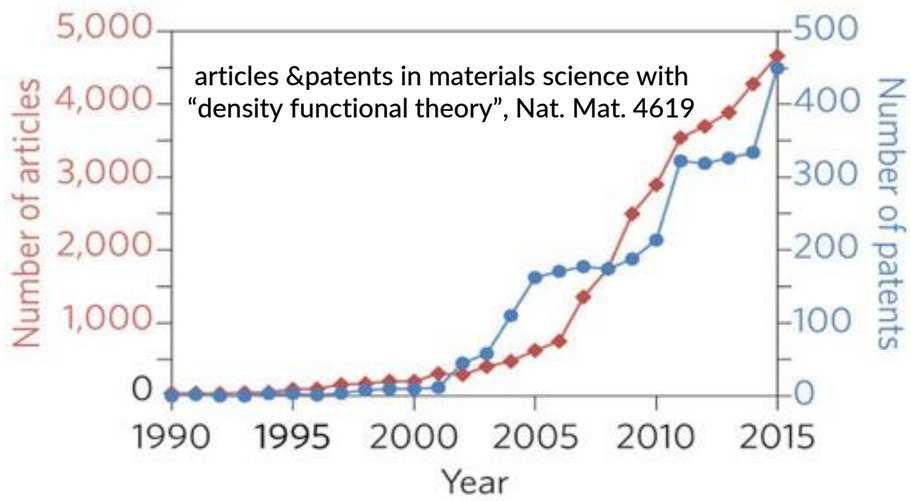
- 2010s: DFTB, ReaxFF, COSMO-RS (Albemarle, DSM)

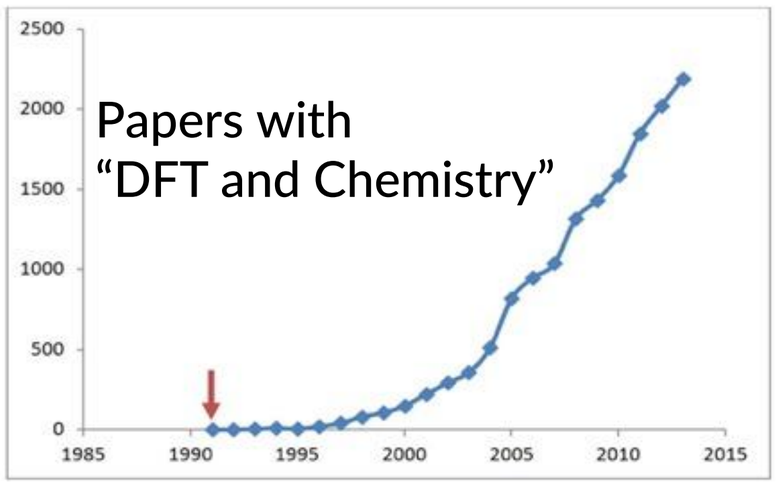
- 2019: Multi-scale: ReaxPro (BASF, Dow, Shell, JM)

- 23 people (19 senior PhD's) + 5 EU fellows

- Many academic collaborators & EU networks

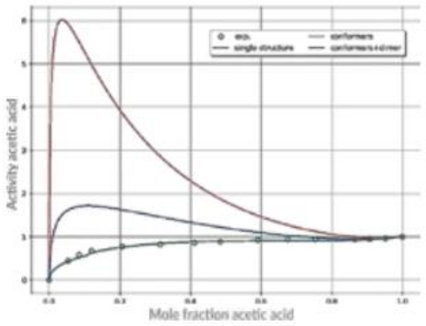
- SCM: development, debug, port, optimize, & <u>support</u>





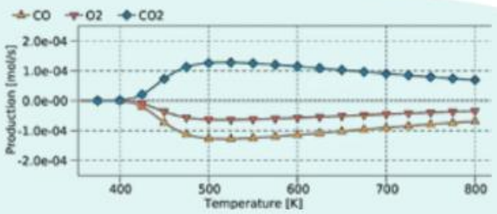
Amsterdam Modeling Suite

Continuum



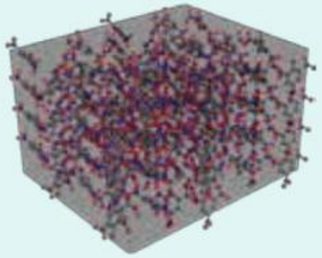
Fluid Thermodynamics
COSMO-RS
COSMO-SAC
UNIFAC

Mesoscale



Kinetics
Kinetic Monte Carlo
Microkinetics

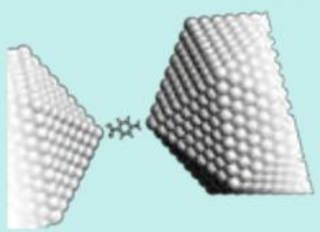
Materials



Force Fields
ReaxFF, GFN-FF
Machine Learning Potentials
Apple & P

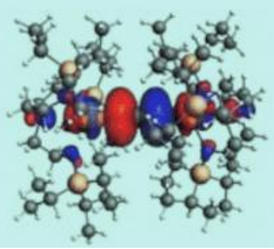
QM/MM
FDE, Hybrid Engine

Nano



Tight binding
GFN-xTB, DFTB

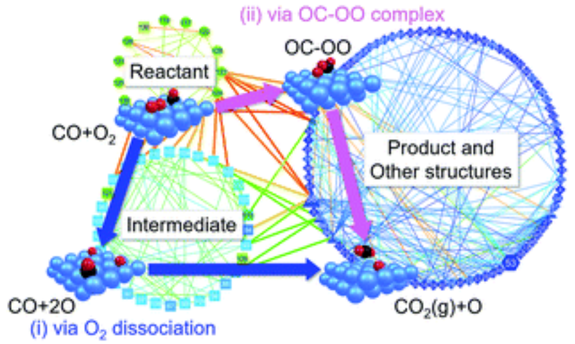
Atomistic



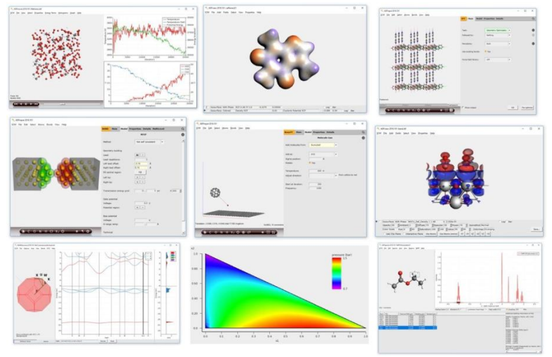
Periodic DFT
BAND, Quantum Espresso

Molecular DFT
ADF

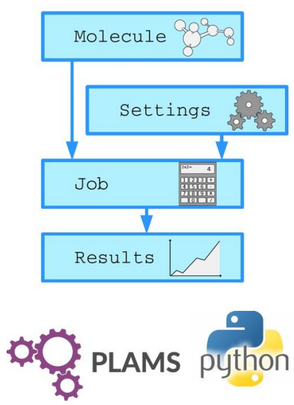
- MD, MC PES exploration

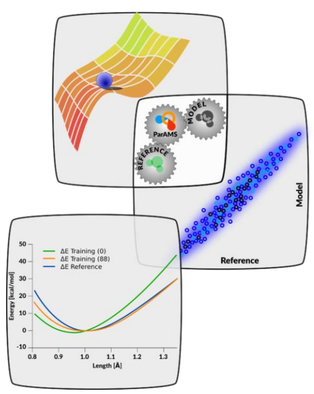


- Integrated GUI



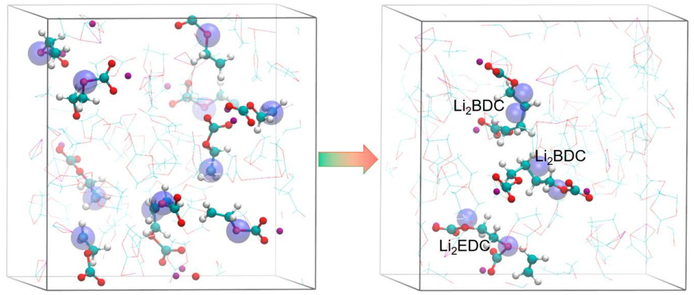
- Python scripting (workflows)



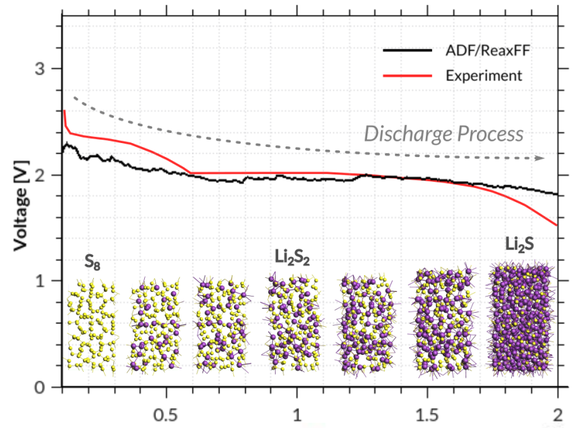


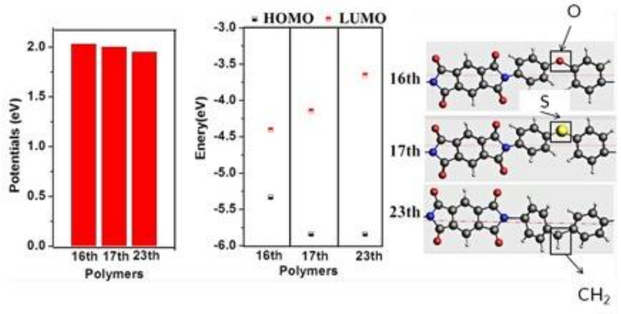
Modeling battery materials

- Charge mobility, diffusion, viscosity
  - ReaxFF, APPLE&P, DFTB-MD
  - NEGF: I-V curves, mobility across interface



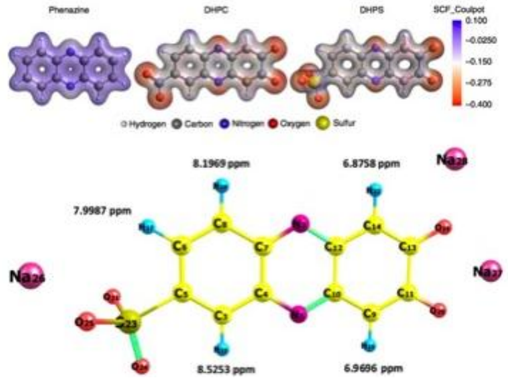
- Electrolyte solubility & electrochemistry
  - Accurate redox potentials (ADF+COSMO-RS), ionization potentials
  - Solubility: COSMO-RS
  - (e)ReaxFF: electrolyte degradation
  - ReaxFF, DFTB, BAND, polymer properties (band gaps)
  - BAND: include solvation, E field



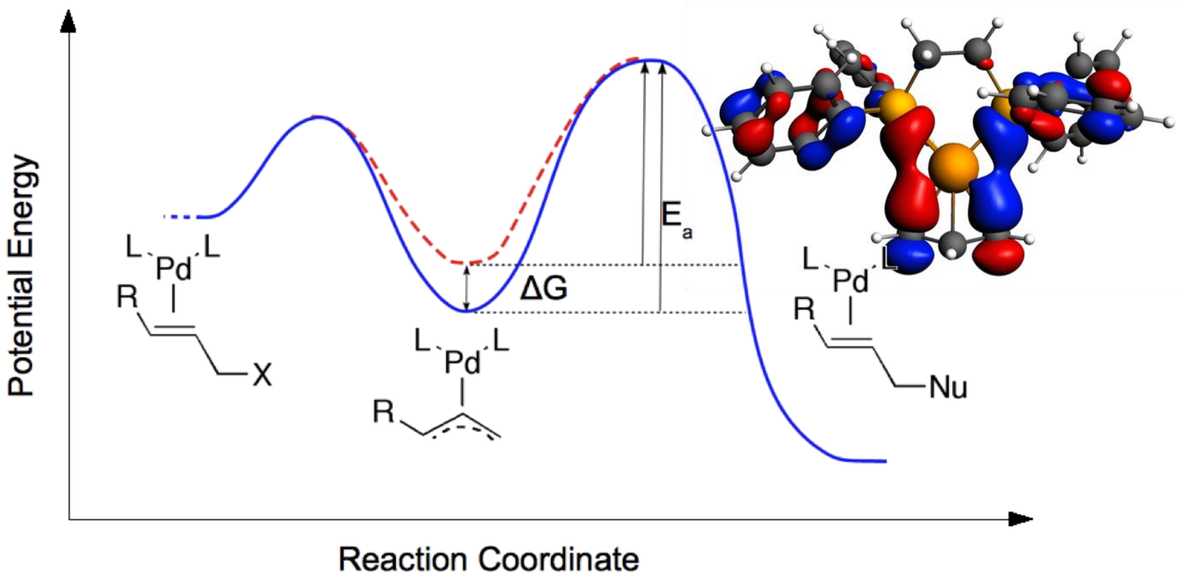


- (Dis)charge processes
  - GCMC with ReaxFF, or DFT(B)

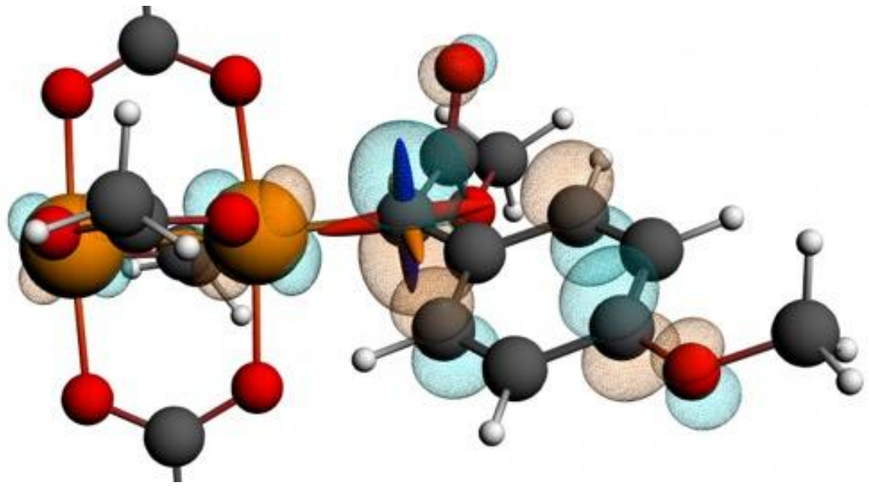
- Understand battery 'operando'
  - ADF - Spectroscopy: NMR, NEXAFS



ADF: Molecular DFT



Bonding analysis: Understanding Catalyst-Substrate Interactions Nature Chem. 2, 417 (2010)



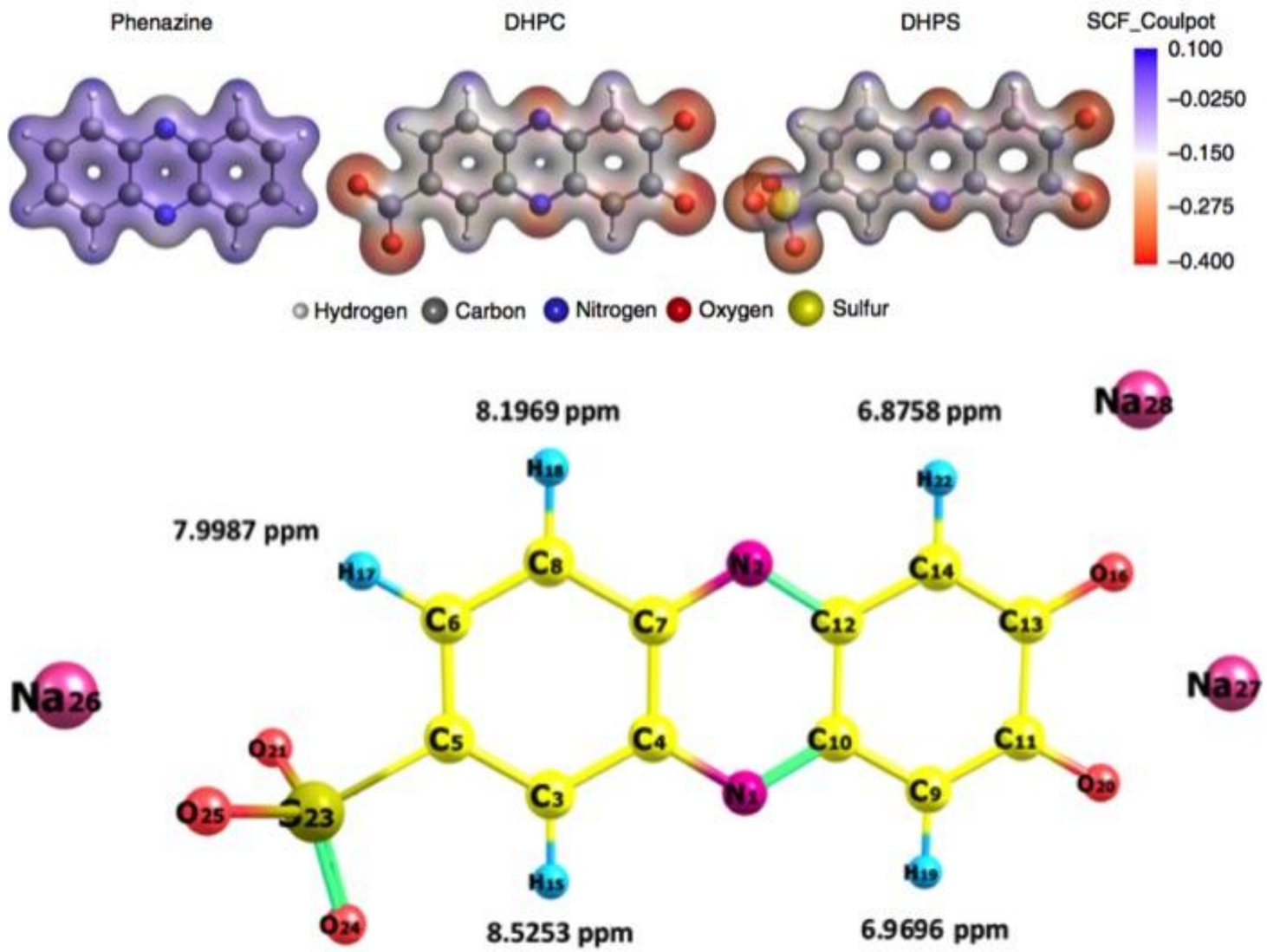
*NMR calculations locate ^{13}C di-Rh carbene catalyst intermediate, Science, **342**, 351 (2013)*

Strong & unique points

- All-electron Slaters, H-Og
- Relativity: ZORA (SR, **SOC**)
- Spectroscopy
  - EPR, NMR, IR (VCD), UVVIS, XANES
- Fast double hybrids (energies), qsGW
- Bonding analysis
  - Fragment-based approach
  - ETS-NOCV, QTAIM, MO diagrams, NCI, Fukui function, dual descriptor
  - Transfer integrals
- Environments
  - Subsystem DFT (FDE), QM/MM, QM/QM', polarizable force fields
  - SM12, COSMO
  - 3D-RISM

Solvation energies, redox potentials, NMR spectra

Developing non-flammable electrolytes:
DFT + Continuum Solvation (COSMO) aids experiments

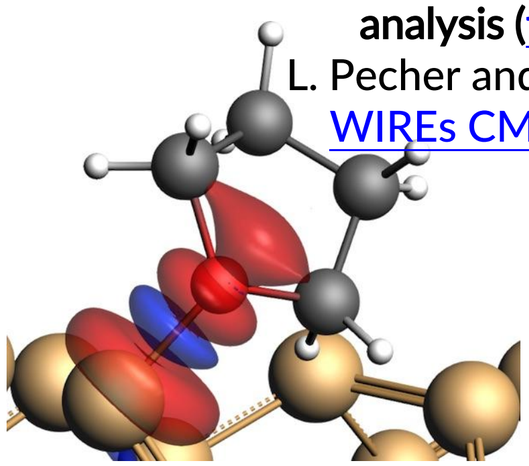


A biomimetic high-capacity phenazine-based anolyte for aqueous organic redox flow batteries, [*Nature Energy 3, 508-514 (2018)*]
Non-flammable electrolytes with high salt-to-solvent ratios for Li-ion and Li-metal batteries, [*Nature Energy 3, 674–681 (2018)*]

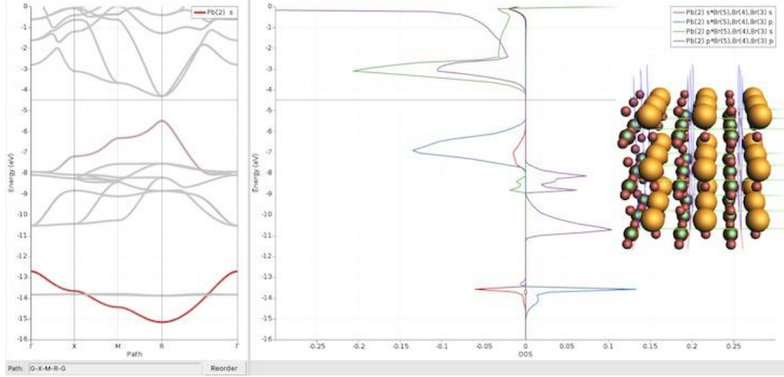
BAND vs. Plane Wave codes (QE)

- Atom centered basis functions, STO or NAO
  - Compare cluster with periodic
  - Basis sets: **no pseudopotentials**, all elements
  - Self-consistent **spin-orbit coupling**
  - Core spectroscopy (core holes)
  - Easy orbital **analysis:** pDOS, COOP, EDA
  - Fast for empty (1D, 2D, porous)
  - xc: r2SCAN, MN15-L, HSE06, GLLB-sc, D4, DFT-1/2
  - Self-consistent NEGF

periodic energy decomposition analysis (tutorial)
L. Pecher and R. Tonner
WIREs CMS, (2018)

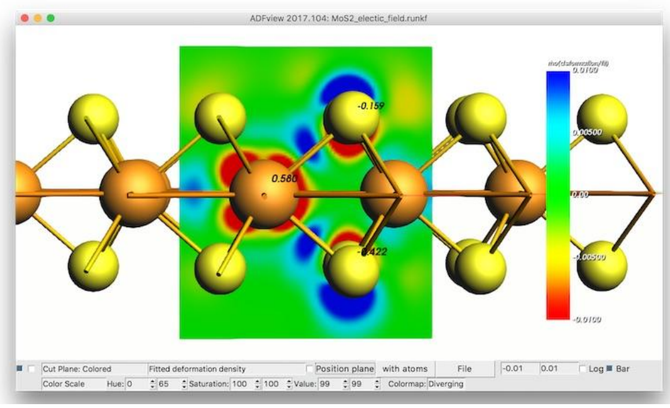


- True 2D surfaces, 1D polymers
  - Solvation: COSMO, **SM12**
  - Electronics (homogeneous E field)
  - **Nanotubes**



COOP in perovskites (tutorial)
Goesten & Hoffmann
JACS (2018)

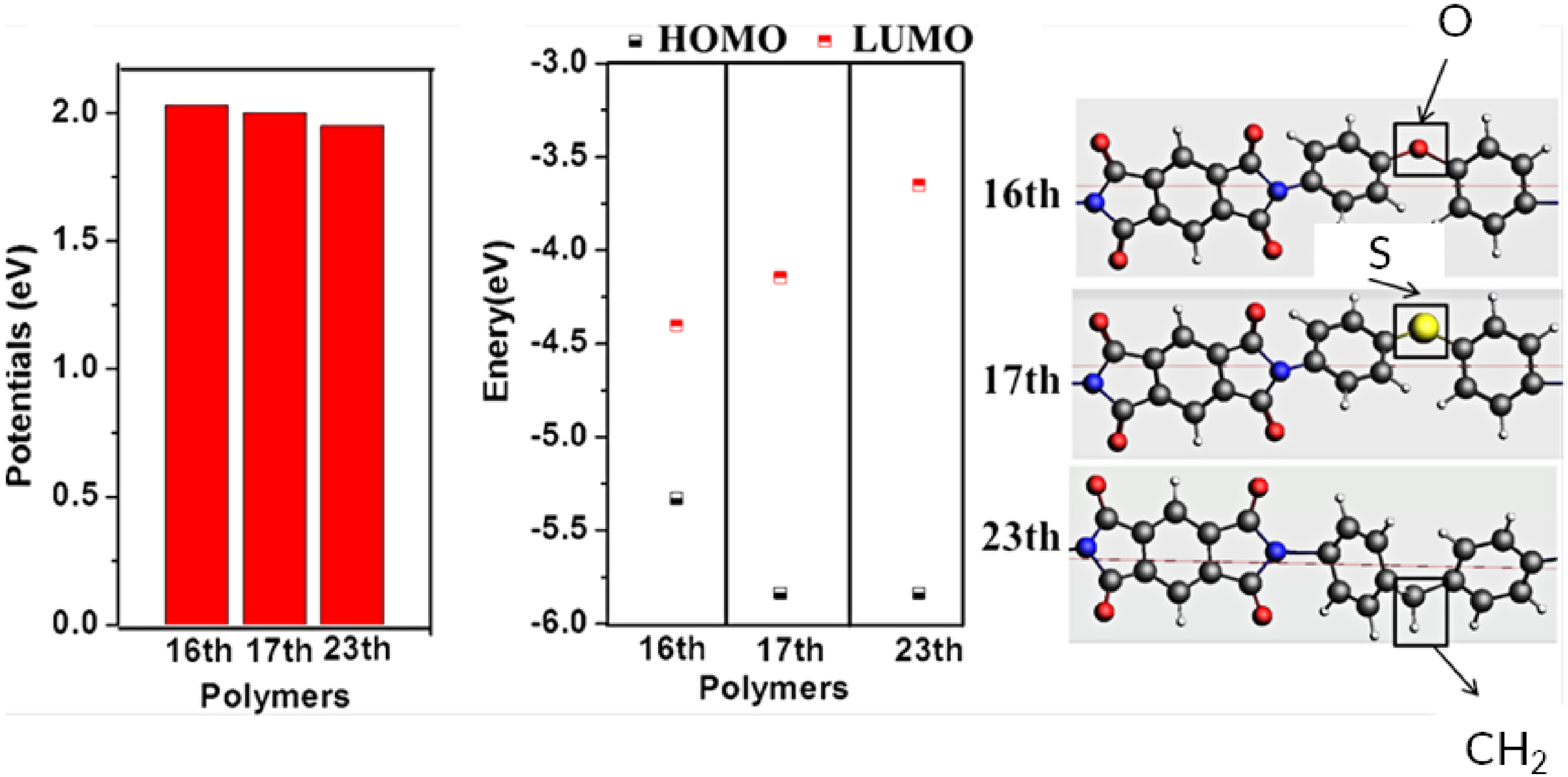
- Integrated Graphical Interface:
  - Easy set up & analysis
  - Switch: ADF, BAND / QE / VASP



Polarizing 2D semiconductor (tutorial)
N. Zibouche et al.
PCCP (2014)

Screening polymers for Lithium Ion Batteries

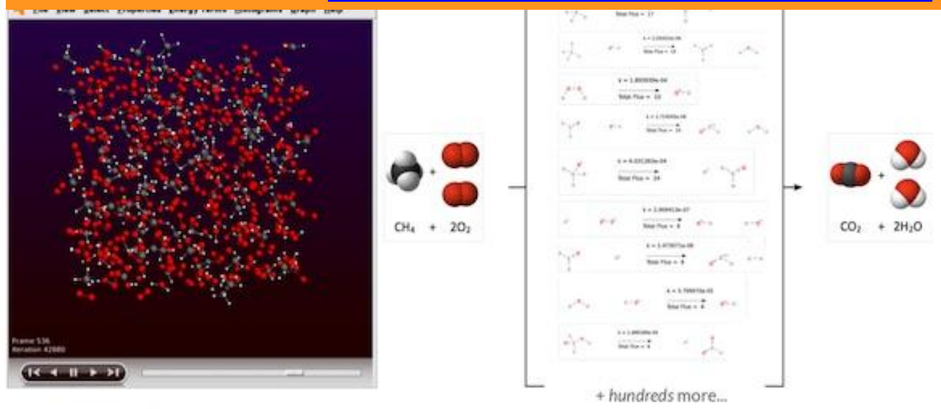
Screening band gaps and lithiation energies with 1D periodic DFT + COSMO



H. Lu, J. Yu, G. Chen, and S. Sun, Theoretical screening of novel electrode materials for lithium–ion batteries from industrial polymers, Ionics (2019)

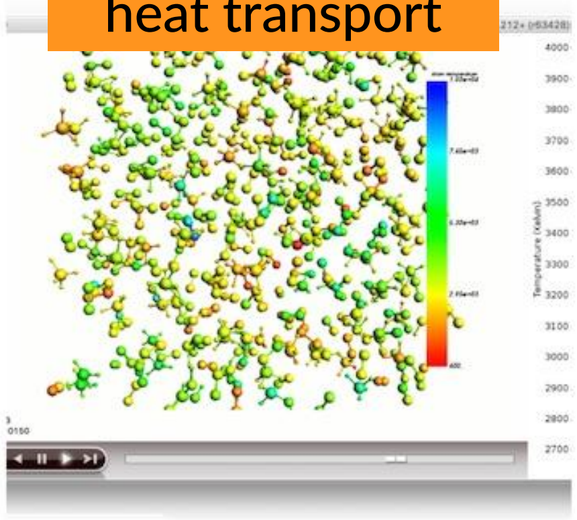
Reactive MD tools Amsterdam Modeling Suite

ChemTraYzer: Automated rates & pathways
New: Analyze surface reactions

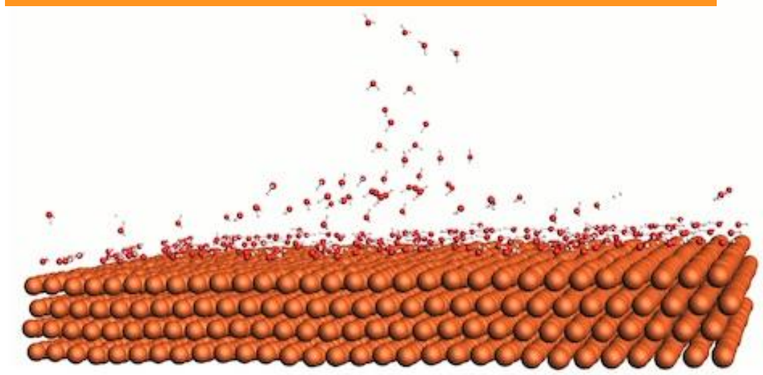


Tools += complete reaction networks
elementary reactions, rate constants, fluxes, timeline

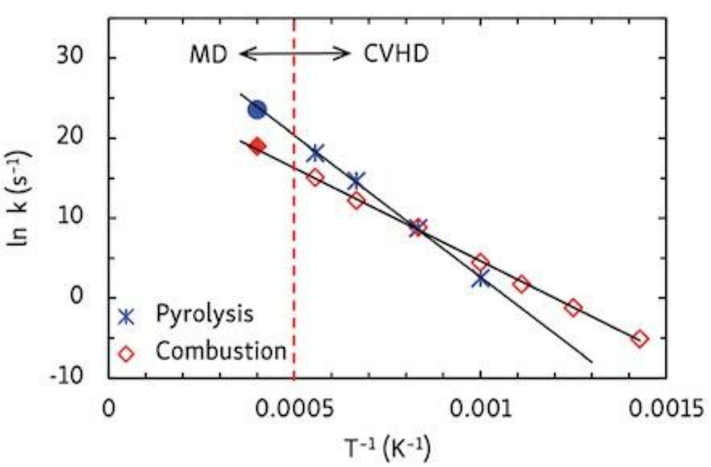
T-NEMD, local T: heat transport

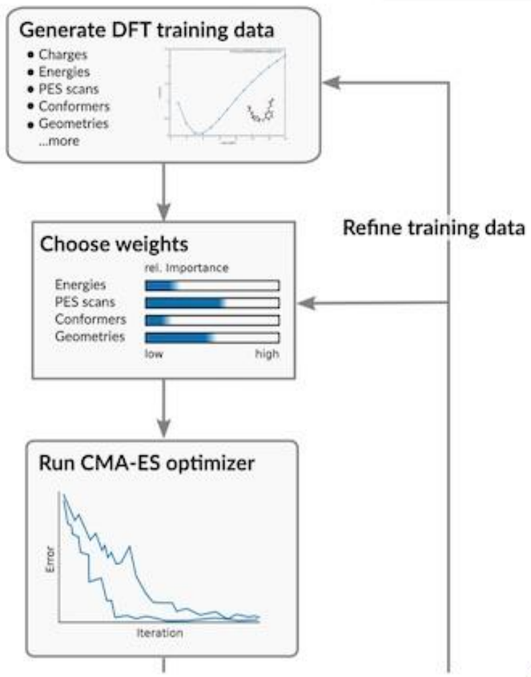


Molecule gun: depositing molecules on surfaces



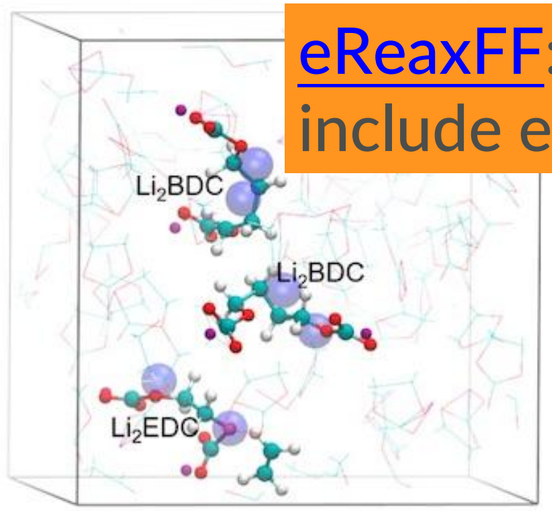
fbMC, CVHD, PRD?: speed up kinetics

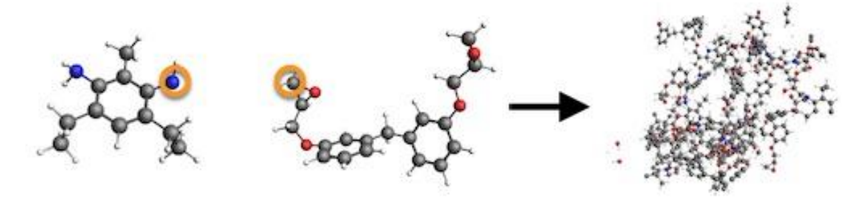




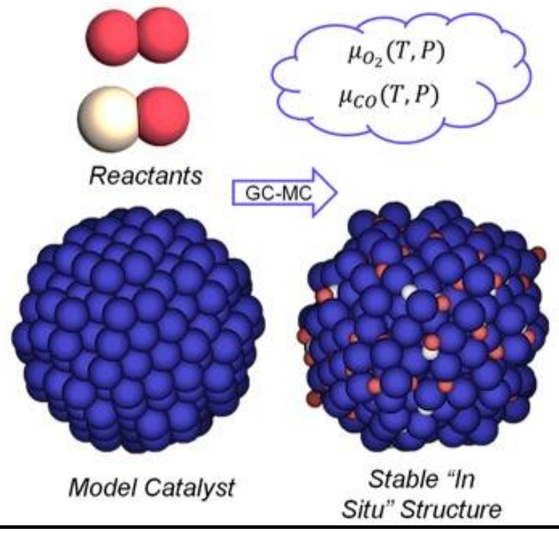
ParAMS - easy ReaxFF& DFTB (re)parameterization

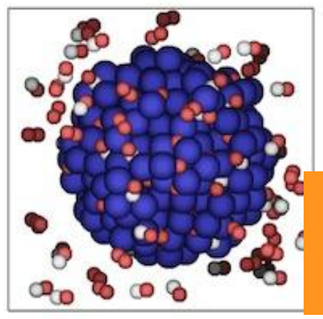
eReaxFF: include e-





bond boost build polymers

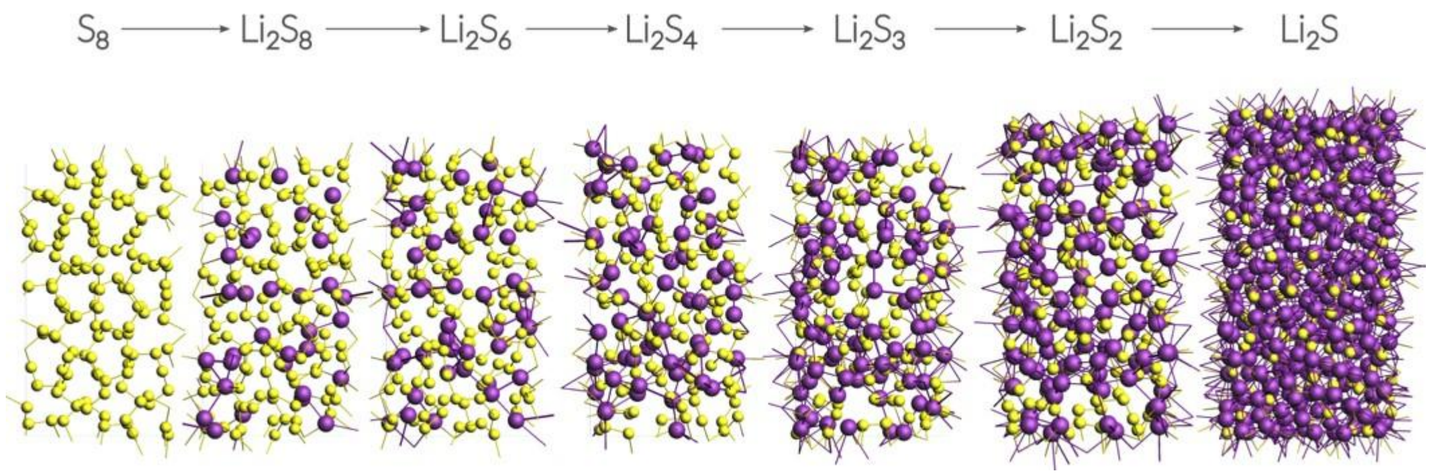




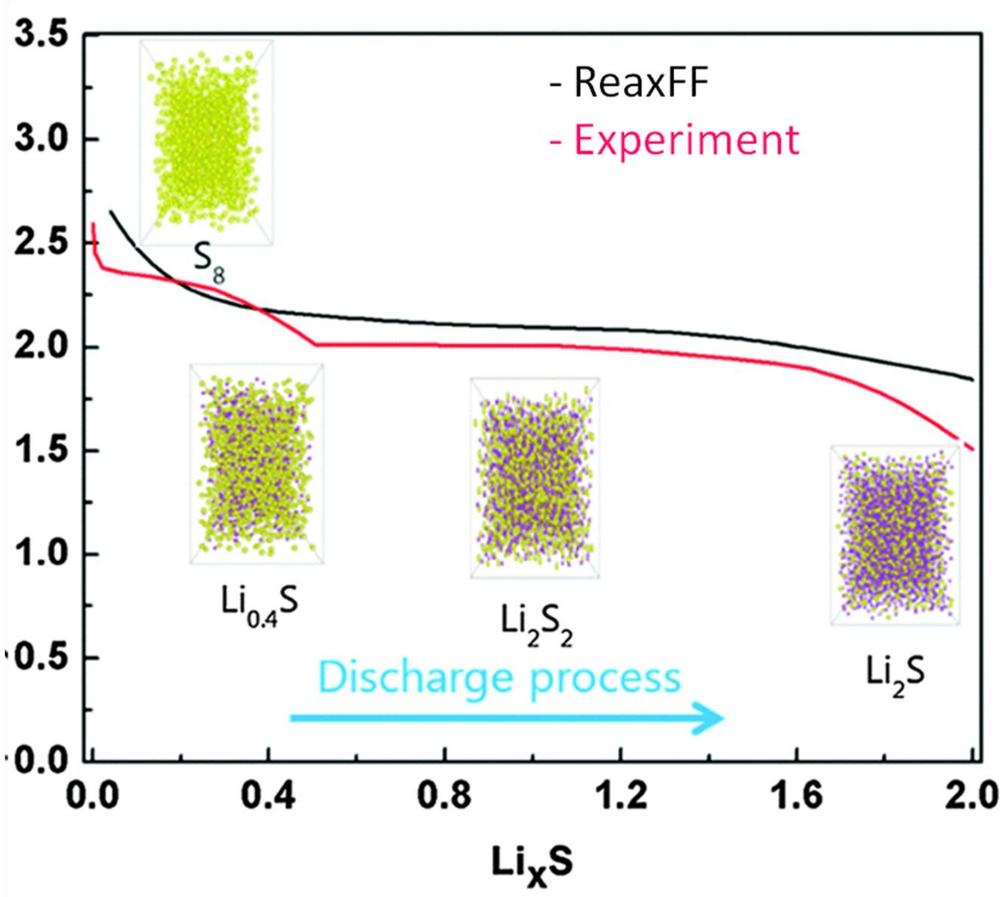
GCMC: speed up thermo

Discharge process Li-S batteries

- Cathode expansion
- Voltage reduction
- Diffusion induced stress



Tutorials: Battery discharge (GCMC) & Li ion diffusion

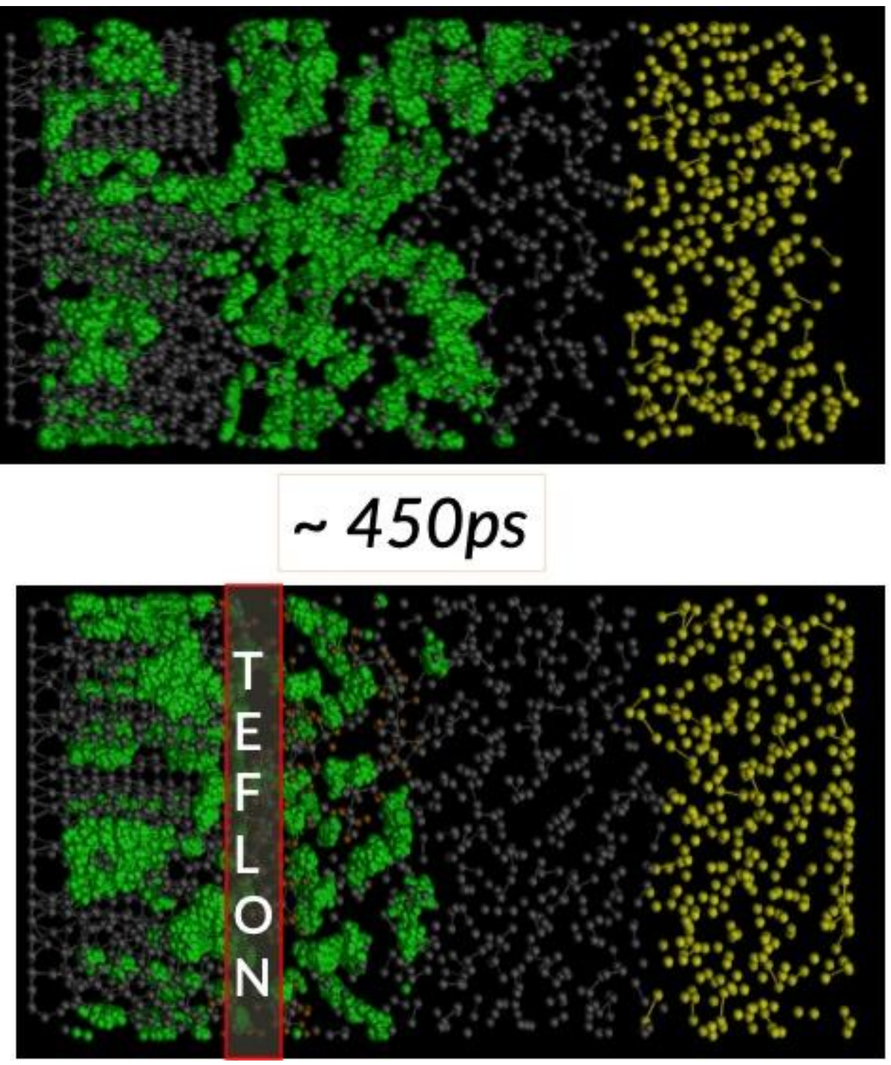


Battery discharge video

M. M. Islam, A. Ostadhossein, O. Borodin, A. T. Yeates, W. W. Tipton, R. G. Hennig, N. Kumar, and A. C. T. van Duin, *ReaxFF molecular dynamics simulations on lithiated sulfur cathode materials*, Phys. Chem. Chem. Phys. **17**, 3383-3393 (2015)

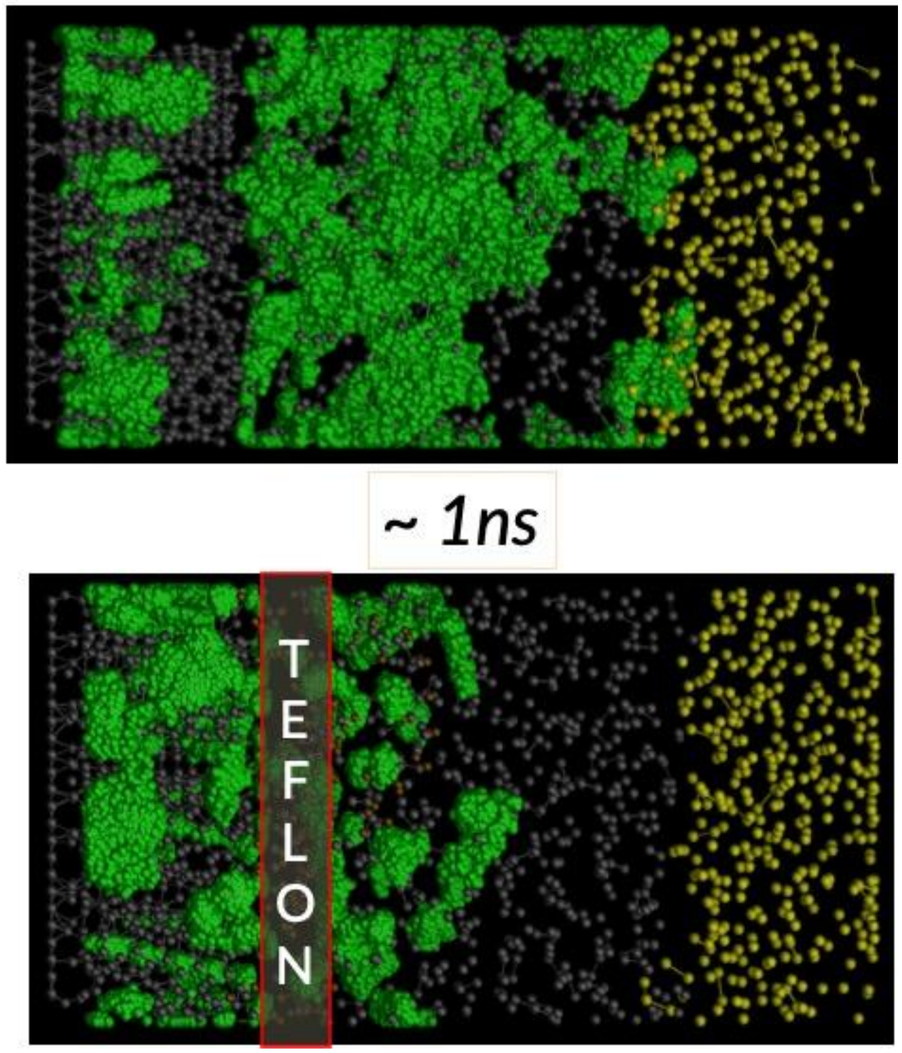
Teflon protects electrolyte in Li battery

Teflon layer on anode-electrolyte interface significantly reduces lithium reactivity and diffusion through the electrolyte phase



NVT, 300K

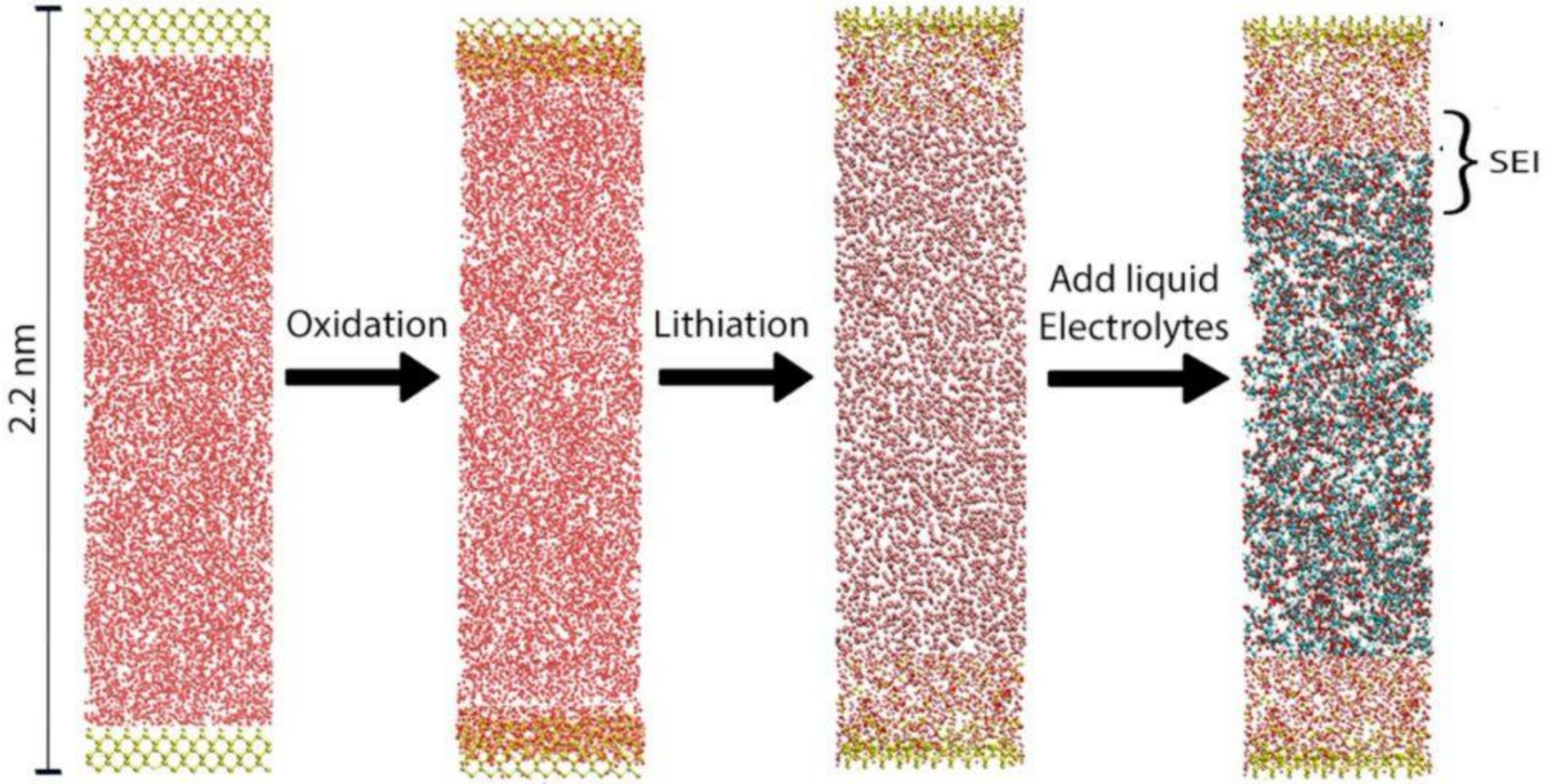
Li discharge



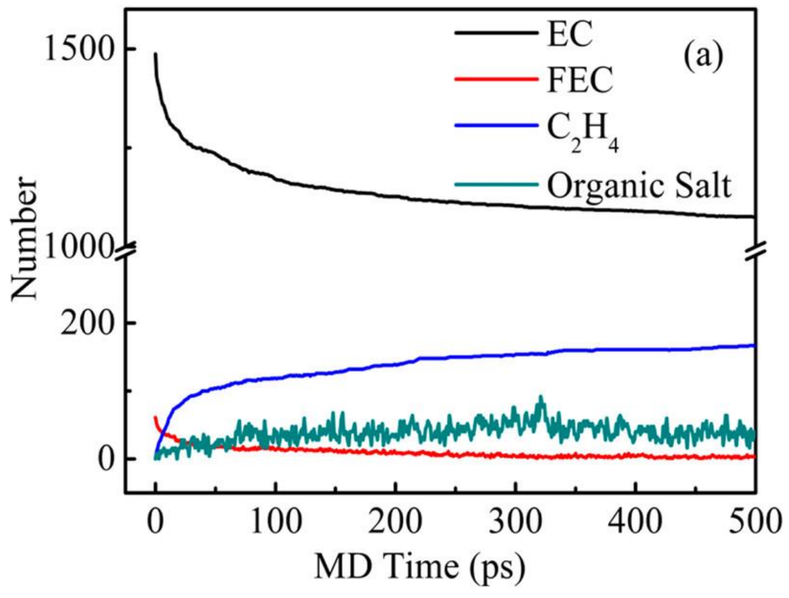
J. Electrochem. Soc. ***161****, E3009-E3014 (2014).*

Solid electrolyte interface formation Lithium Ion Batteries

ReaxFF protocol to study the initial formation stages of SEI formation

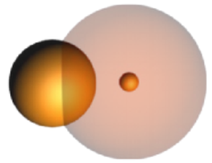


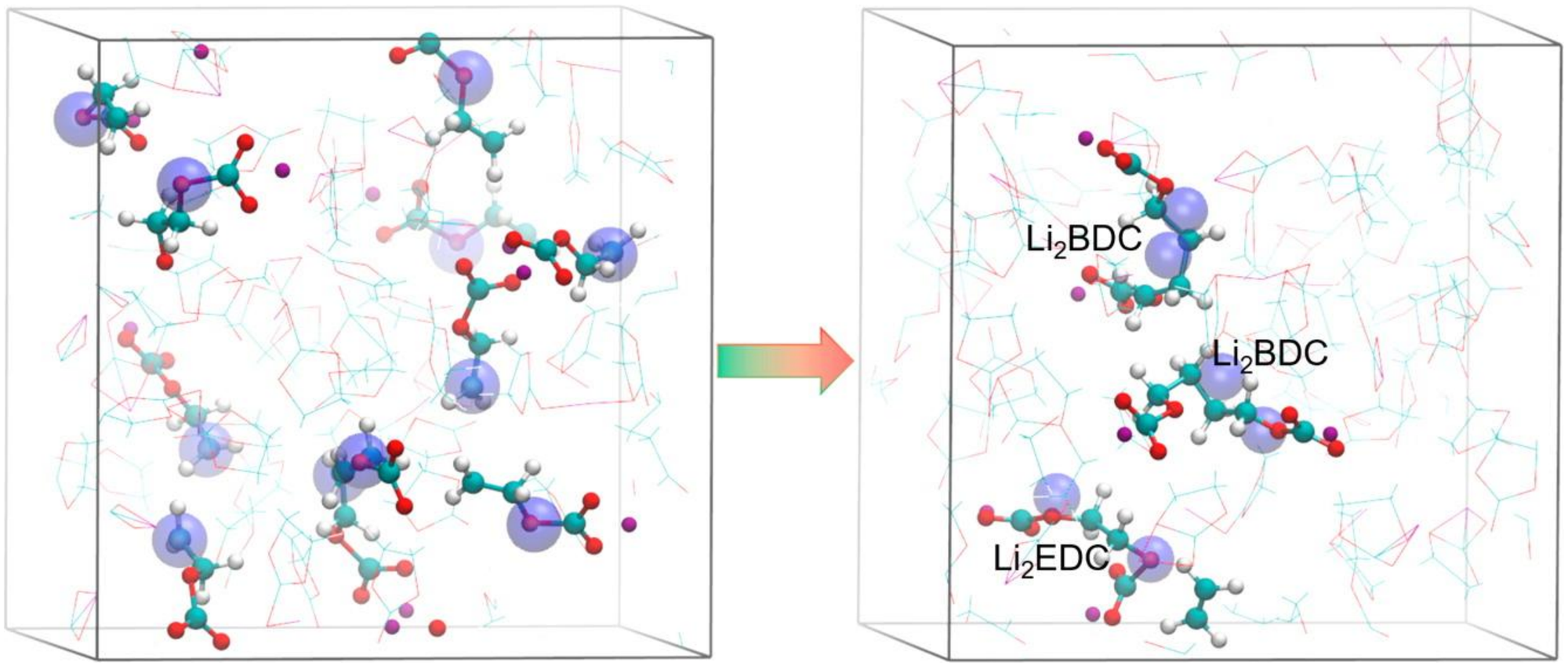
Wang J, Liun Y, Tu Y, Wang Q, Reductive Decomposition of Solvents and Additives toward Solid-Electrolyte Interphase Formation in Lithium-Ion Battery, J. Phys. Chem. (2020).



eReaxFF – Solid-Electrolyte Interfaces

Reductive decomposition of ethylene carbonate in Li ion batteries

Setup: Li-atom →  (Li^+/e^- -pair)

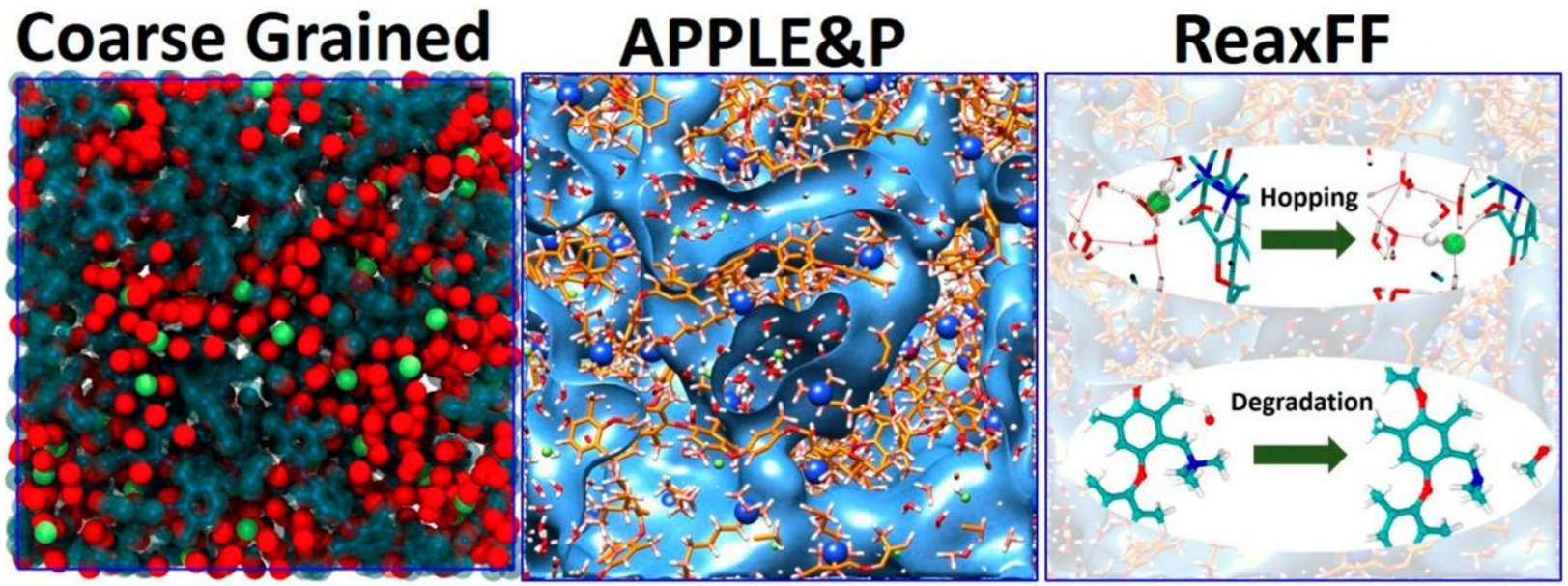


[eReaxFF video]

M. M. Islam and A. C. T. van Duin, J. Phys. Chem. C 2016, 120 (48), 27128-27134.

Extending to larger scales

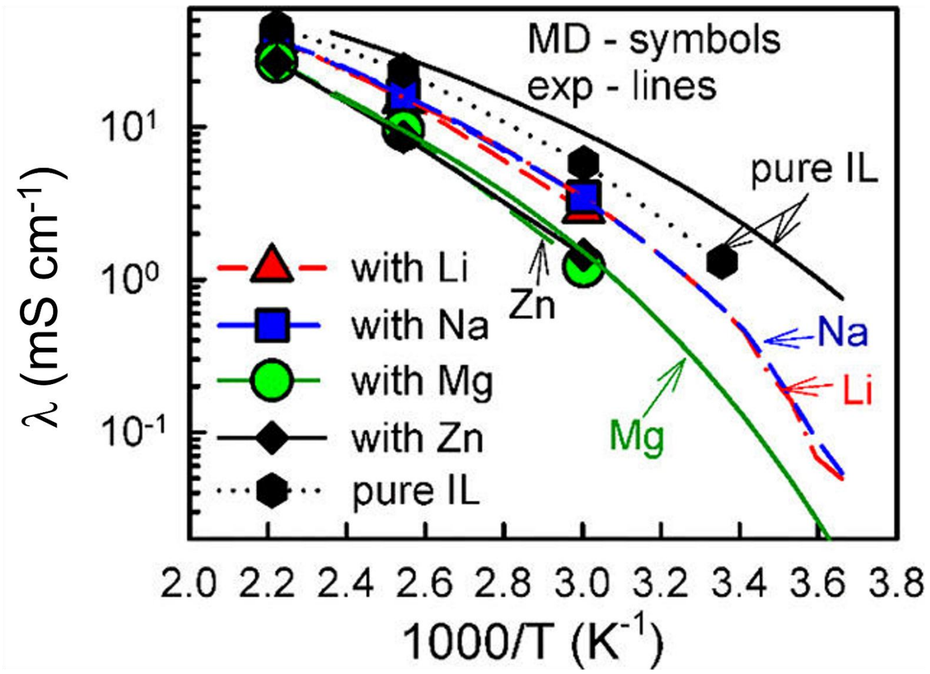
- Collaboration

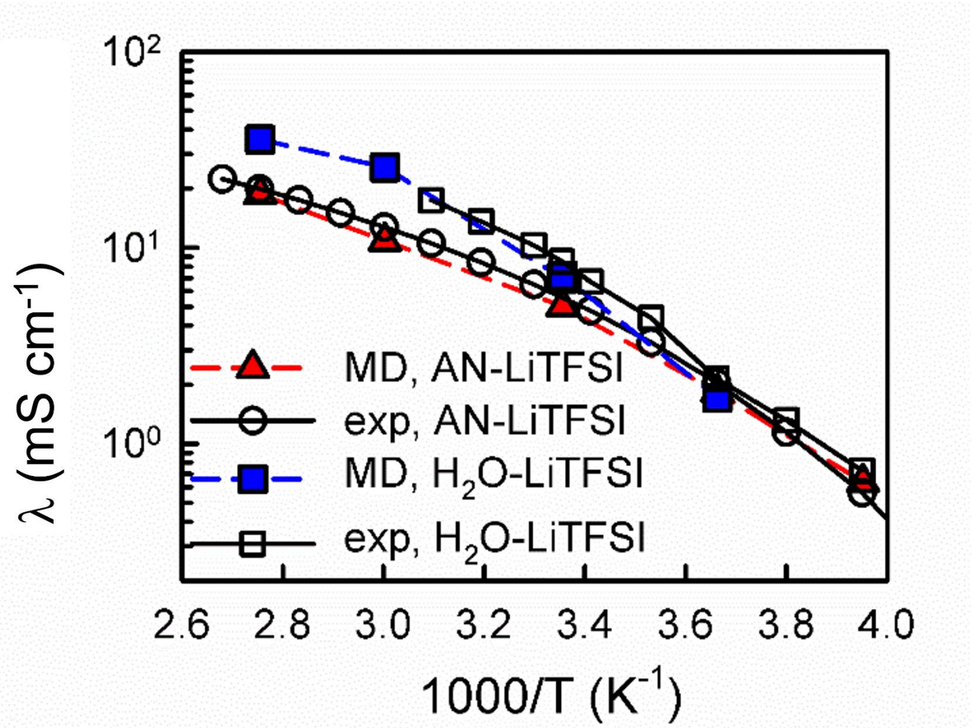


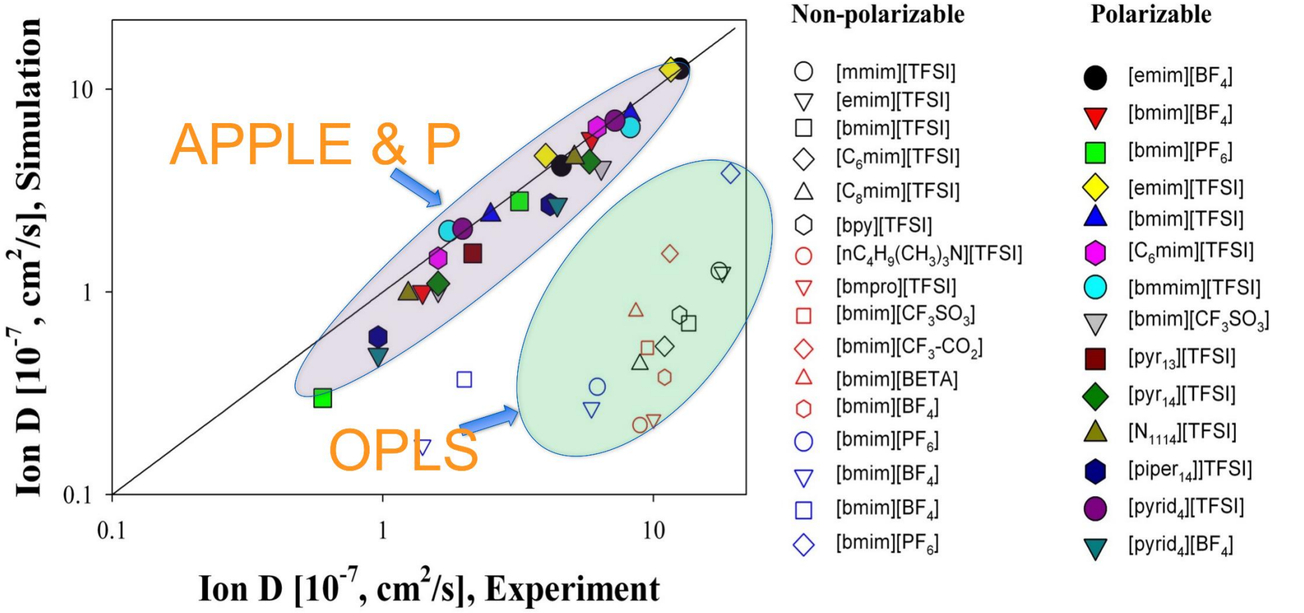
- Atomistic -> meso-scale Coarse Grained - Polarizable FF - Reactive FF

- Applications to **batteries** and **fuel cells**
  - Fuel cell membranes: Polymers 2018, 10, 1289; doi:10.3390/polym10111289

Diffusion, Conductivity with APPLE&P







Polarizable force field for batteries, ionic liquids, fuel cells

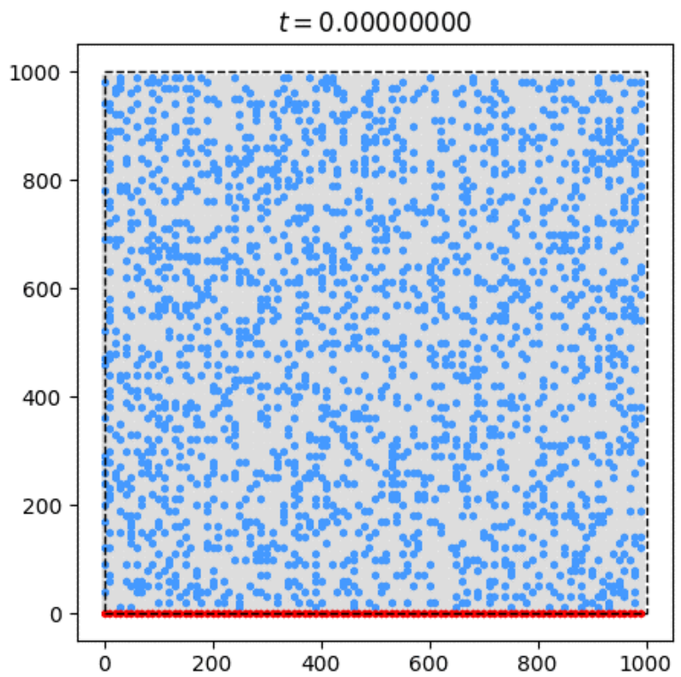
APPLE&P MD simulations: ion dynamics **within 15-20% from experiment**

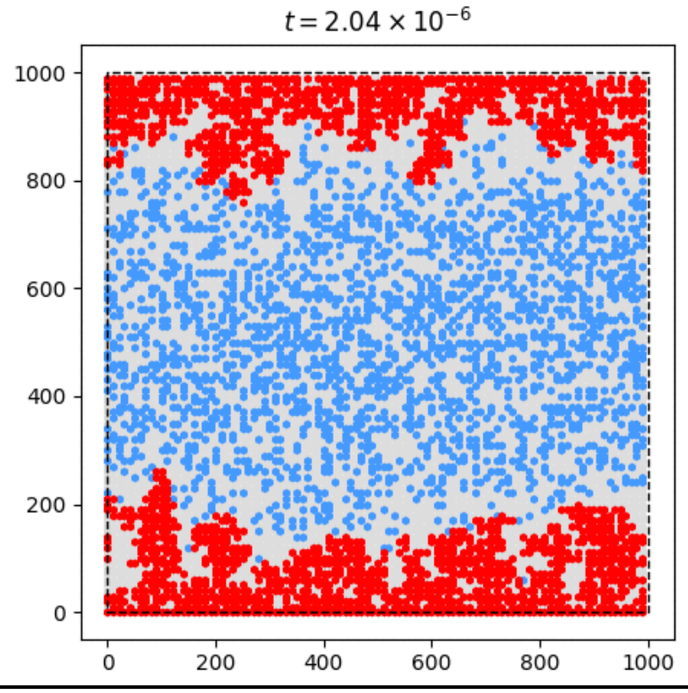
More transferable than non-polarizable force fields with rescaled ion charges

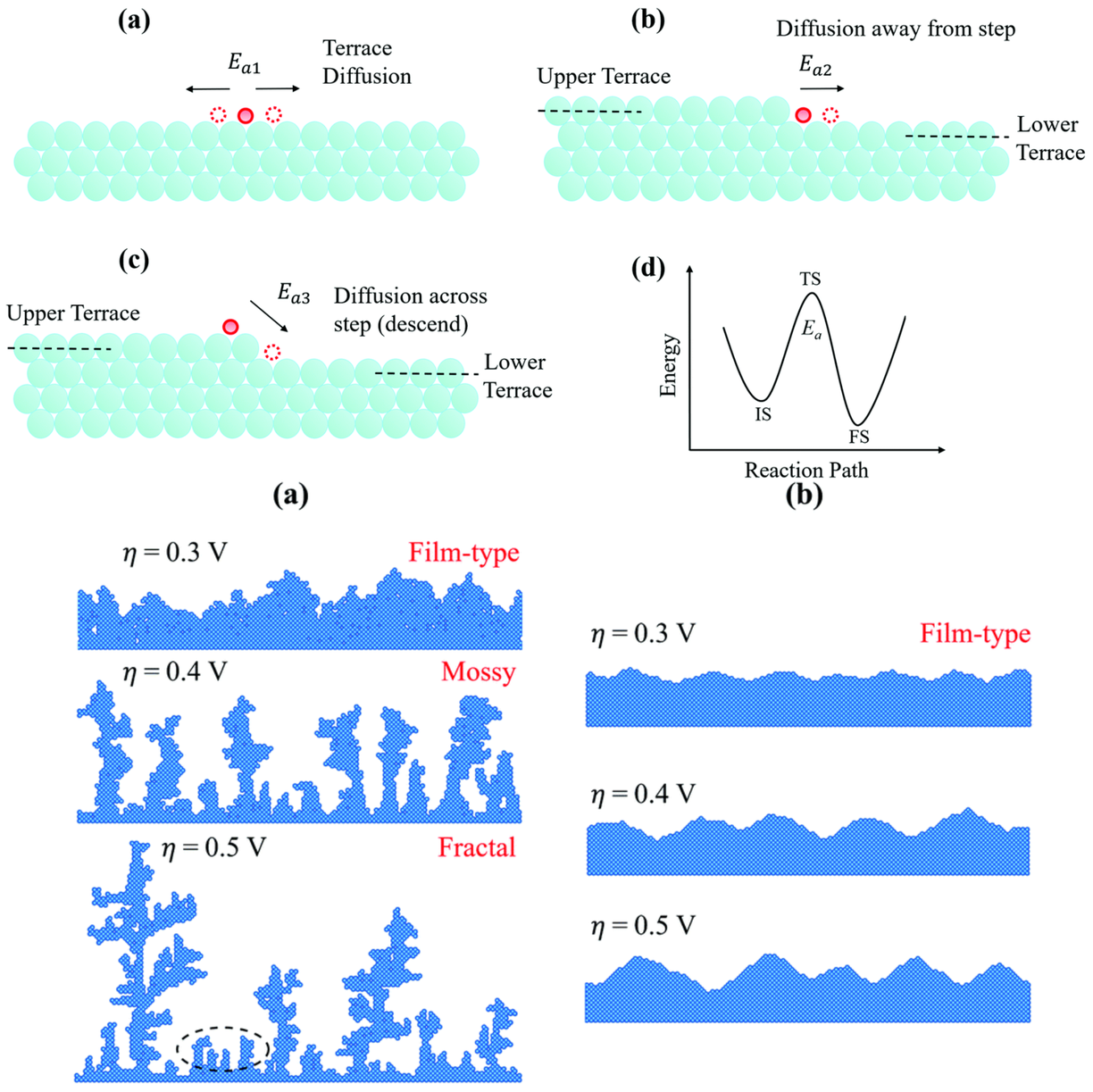
Chem. Rev. 2019, **119**, 7940

Kinetic Monte Carlo: SEI dendrite formation

Interface to Zacros kMC to study dendrite formation







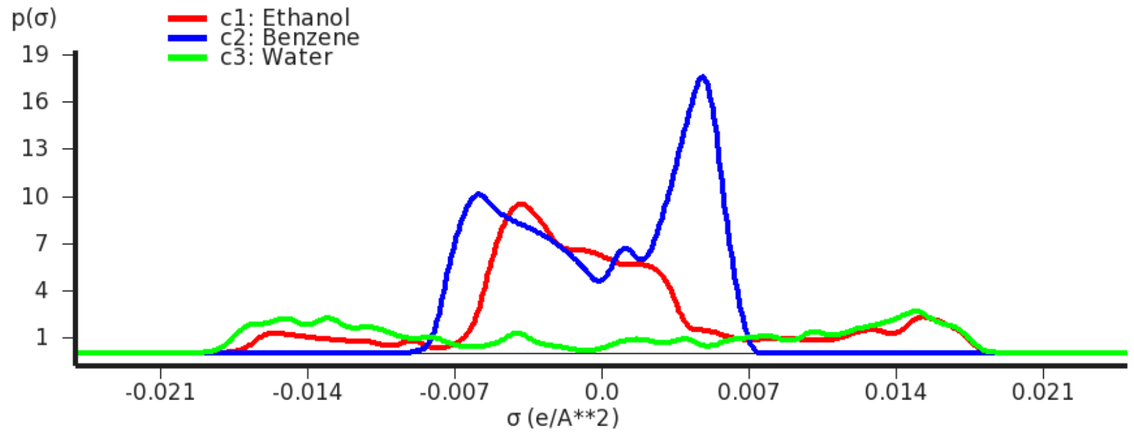
Inspired by **Surface diffusion manifestation in electrodeposition of metal anodes, PCCP 2020 (22), 11286**

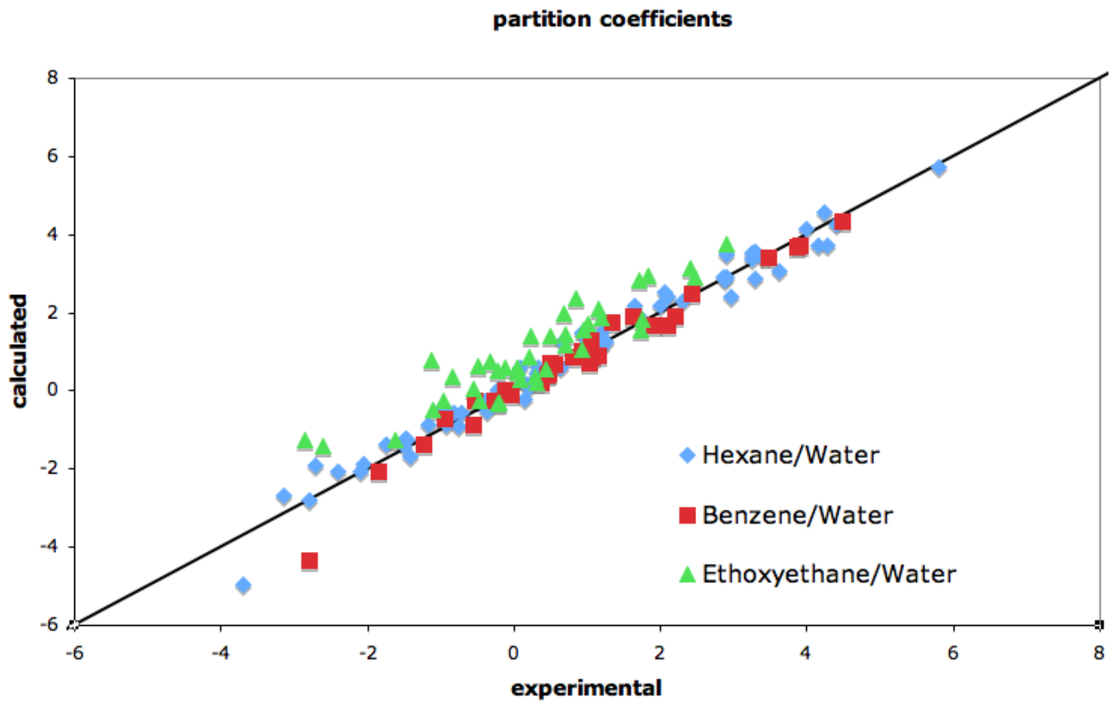
COSMO-RS/SAC: thermodynamic properties of fluids

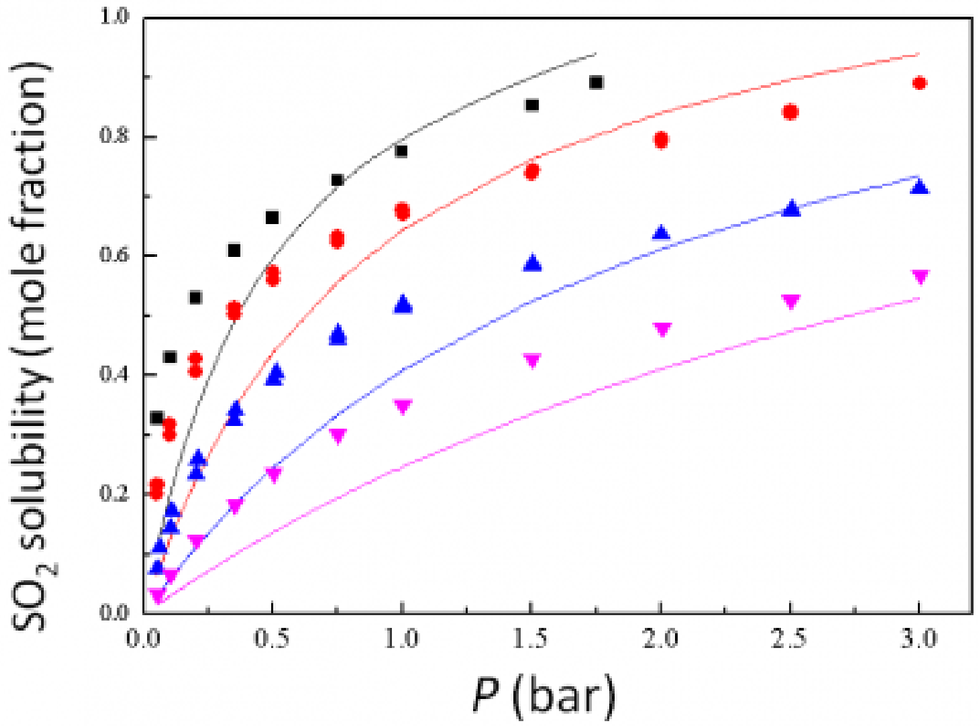
Quantum Chemistry & QSPR for quick property predictions

COntinuum Solvation MOdel + RS (Klamt), SAC (Sandler)
chemical potential => activity coefficients => instantaneous properties

- Solvation & excess energies, pKa
- Solubilities, LLE, VLE, boiling points
- Optimize mixtures: solubility, LLE
- Polymers: Flory-Huggins X







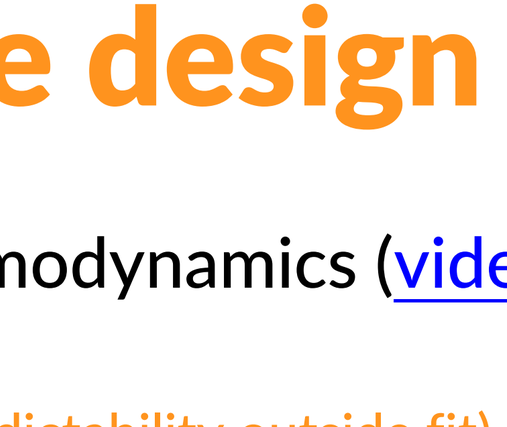
Polymer mixture design with COSMO-RS

- COSMO-RS: mixture thermodynamics (video demo)
  - Liquid-activity coefficient
  - Fit to experimental data (predictability outside fit)
  - Pseudo-chemical potential from quantum mechanics (surface charges)
- Existing design approaches focus on property targeting (QSPR)
- Mixing => requires free energies, activities

Organic electronics

- Phase stability
- Polymeric batteries

Drug Solubility

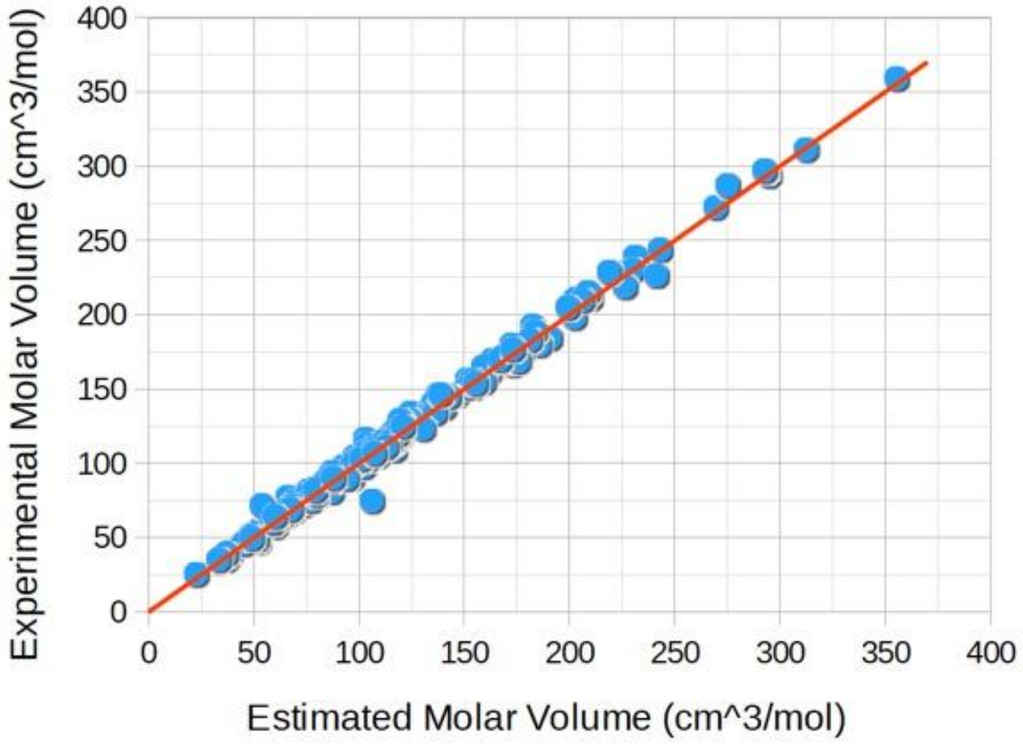


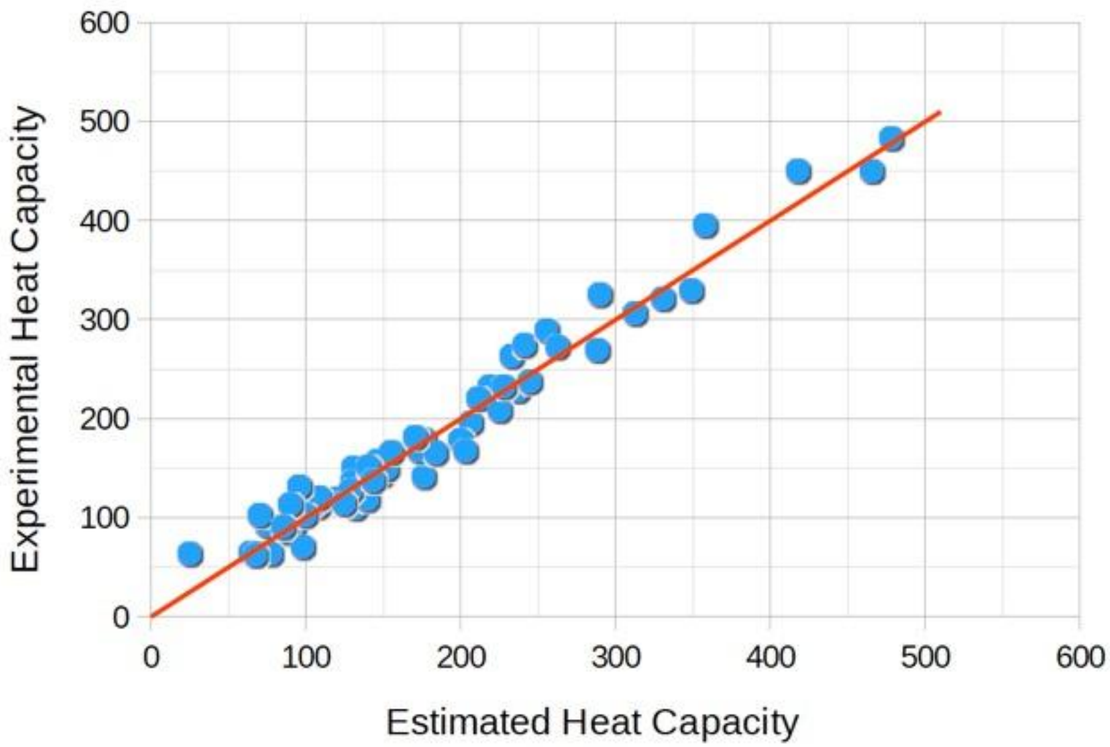
- Poorly water-soluble drugs
- Bioavailability

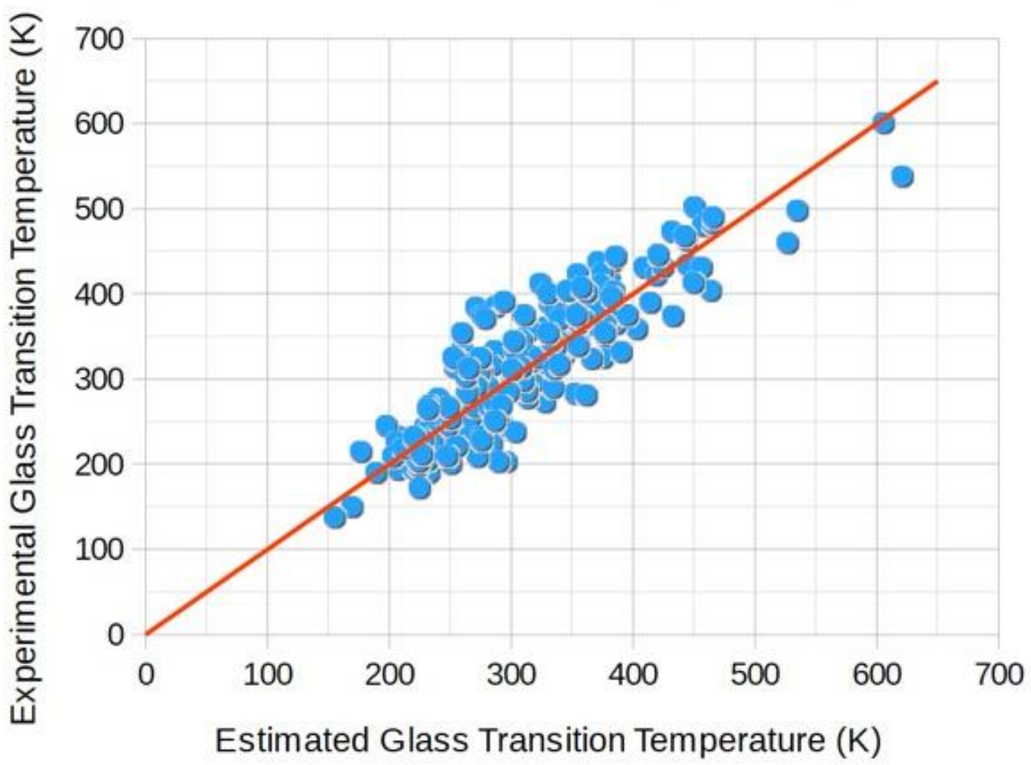
Polymer/Solvent-Polymer interactions

- Flory-Huggins parameters
- Swelling, solubility of polymer blends, etc.

QSPR with COSMO sigma-moments: polymers







Basic statistics

r^2:

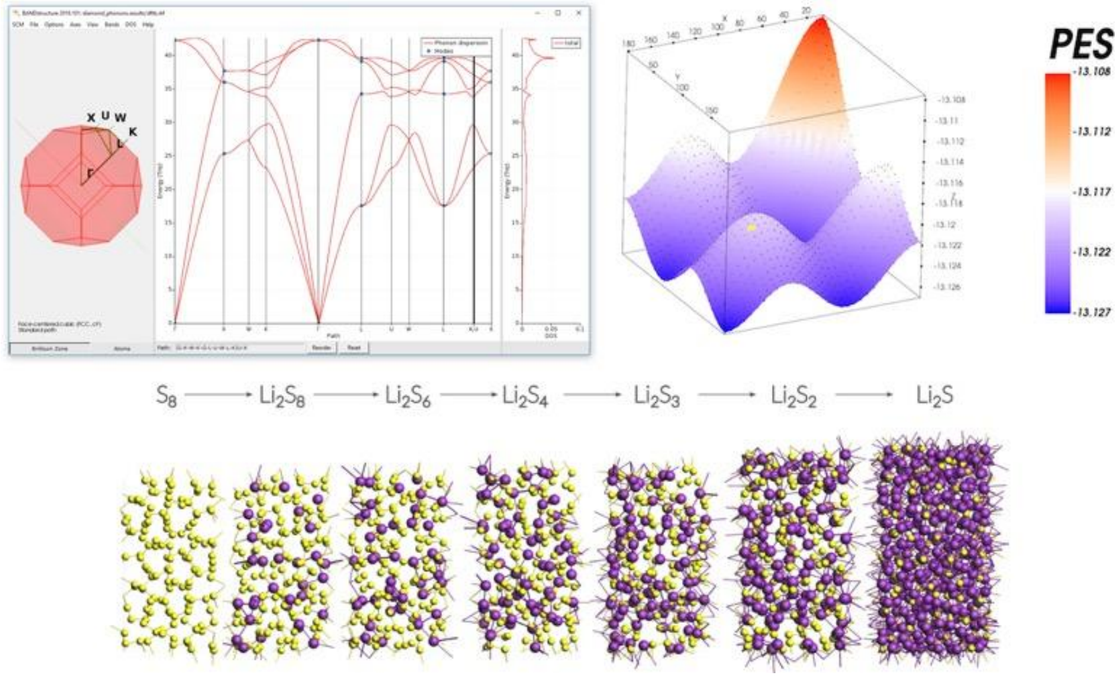
- Molar volume: > 0.99
- Heat Capacity: 0.96
- Glass Transition Temp. : 0.81

Average absolute error:

- Molar volume: 3.1 cm^3/mol
- Heat Capacity: 13.8 J/(mol K)
- Glass Transition Temp. : 45 K

Machine learning in materials science

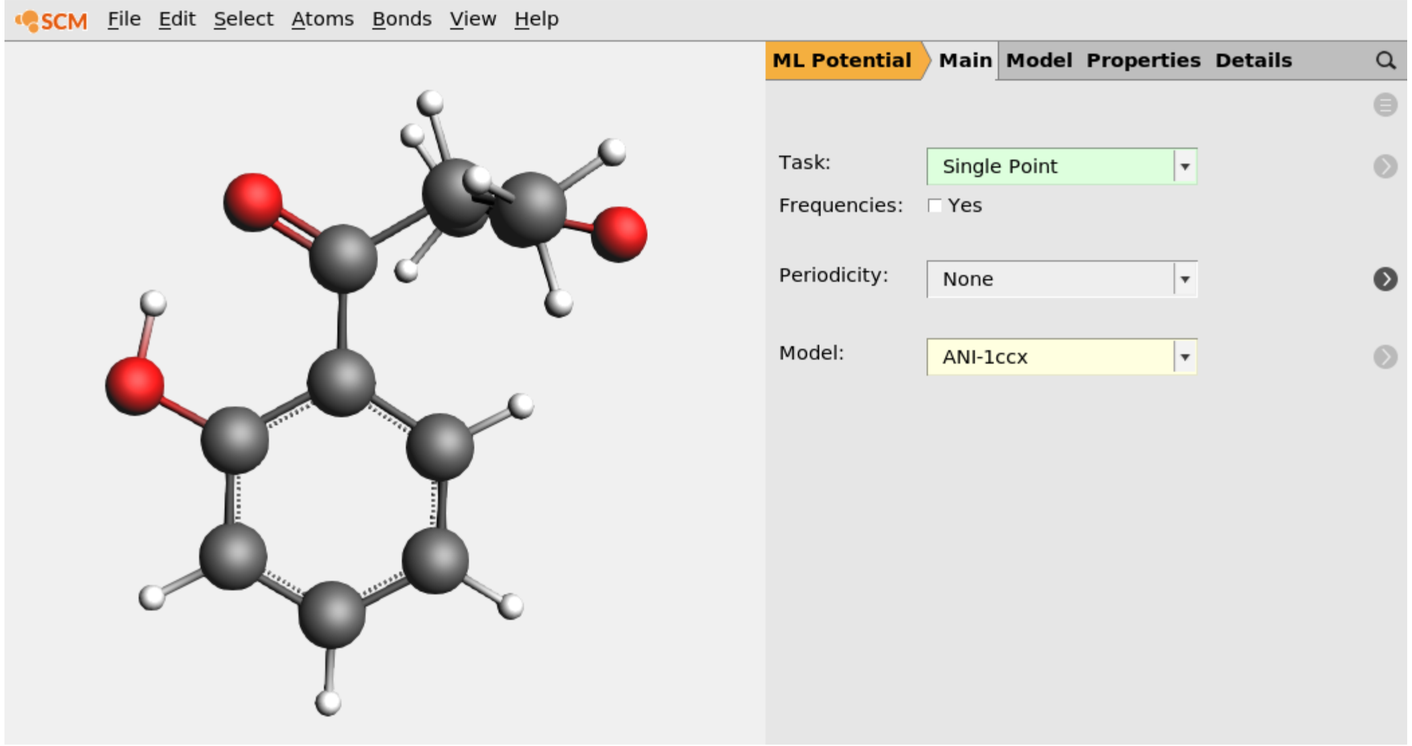
1. Direct Structure-Property prediction ‘QSPR’
2. Predict **Potential Energy Surface** (atomic coordinates)
   - Determine how atoms move in space & time
   - Predict various properties:
     - Chemical reactivity
     - Thermodynamics
     - Mechanical properties
     - Mobility rates
     - ……



3. Hybrid/other: e.g. predict reaction networks, interpolate kinetics (kMC) under different conditions

Machine Learning Potentials

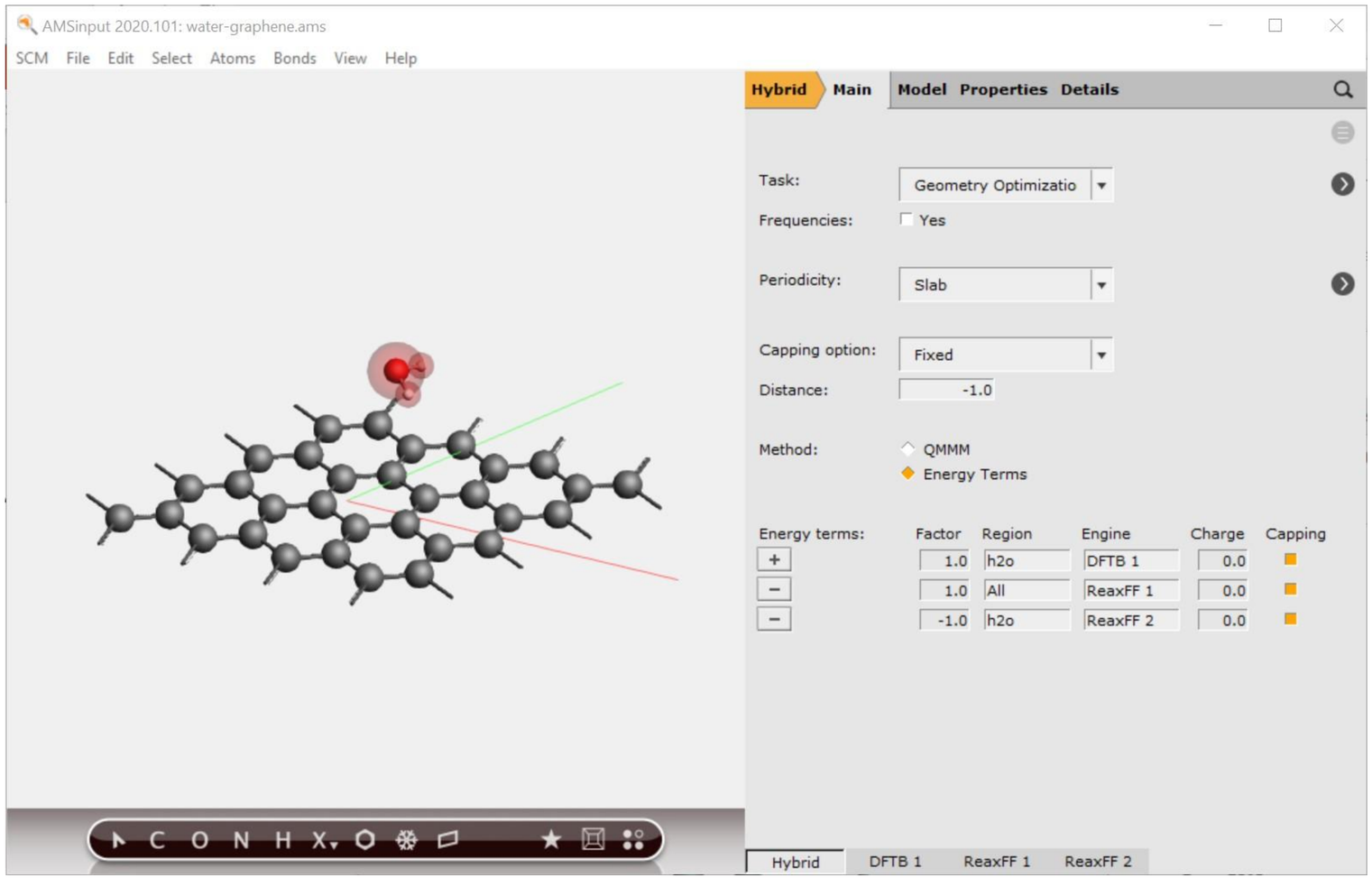
- Use machine learning potentials with AMS driver
  - PES scans, conformers, MD, reaction energies, …
- Automatically install popular ML Backends
  - SchNetPack, sGDML, PiNN, TorchANI
  - Pre-parametrized neural network potentials ANI-1ccx, ANI-2x
  - CUDA-enabled PyTorch and Tensorflow can be used
  - Development: on-the-fly ML force fields (Flare, nequip), M3GNet

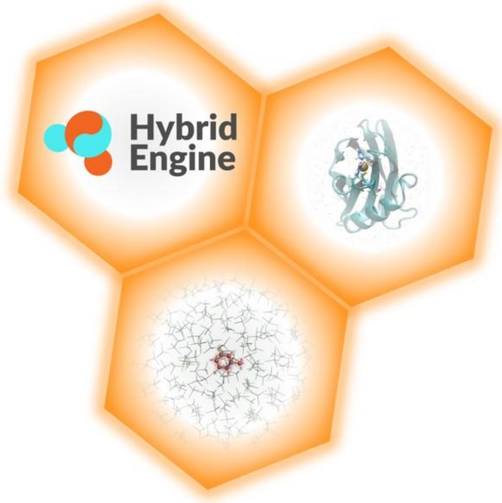


Demo video

Hybrid Engine: combine methods

- Multi-layer (subtractive, QUILD, ONIOM)
  - combine **any** periodicity, number of layers, and QM or MM methods
- 2-layer: (additive) QM/MM
  - any periodicity
  - QM: ADF, DFTB, BAND, MM: Force Field engine (ReaxFF, GAFF, MLPot)



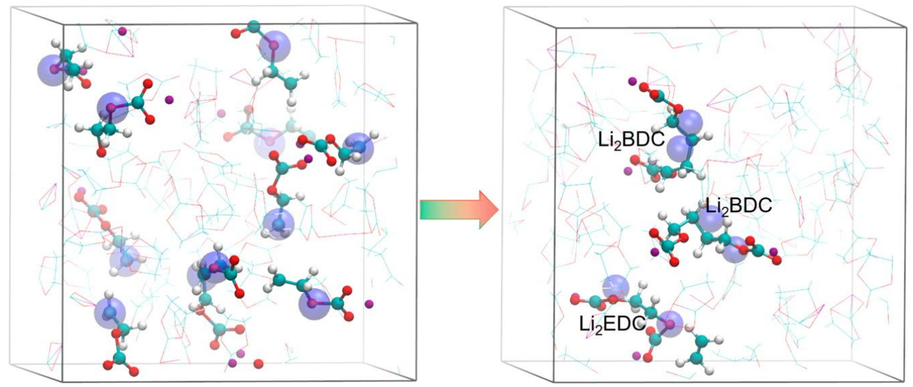


[Demo video]

Summary: SCM & Amsterdam Modeling Suite

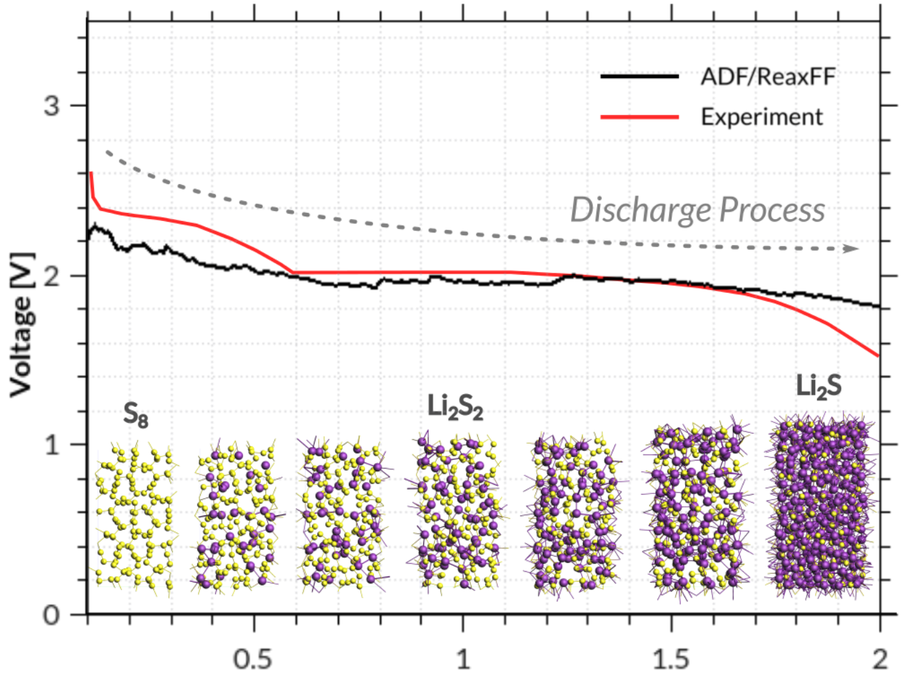
- Long history / track record computational chemistry
  - Pragmatic software development: fast, accurate, user-friendly

- Unique features, specific strengths
  - Apply different levels of theory in same suite
    - spectroscopy, dynamics, electronics, solubility



- Further expanding into multi-scale
  - Keep control & expertise + **collaborations**

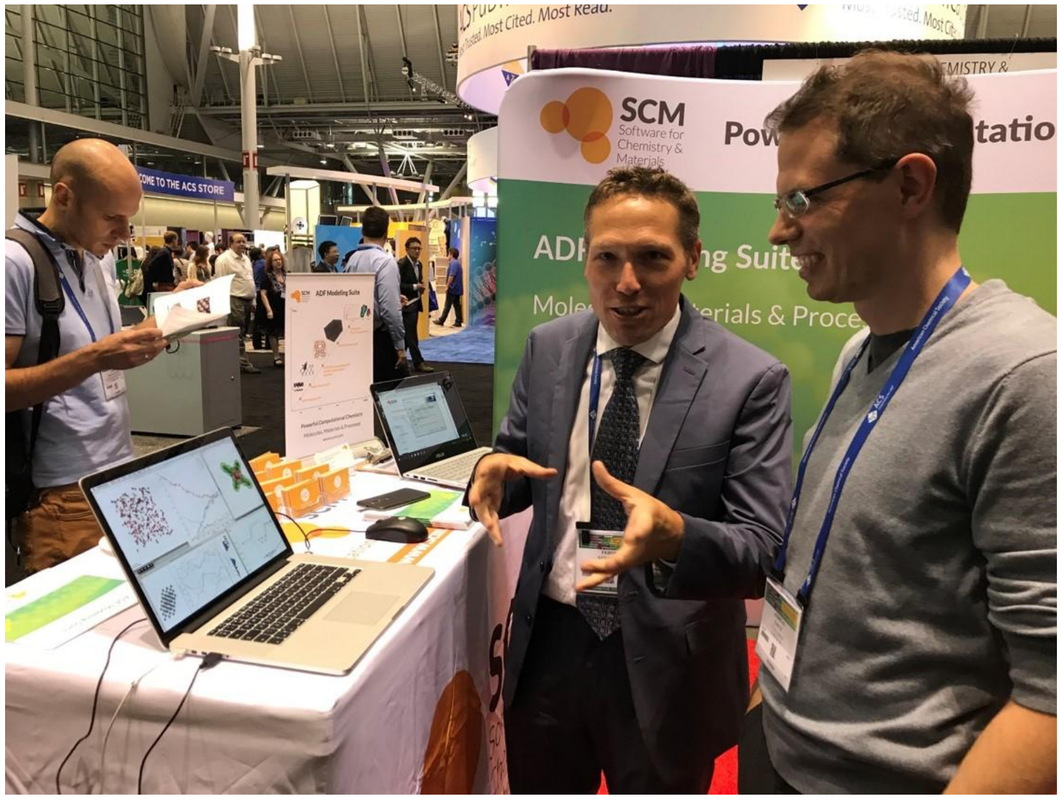
- Machine learning
  - ML potentials
  - Property prediction, accelerated optimization



Fedor Goumans, goumans@scm.com

Discuss your research challenge!

Which materials are you working on?
What are the key properties?



We love to hear from you!
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