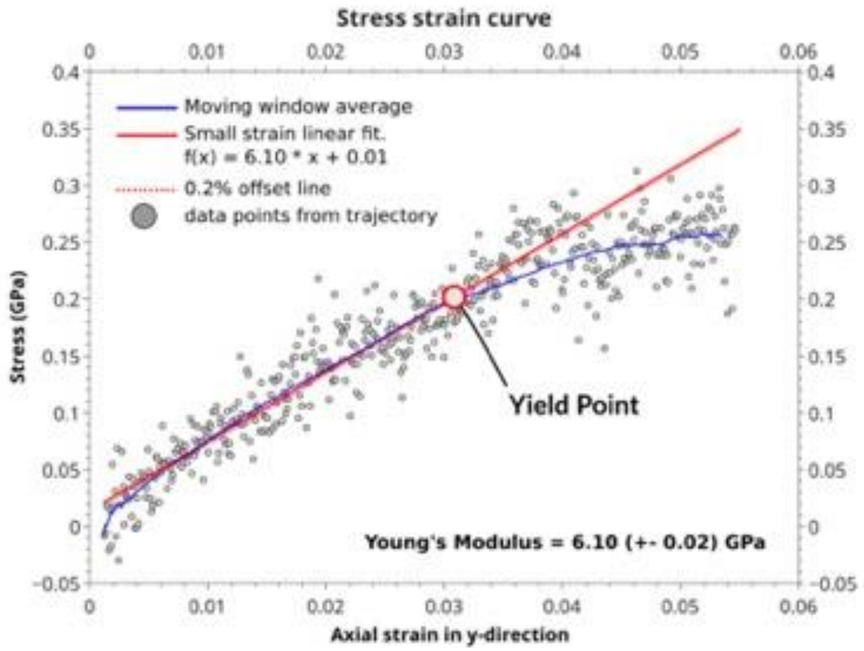
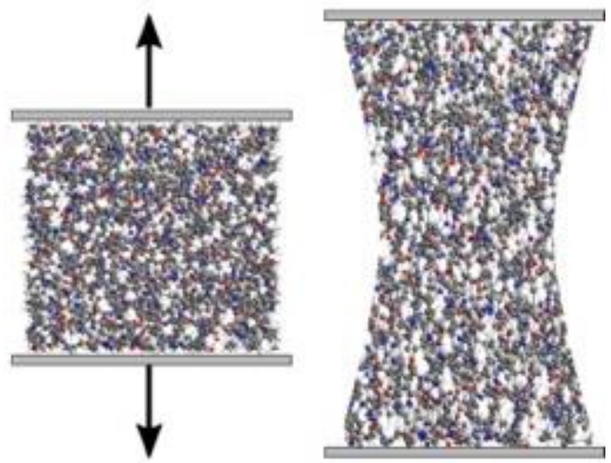
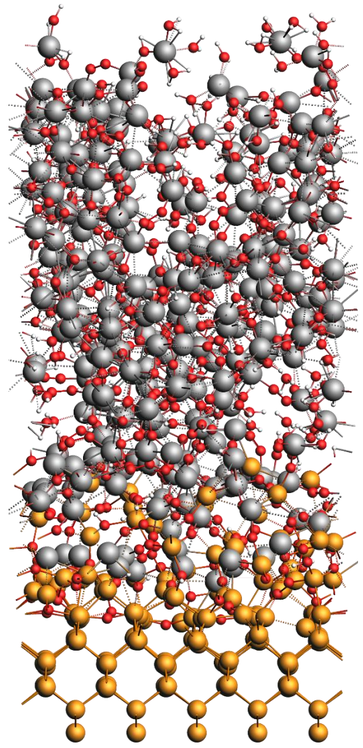
Amsterdam Modeling Suite

Atomistic & Multiscale Simulations for Chemistry & Materials







SCM
Software for Chemistry & Materials

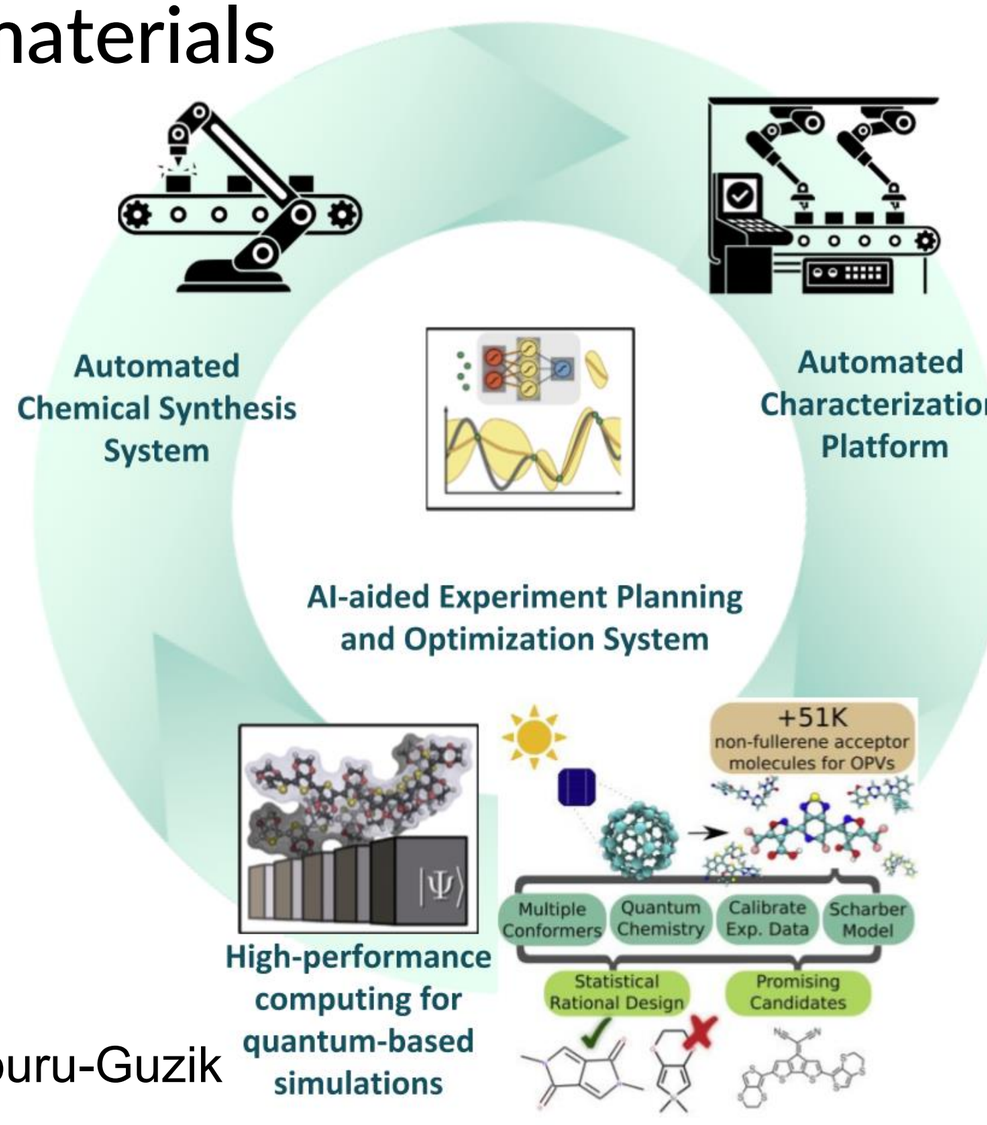
Fedor Goumans, Chief Customer Officer, goumans@scm.com

AMS Workshop, NTU, Taiwan, 28 August 2023

New materials discovery too slow

- 8-19 years to develop materials solutions in new markets
- 80-85% R&D programs fail
- >50% R&D spending only incremental improvement
- **Catch 22**: slow discovery ⇔ few new materials

https://www.mckinsey.com/industries/chemicals/our-insights/chemical-innovation-an-investment-for-the-ages



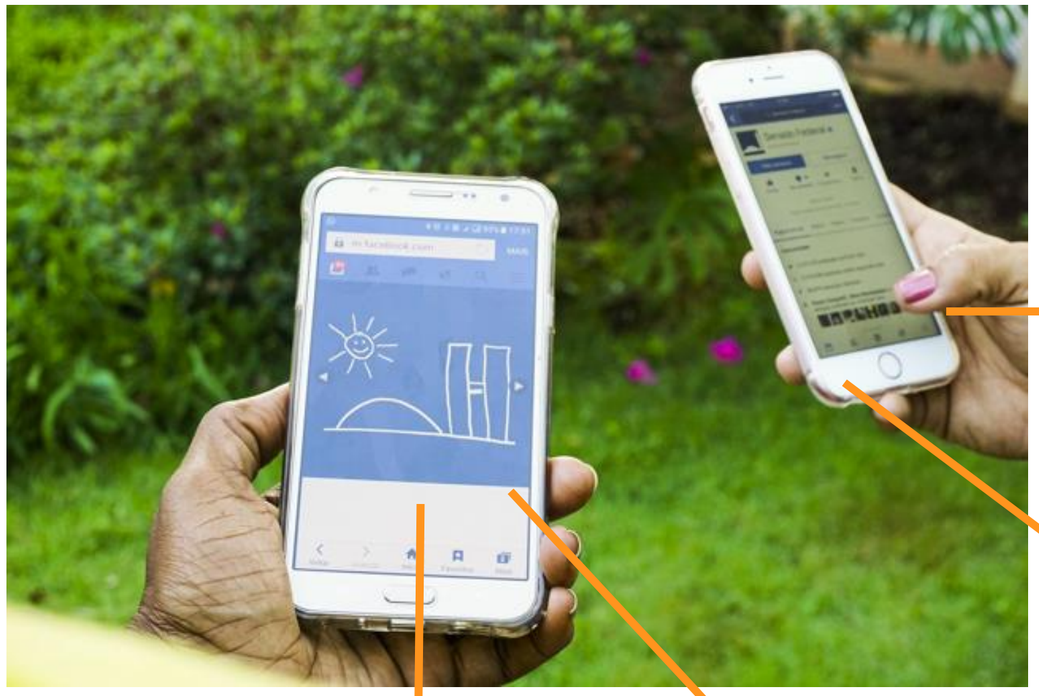
Simulations -> predict new materials
Robots -> make new materials
AI -> improve simulations and DOE

the Matter Lab - Aspuru-Guzik

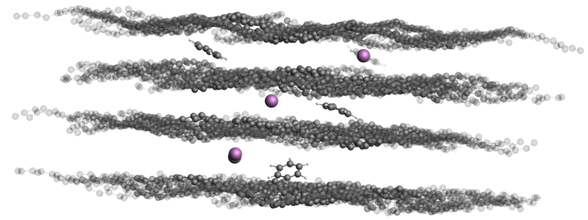
Bottom up Property Prediction

Atomistic modeling
- Decreases search space
- Develops understanding

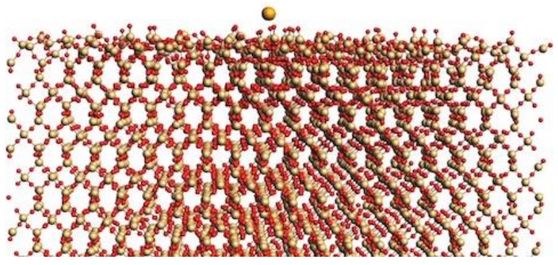
Input for meso & macro



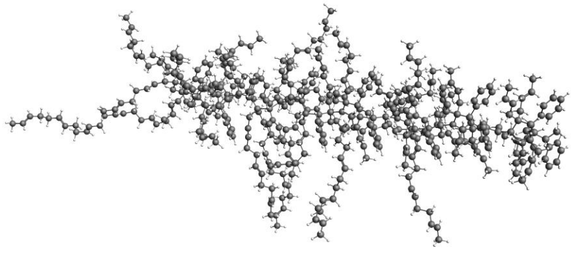
Batteries:
fast recharge,
high capacity

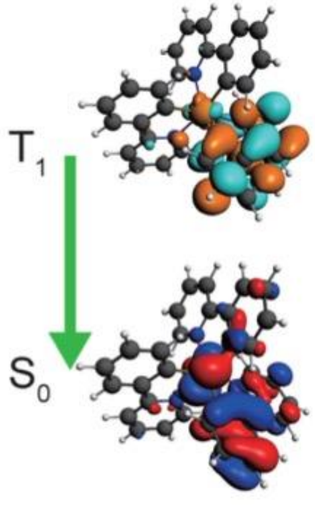


(m)CPU: high
capacity, I-V profiles



Glass, coating: optical
properties, conductivity

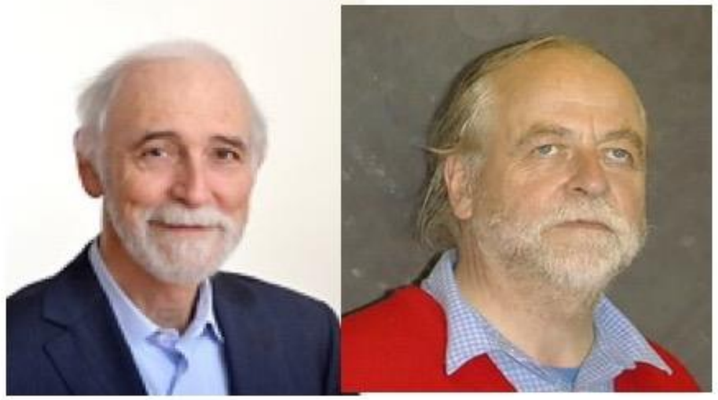


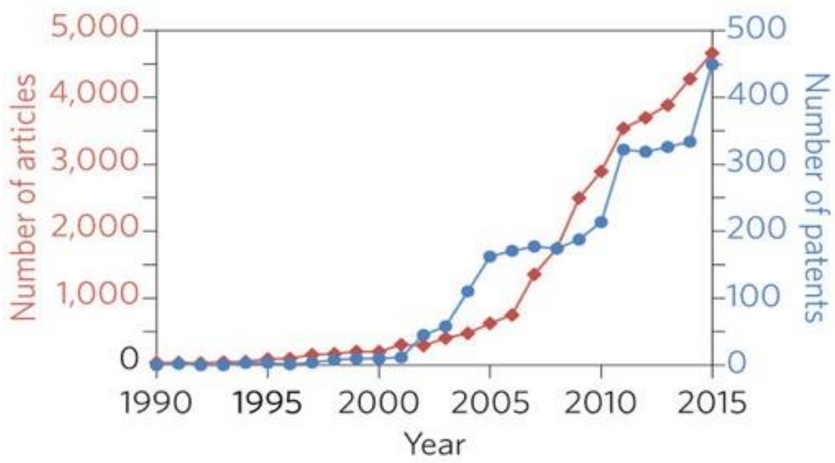


OLEDs, QLEDs: color,
lifetime & efficiency

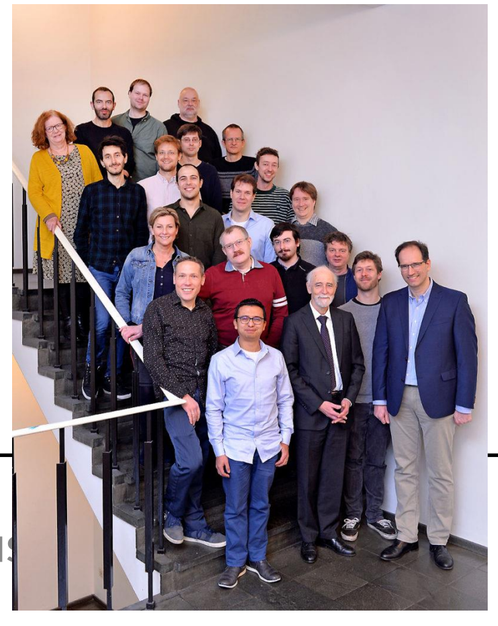
History: Sofware for Chemistry & Materials

- 1970s ADF: first DFT code to **understand chemistry**
  - Baerends@VU & Ziegler@Calgary(+)
- 1980s: Mitsui, Shell, Akzo, Unilever:
  - Training in Amsterdam -> optimize catalysts
- 1995 SCM: Spin-off company
  - Continuous support & Development
- 2010s: DFTB, ReaxFF, COSMO-RS (Albemarle, DSM)
- 2019: Multi-scale: ReaxPro (BASF, Dow, Shell, JM)
- 2023: ~30 people (19 senior PhD's)
  - Many collaborations nonprofit & industry
  - SCM: development, debug, port, optimize, & support



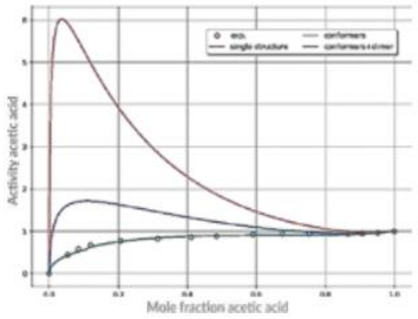


articles &patents in materials science with "density functional theory", Nat. Mat. 4619

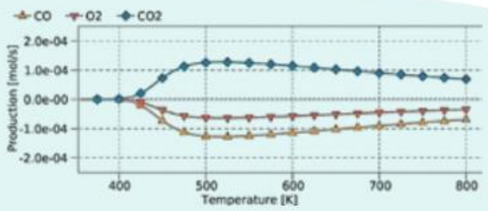


Amsterdam Modeling Suite

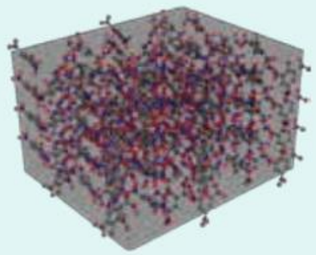
Continuum



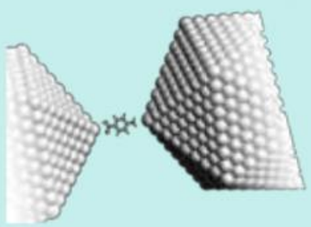
Mesoscale



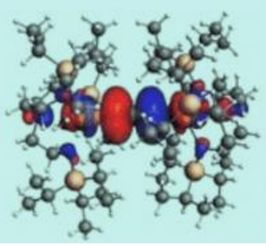
Materials



Nano



Atomistic



Fluid Thermodynamics
COSMO-RS
COSMO-SAC
UNIFAC

Kinetics
Kinetic Monte Carlo
Microkinetics

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

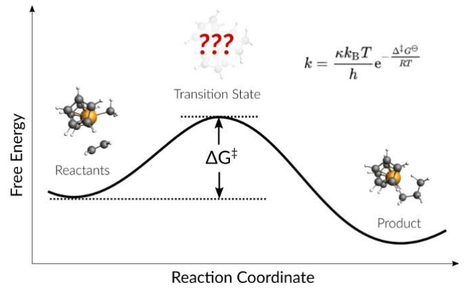
QM/MM
FDE, Hybrid Engine

Tight binding
GFN-xTB, DFTB

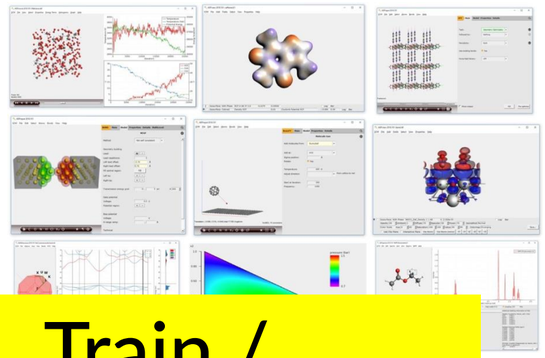
Periodic DFT
BAND, Quantum Espresso

Molecular DFT
ADF

AMS Driver: PES exploration, MD, MC, TS, IR, phonons, …

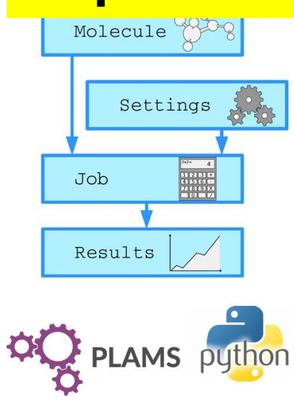


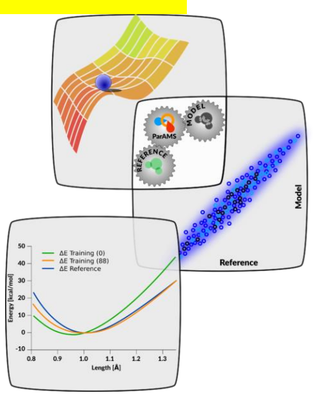
Integrated GUI, remote jobs



Py parametrize

Train / parametrize

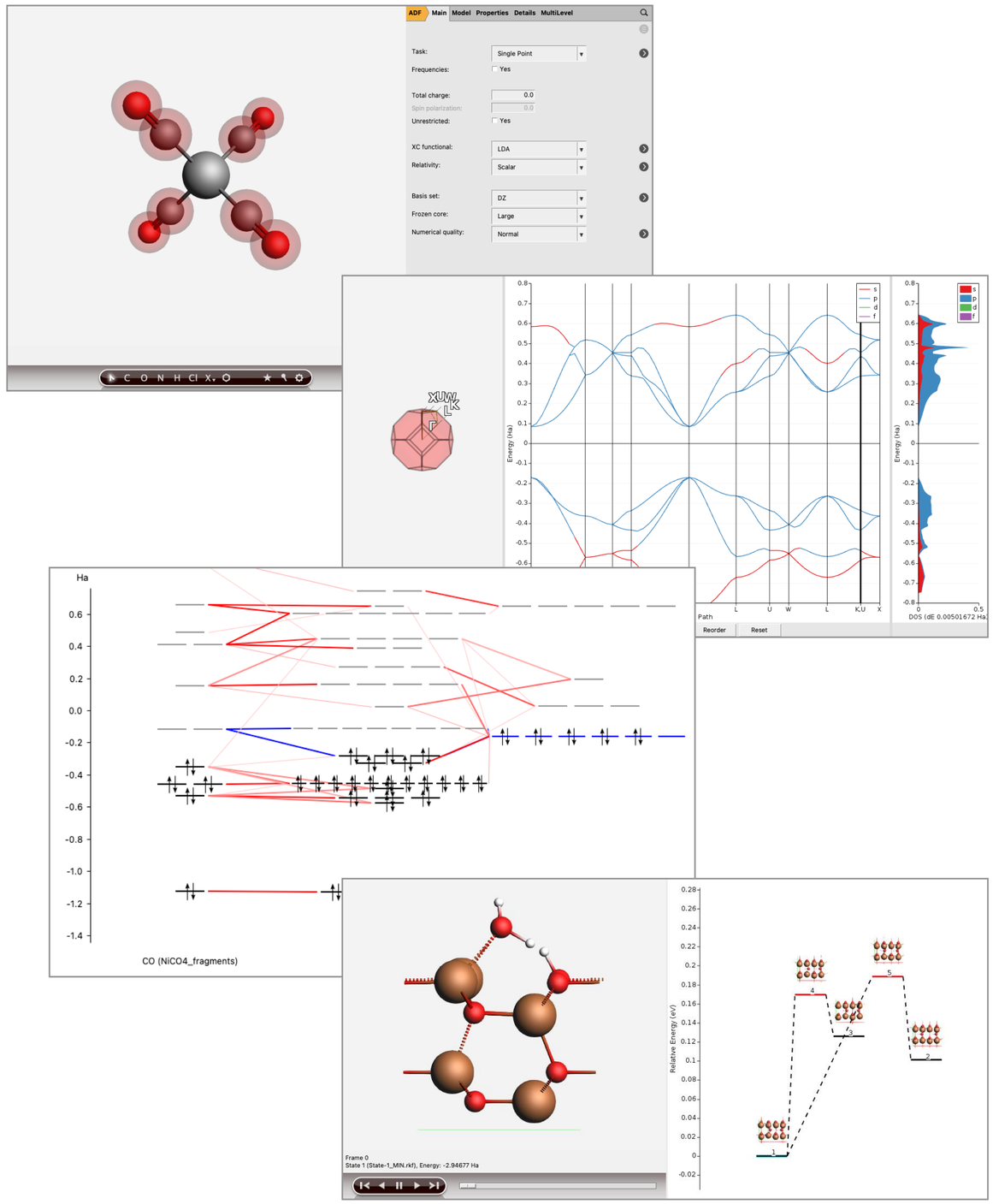




The graphical user interface (GUI)

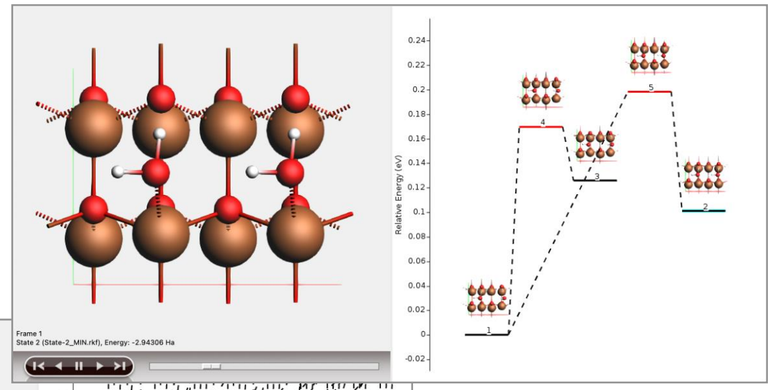
Setup & analyze calculations

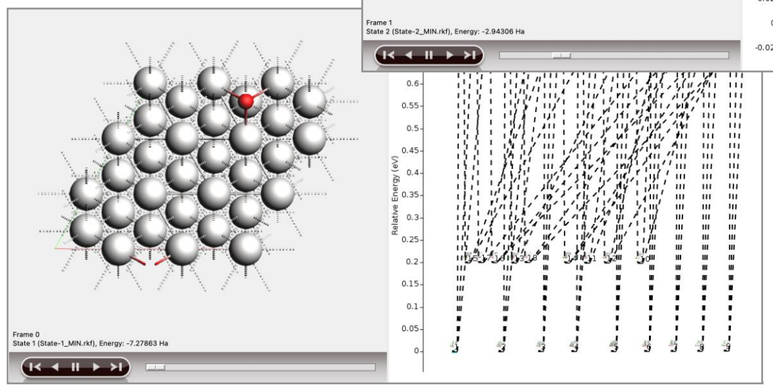
- AMSjobs
  - manage jobs, locally or remotely
  - extract summaries
- AMSinput
  - build molecules, periodic systems, polymers, etc.
  - import structures from many formats
- AMSview, AMSlevels, AMSspectra, etc.
  - analyze results
  - visualize structures, transition states, orbital densities, DOS, band structure, spectra, etc.



AMS driver: MD with ‘anything’

A unified driver to explore the Potential Energy Surface (PES)





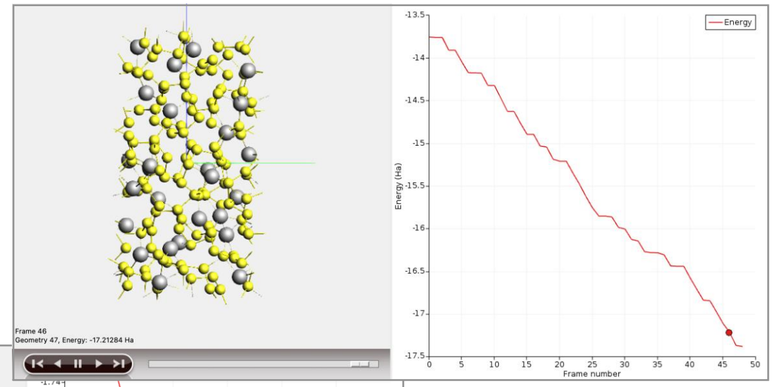
Tasks

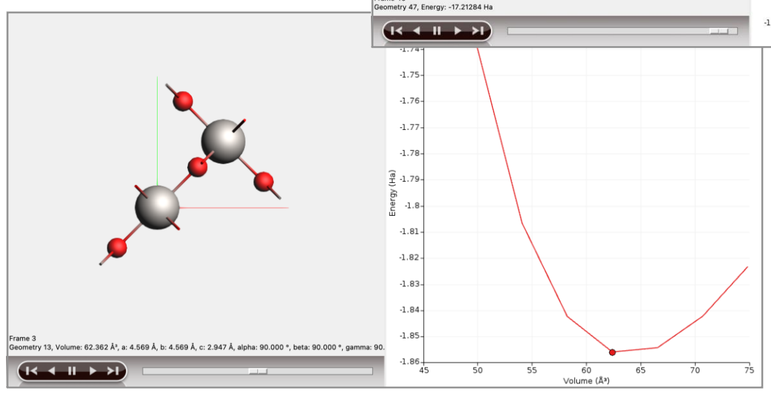
- Molecular dynamics
- Frequencies & phonons
- Stress & elastic tensors
- Scan coordinates & constraints
- Monte Carlo, etc.

AMS driver

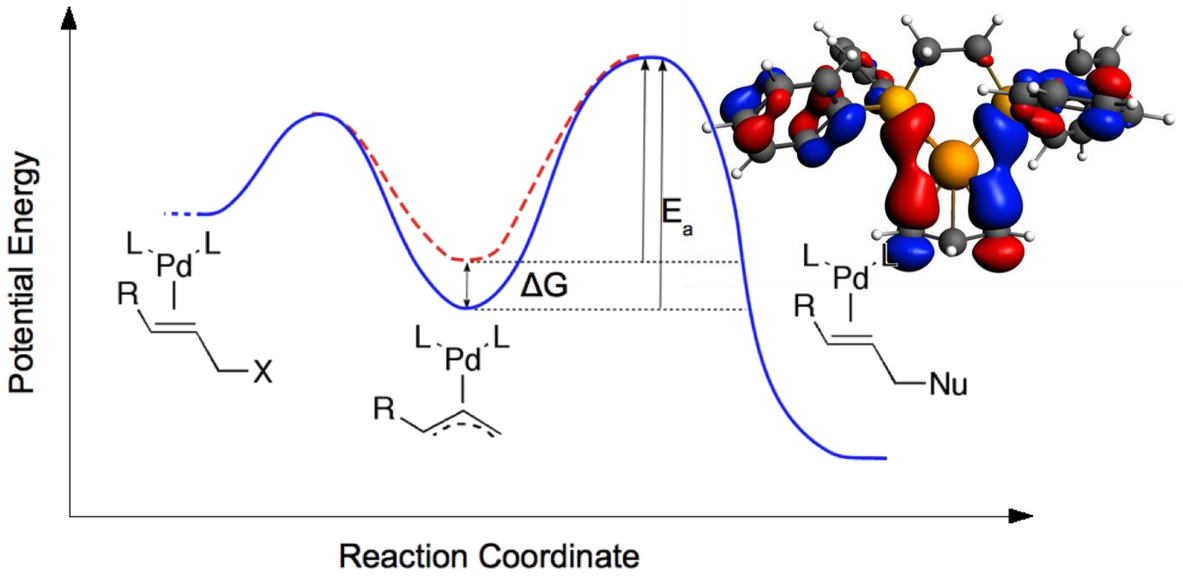
Engines

- ADF
- BAND
- DFTB
- ReaxFF
- MLP

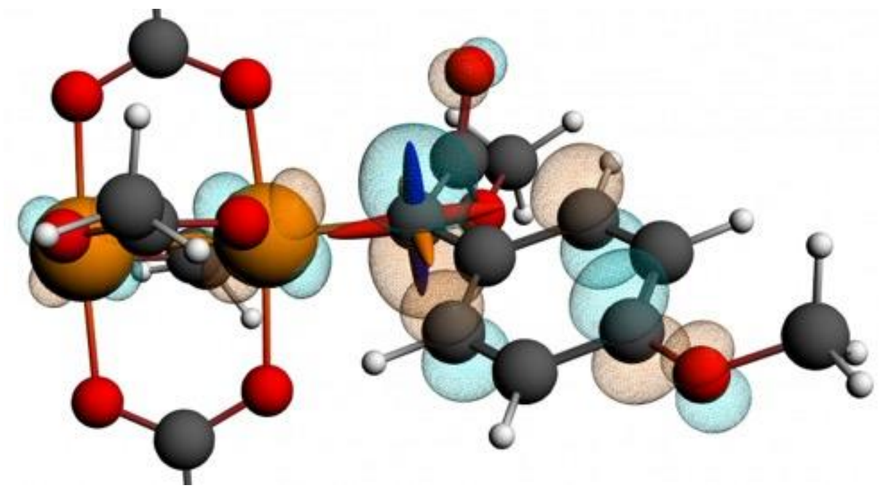




ADF: Molecular DFT



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, **all elements**
- ZORA & X2C scalar relativistic
- **Spin-orbit coupling**
- Fast GW, BSE, double hybrids

Spectroscopy

- EPR, NMR, IR, Raman, UV/VIS, XANES,

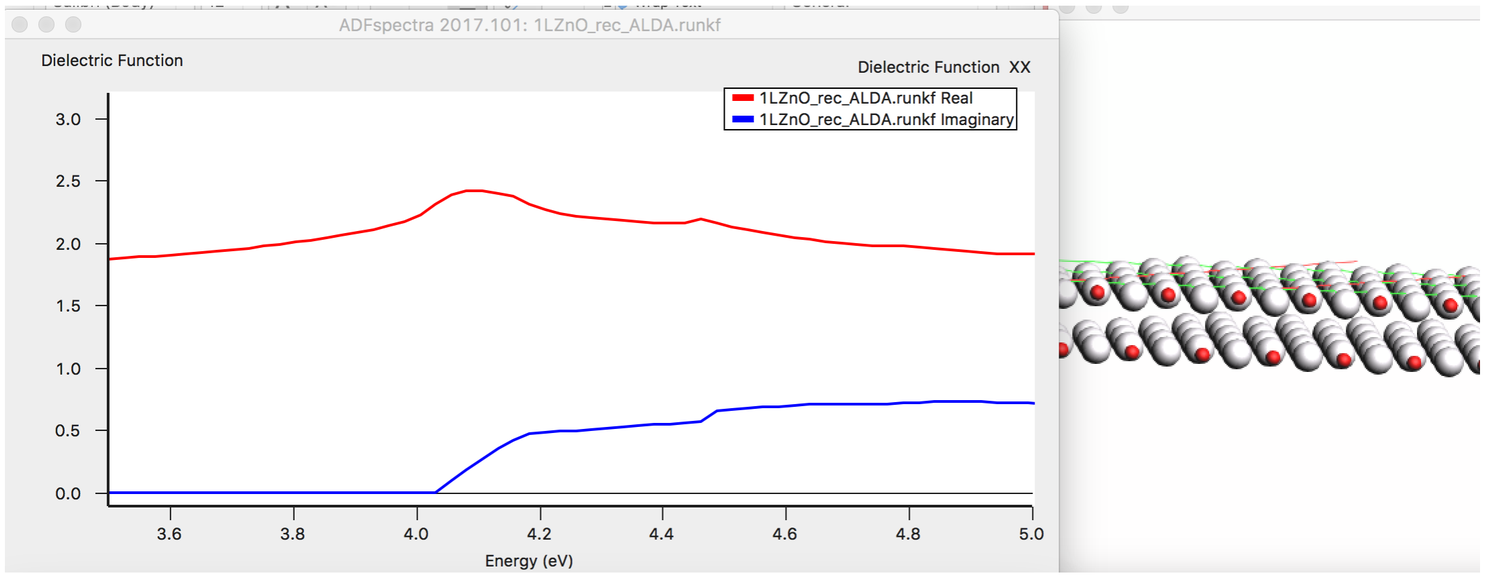
Bonding analysis

- ETS-NOCV, QTAIM, MO diagrams, NCI
- NTOs, CT descriptors
- TDM between excited states
- Transfer integrals

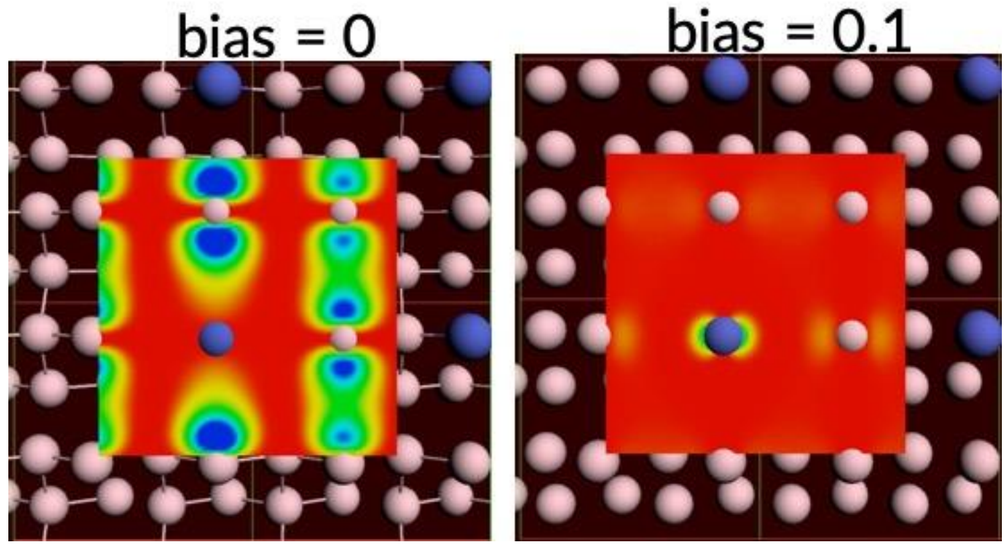
Environments

- COSMO, SM12, 3D-RISM
- Subsystem DFT (FDE)
- DRF, SCRF, DIM/QM, QM/FQ, multi-layer

BAND Periodic DFT with AOs

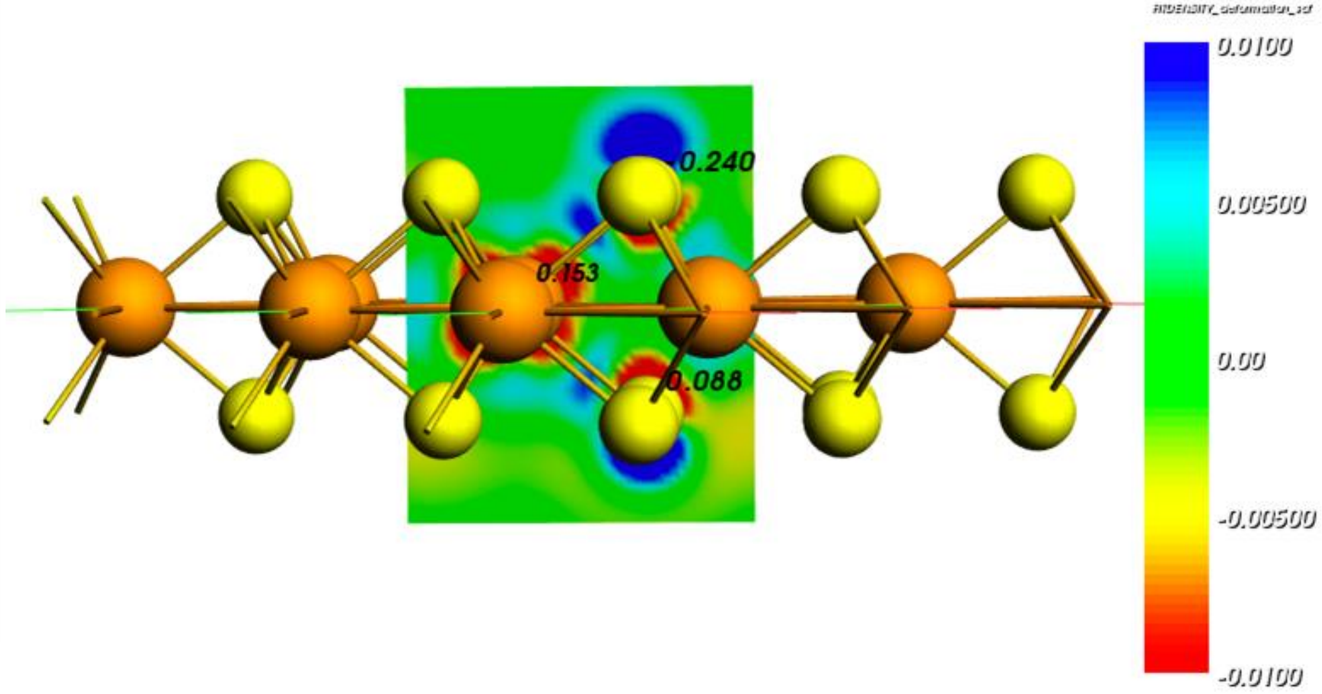


Dielectric function ML ZnO (2D TDCDFT)



LDOS: STM PtGe(100)

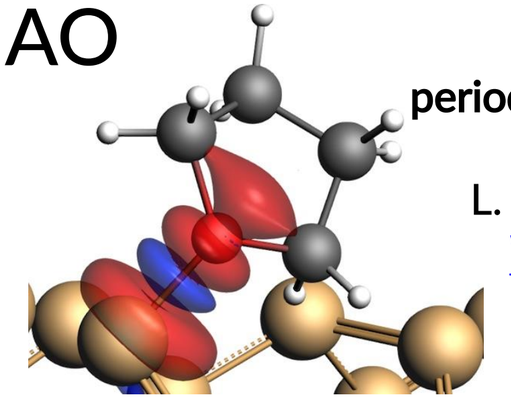
Band structure, pDOS, fat bands ZnS



Polarizing MoS_2 with an electric field

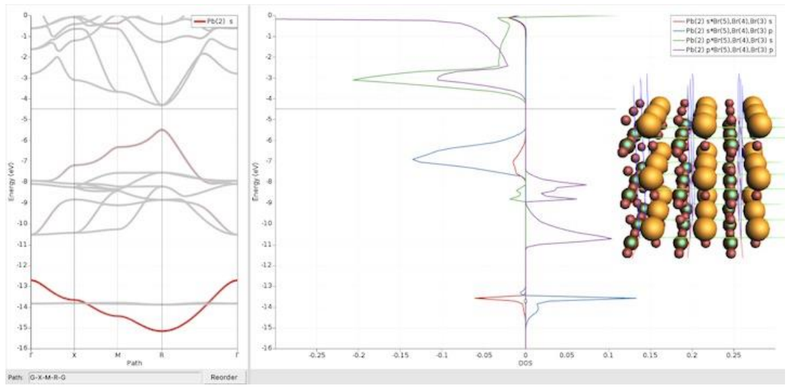
BAND + Plane Wave codes (QE)

- Atom centered basis functions, STO + NAO
  - Compare cluster with periodic
  - No pseudopotentials, basis sets for all elements
  - Core spectroscopy (core holes)
  - Easy orbital **analysis:** pDOS, COOP, EDA
  - Dielectric function: polarization functional, TDCDFT
  - xc: TASK, SCAN, MN15-L, HSE06, GLLB-sc, D3(BJ), DFT-1/2, r2SCAN-D4
  - Self-consistent spin-orbit coupling
  - Self-consistent NEGF (biases, spin)



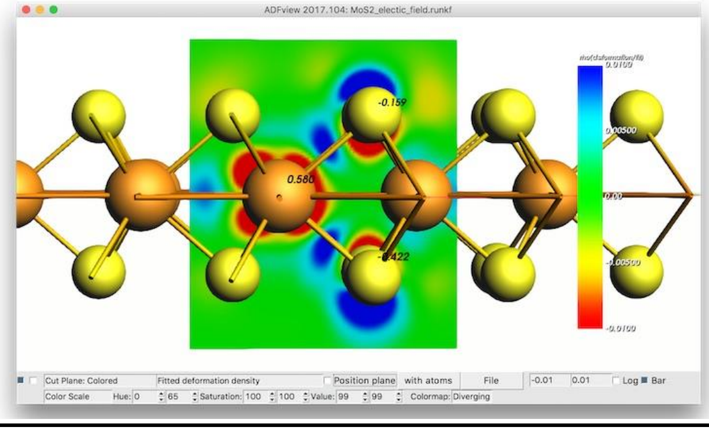
periodic energy decomposition analysis (tutorial)
L. Pecher and R. Tonner
WIREs CMS, (2018)

- True 2D surfaces, 1D polymers
  - Solvation: COSMO, **SM12**
  - Polarization in z-direction
  - 1D, 2D electronics (homogeneous E field)
  - Nanotubes



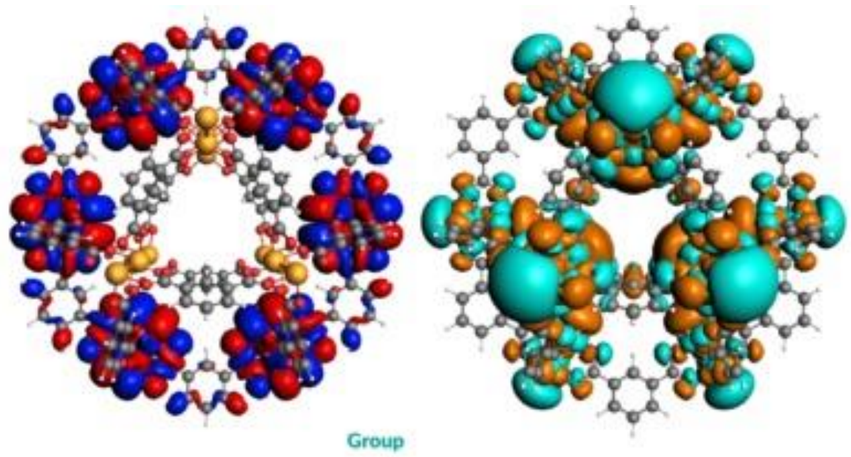
COOP in perovskites (tutorial)
Goesten & Hoffmann
JACS (2018)

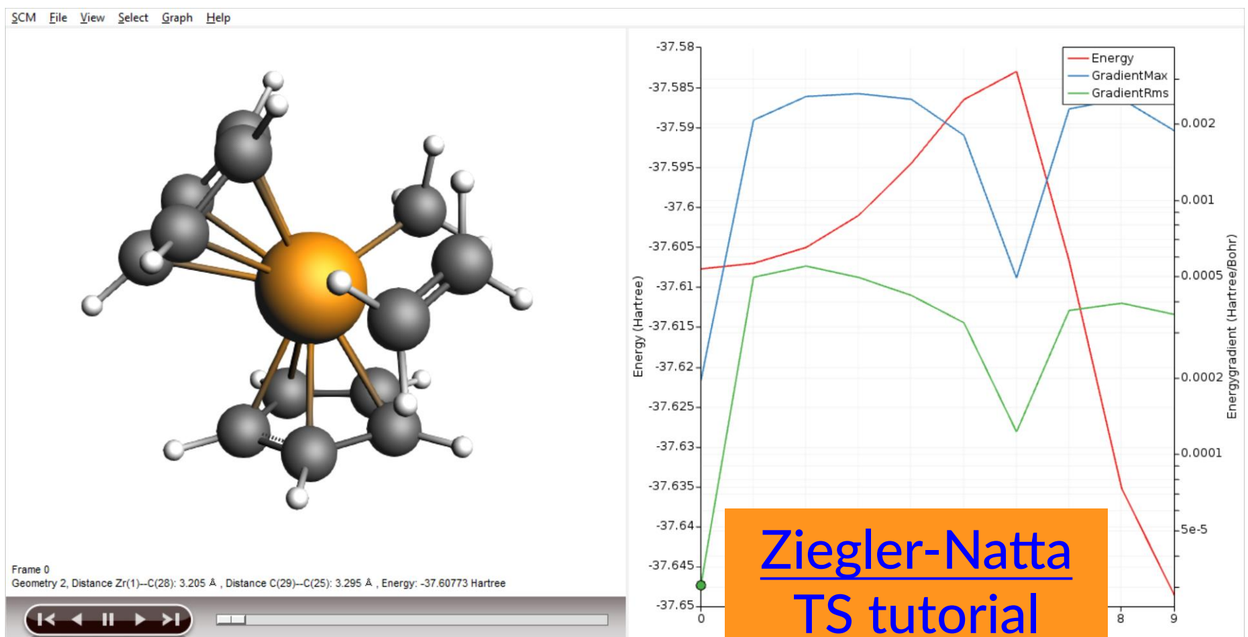
- Integrated GUI + Python I/F
  - Easy set up & analysis
  - Switch: ADF, DFTB, BAND, QE (VASP)
  - Parametrize DFTB, ReaxFF, MLP



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

DFTB: ‘fast DFT’ for molecules & periodic





[Ziegler-Natta TS tutorial]

Approximated DFT

- Nearest neighbor & minimal basis
- Tabulated elec & rep. parameters:
  - Grimme GFN-xTB (Z = 1-86)
    - GBSA solvation
  - QuasiNaNo & DFTB.org

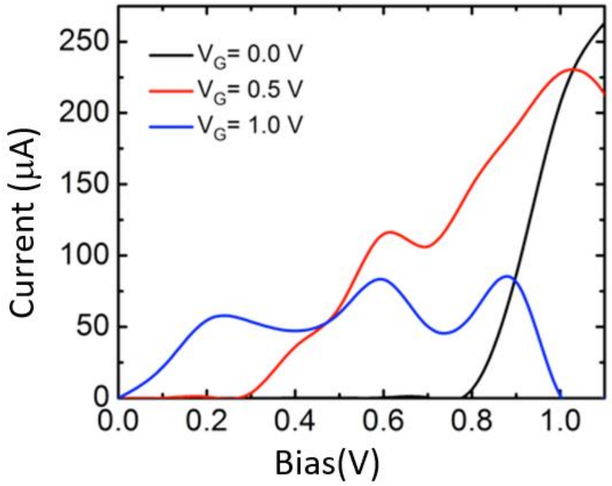
Capabilities & Features

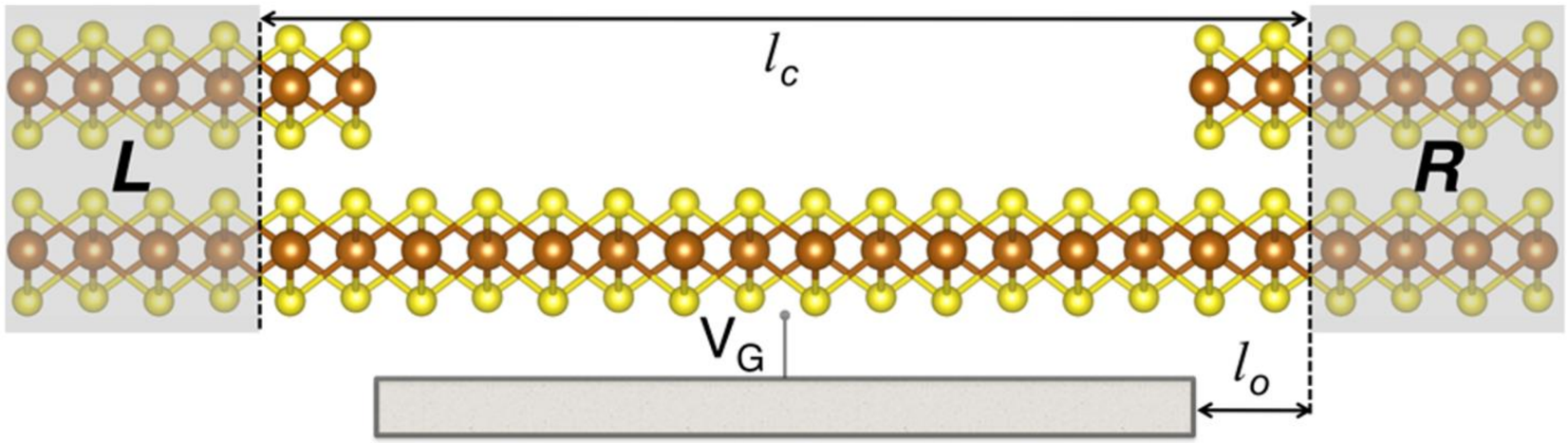
- UV/VIS for molecules (fast!)
- MOs, Band structures, DOS
- Molecules, 1D polymers, bulk
- NEGF: transmission

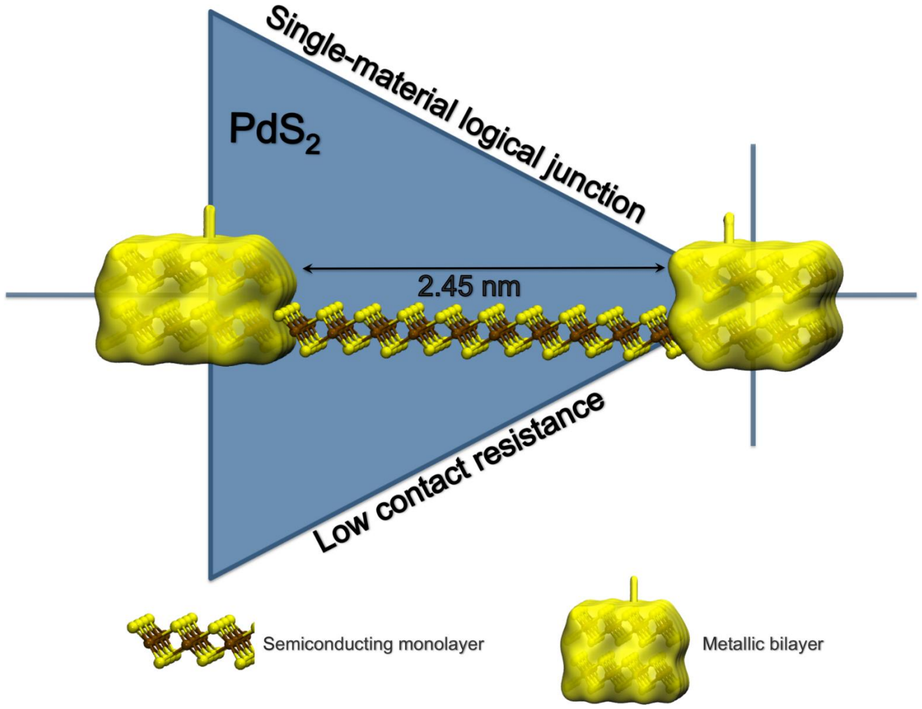
DFTB & MOPAC + AMS driver

- Geometries, frequencies, phonons
- Stress tensors (optimize under p)
- Advanced MD, PES scans
- GCMC, molecule gun (deposition)

Single 2D semiconductor material diode







2D PdS_2 is

- semiconducting as ML
- semimetallic as BL

DFTB-NEGF

- channel lengths of about 2.5 nm
- I–V characteristics tunable by gate voltage

M.Ghorbani-Asl, A. Kuc, P. Miró, and T. Heine, *A Single-Material Logical Junction Based on 2D Crystal PdS2*, Adv. Mater. (2016)

ReaxFF – reactive molecular dynamics

- Single atom type, bond-order based
- Charge equilibration
- Continuous F & E

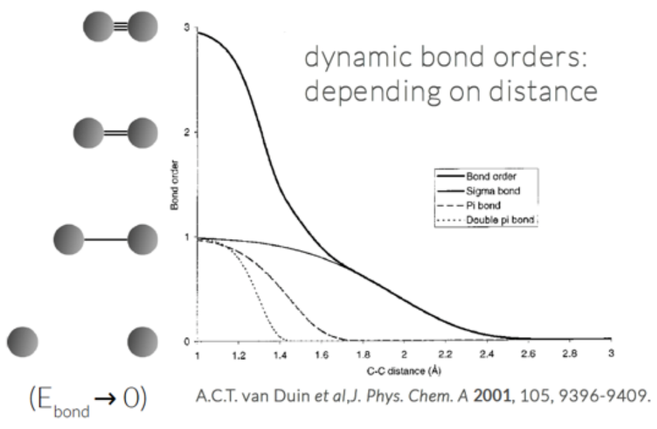
$$E_{\text{system}} = E_{\text{bond}} + E_{\text{lp}} + E_{\text{over}} + E_{\text{under}} + E_{\text{val}} + E_{\text{pen}} + E_{\text{coa}} + E_{\text{C}_2} + E_{\text{tors}} + E_{\text{conj}} + E_{\text{H-bond}} + E_{\text{vdWaals}} + E_{\text{Coulomb}}$$

$$BO'_{ij}(r_{ij}) = \exp\left[p_{bo,1} \cdot \left(\frac{r_{ij}}{r_o}\right)^{p_{bo,2}}\right] + \exp\left[p_{bo,3} \cdot \left(\frac{r^{\pi}_{ij}}{r_{o,\pi}}\right)^{p_{bo,4}}\right] + \exp\left[p_{bo,5} \cdot \left(\frac{r^{\pi\pi}_{ij}}{r_{o,\pi\pi}}\right)^{p_{bo,6}}\right]$$

In: distance between atoms, r_{ij}
Out: 1, 2, 1.42, etc...

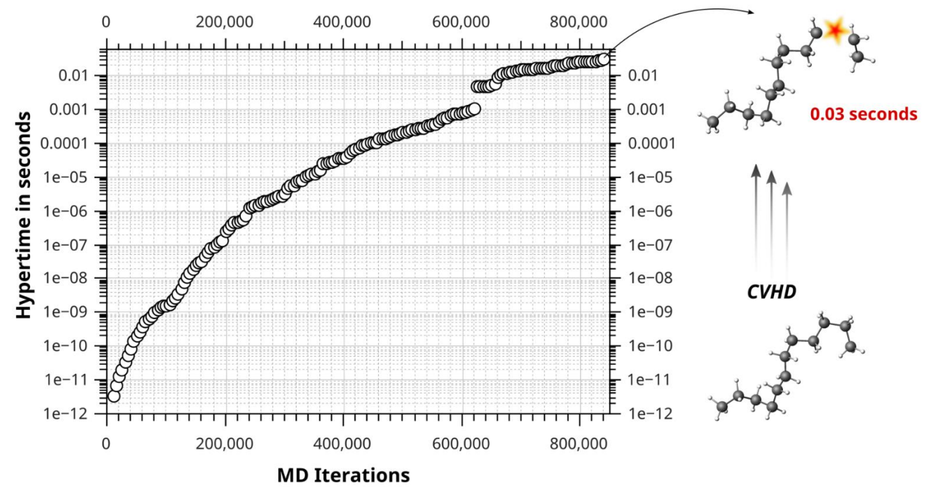
Parameters = 16
$p_{bo,1}, p_{bo,2}, p_{bo,3}, p_{bo,4}, p_{bo,5}, p_{bo,6}, r_o, r_{o,\pi}, r_{o,\pi\pi}$
$val_1, val_2, \lambda_1, \lambda_2, \lambda_3, \lambda_4, \lambda_5$

Correction terms f_1, f_2, f_3: $BO_{ij}(r_{ij}) = BO'_{ij}(r_{ij}) \cdot f_1(BO'_{ij}) \cdot f_2(BO'_{ij}) \cdot f_3(BO'_{ij})$

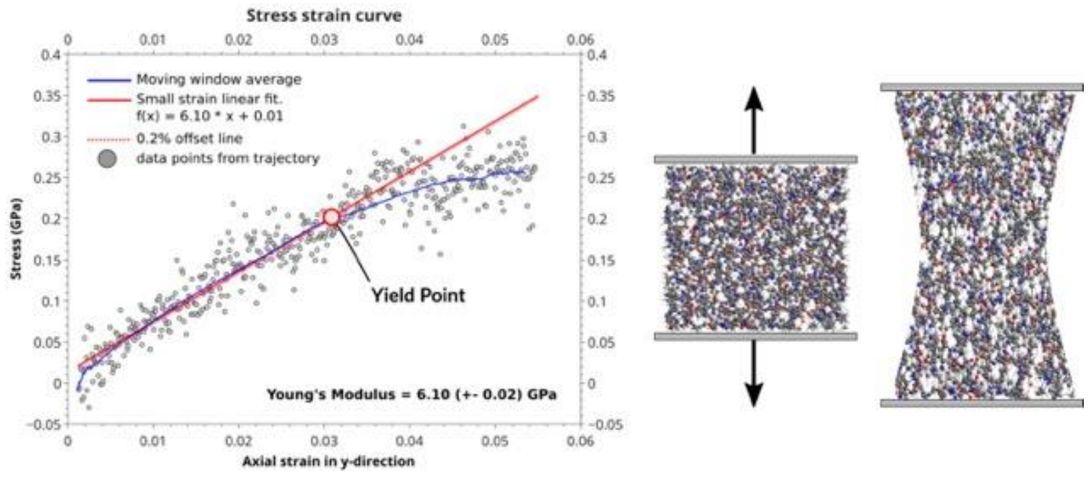


($E_{bond} \rightarrow 0$) A.C.T. van Duin *et al*, *J. Phys. Chem. A* **2001**, 105, 9396-9409.

Review NPJ Comp. Materials (2016):
The ReaxFF reactive force field: development, applications and future directions



Combustion, pyrolysis
Chem. Sci., 2016, 7, 5280

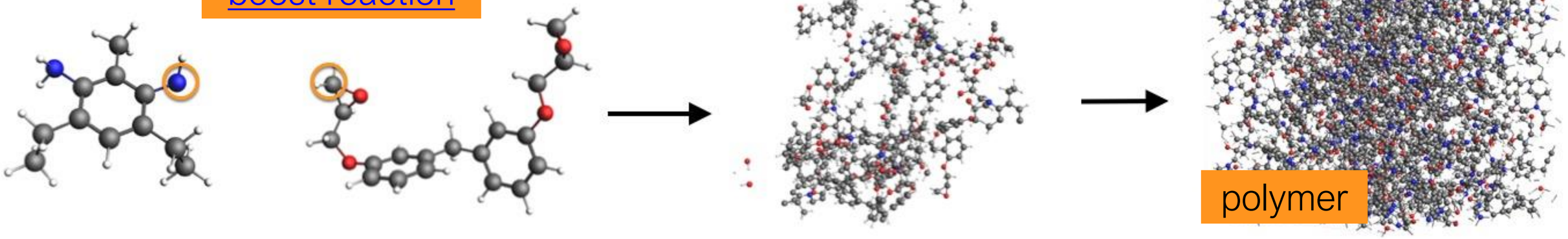


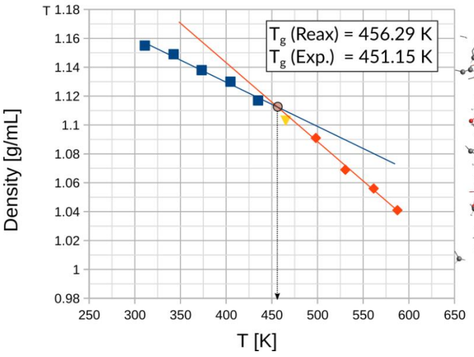
Polymer formation & Tg, CTE, yield point
J. Polym. Sci. B 2018, 56, 255

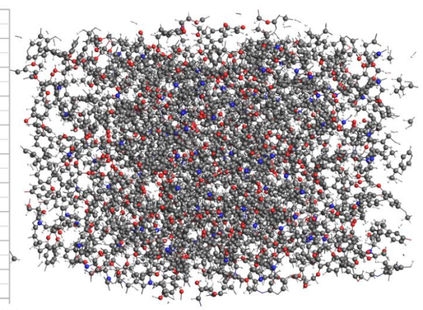
Epoxy polymers: structure & properties

- Understand & predict how cross-linking effects mechanical properties
  - Exp = slow: minutes to hours to reach ~80% cross-linking
  - ReaxFF: simulate few ns => accelerate kinetics to get highly xlinked structures

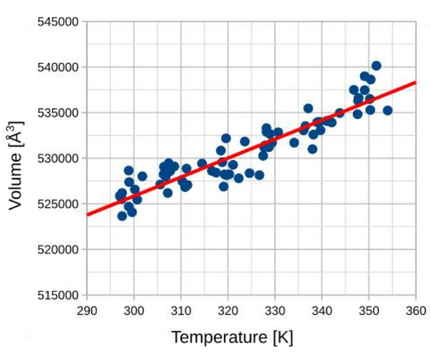
epoxy + amine
boost reaction

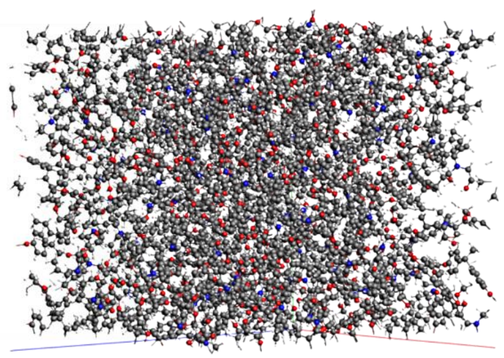




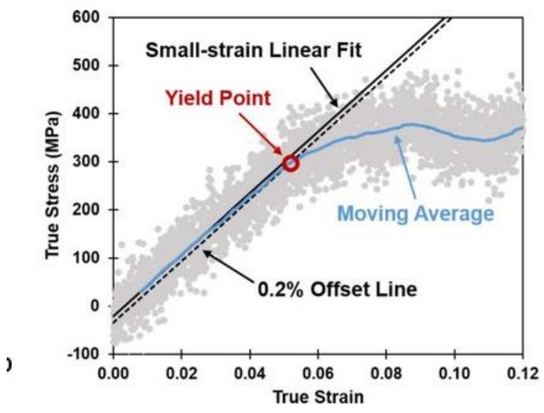


density(T) using npT
glass transition T_g



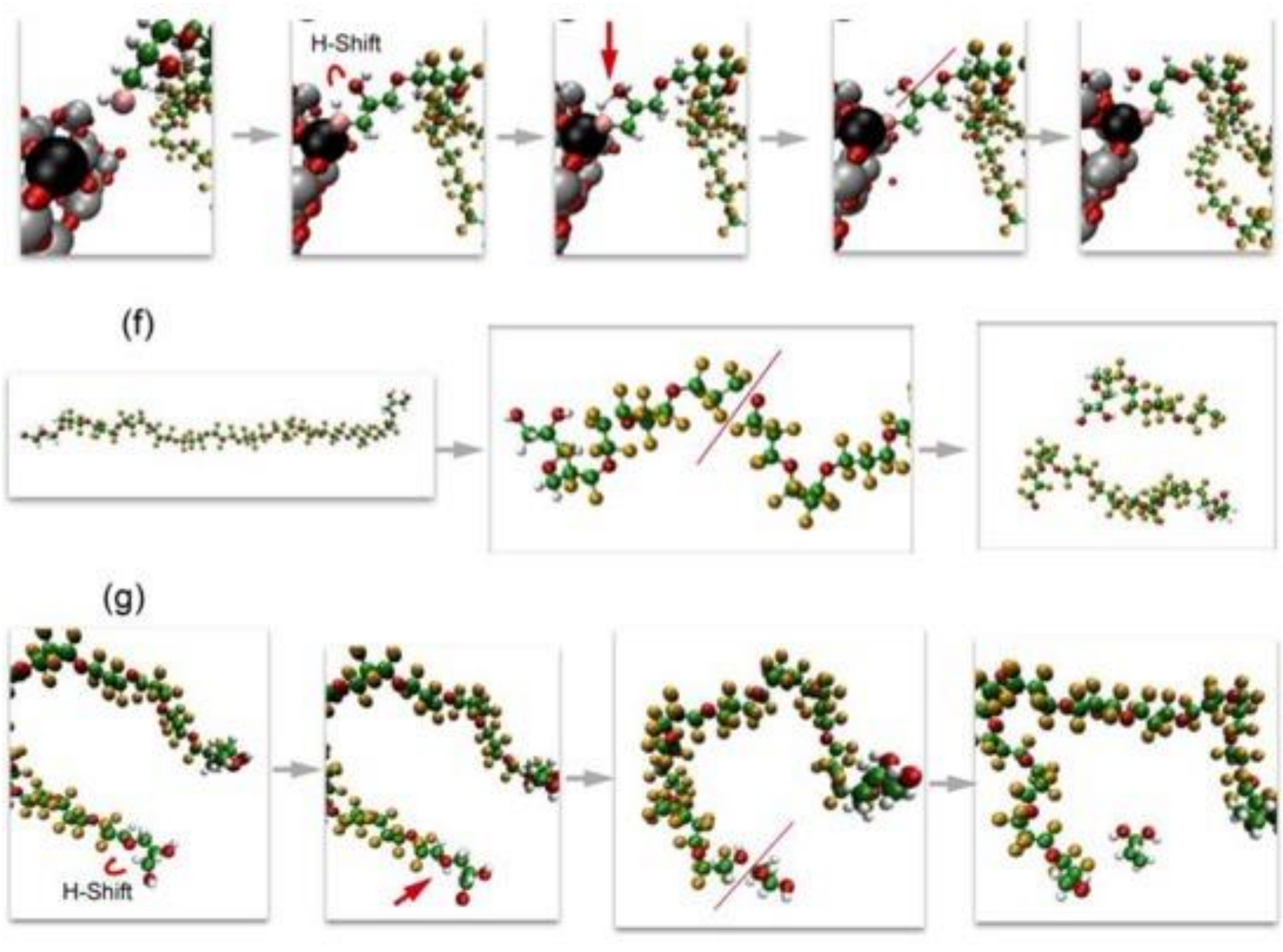


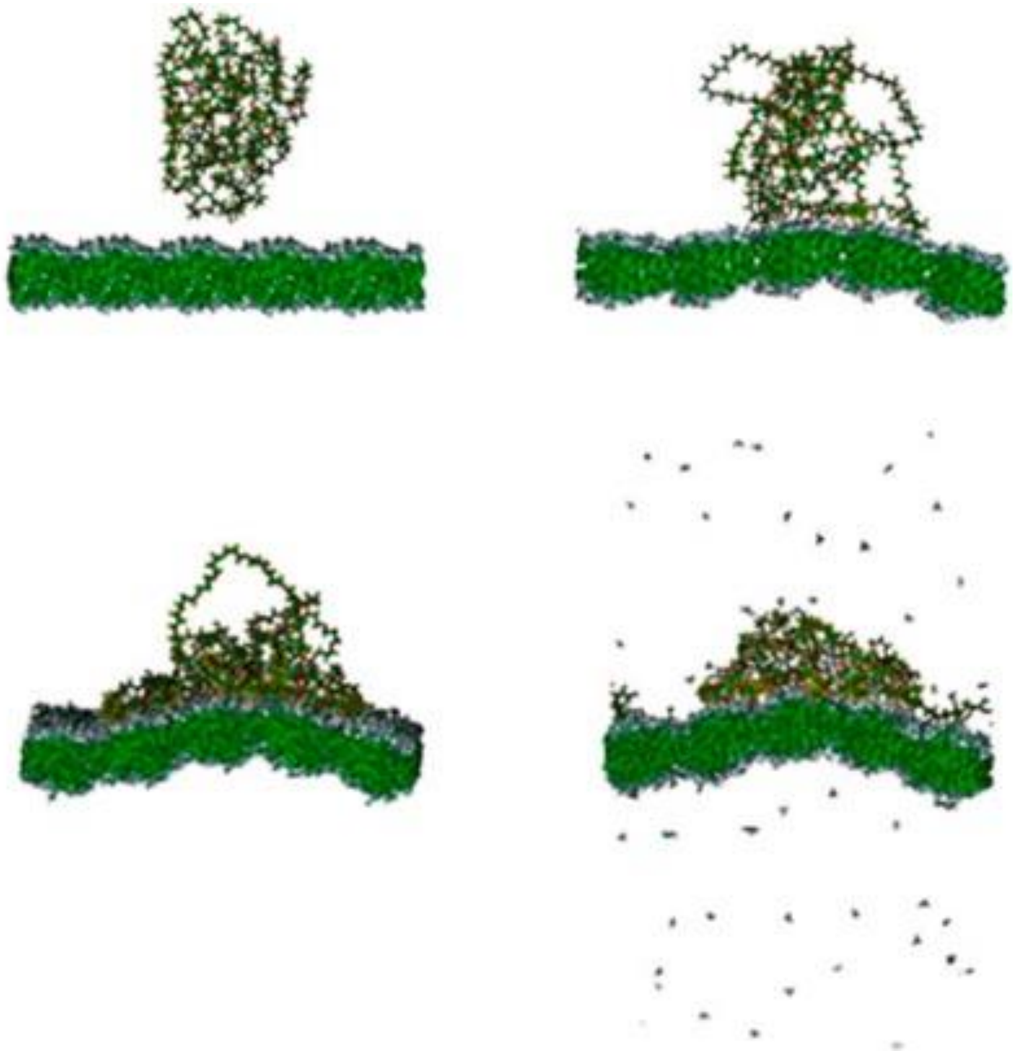
volume(T) npT
CTE



Stress-strain:
yield point, Poisson ratio

ReaxFF simulations: degradation





Degradation mechanisms perfluoropolyether lubricant on SiO_2, Fe_2O_3 nanoparticles & DLC, effect of oxygen and water. Work by van Duin (Penn State/RxFFconsulting) with **Western Digital**

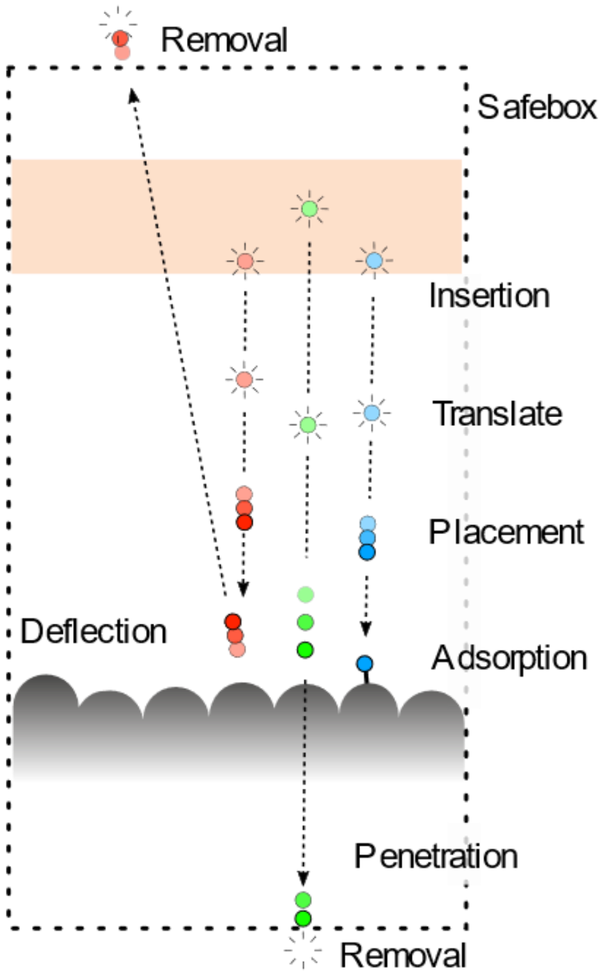
J. Phys. Chem. C, **120**, 27433 (2016)

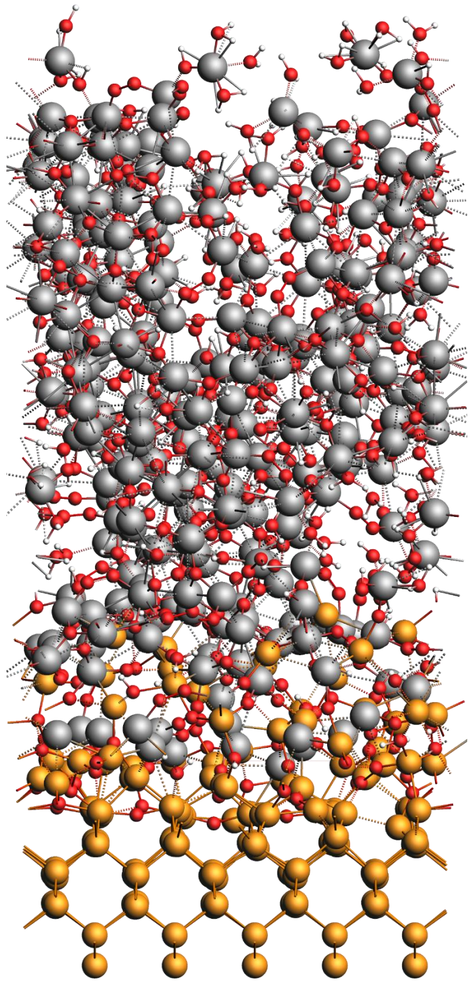
J. Phys. Chem. C, **122**, 2684 (2018)

Sputtering, etching, CVD, ALD, PVD

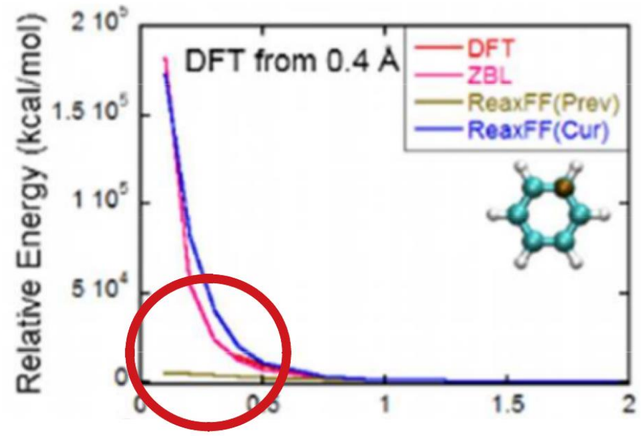
Shoot molecule/atomic targets at surface

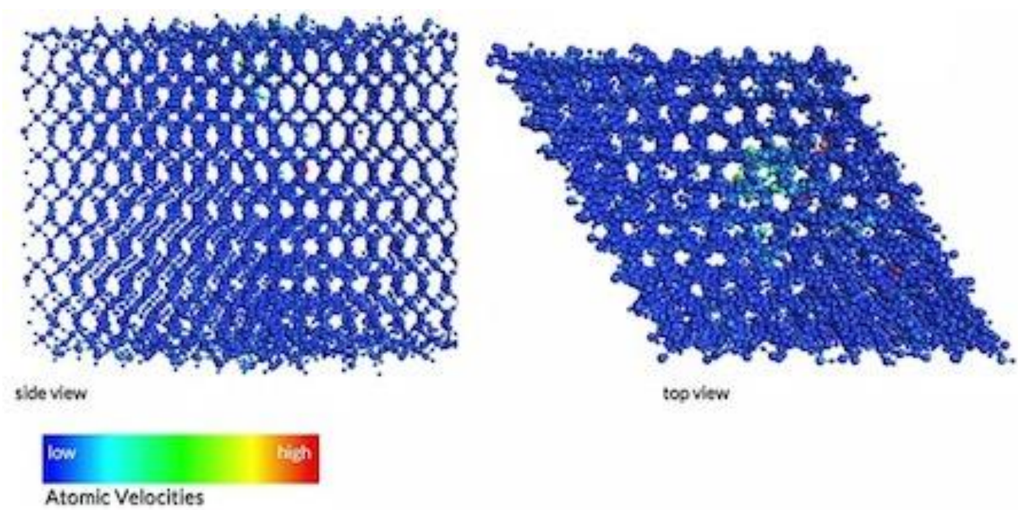
- High E: sputtering, (plasma/photo?)etching
- Low E: CVD, ALD, PVD





- Need good force fields: ReaxFF refit short distances
- Sputtering yields (E, angle) (movie: Ar / SiO_2)

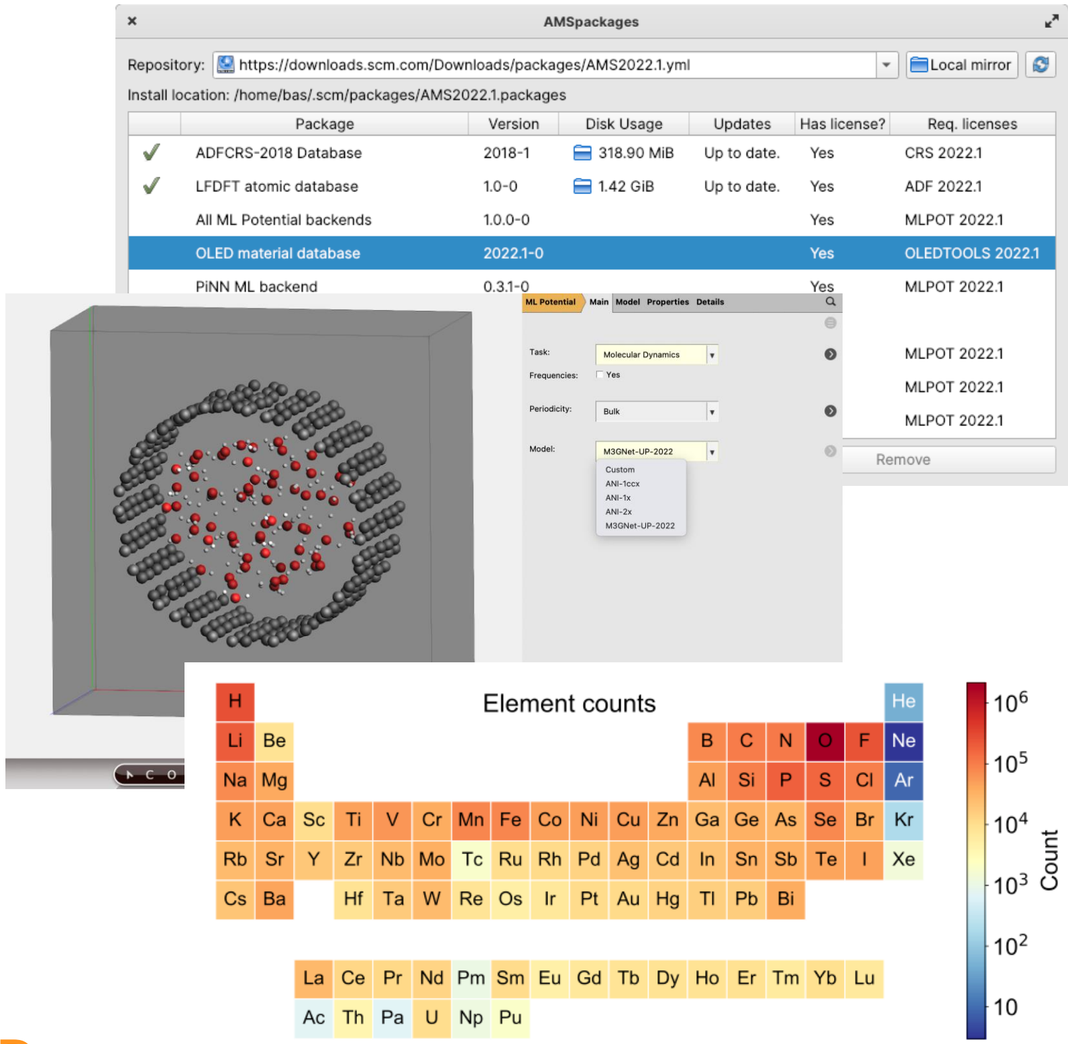




Tutorial CVD: Alumina / Ge(001)

Machine Learning Potentials

- Automatically install popular ML Backends
  - Pre-parametrized
    - i) ANI-1x and 2x (H, C, N, O, F, S, Cl)
    - ii) M3GNet (“Universal”)
  - Backends, via ASE
    - iii) NEquIP, FLARE (on-the-fly)
    - iv) sGDML
    - v) SchNet
    - vi) CHGNet
- Use MLP with all the tasks in the AMS driver
  - PES scans, reaction discovery, conformers, IR, phonons, MD, MC etc.
  - Hybrid (multi-layer): combine with other methods
- CUDA-enabled PyTorch and Tensorflow

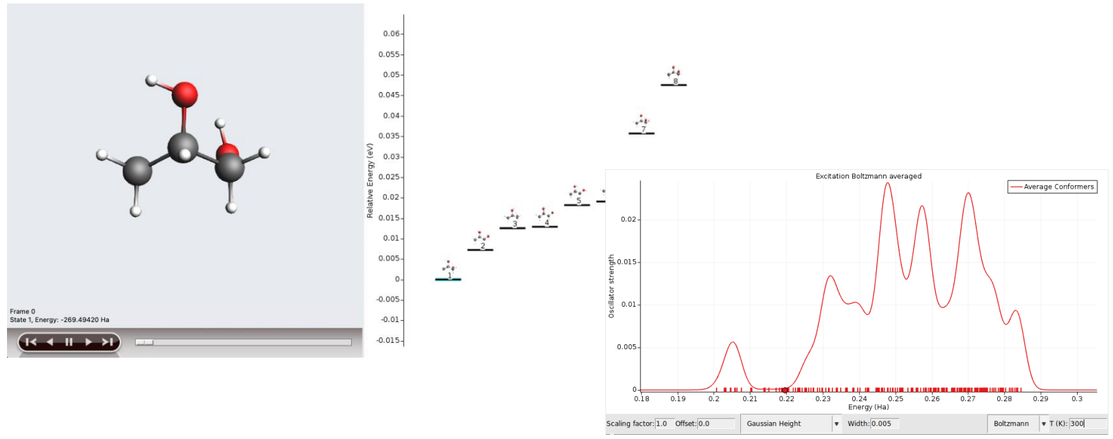


i) O. Isayev et al. Chem. Sci., 2017, 8, 3192–3203
ii) C. Chen, S.P. Ong., Nature Comp. Sci. 2, 718–728 (2022)
iii) S. Batzner et al. Nature Comm. 13: 2453 (2022)
iv) S. Chmiela et al. Comp. Phys. Commun. 240 (2019) 38-45
v) K. T. Schütt et al., J. Chem. Theory Comput. 15 (2019) 448-455

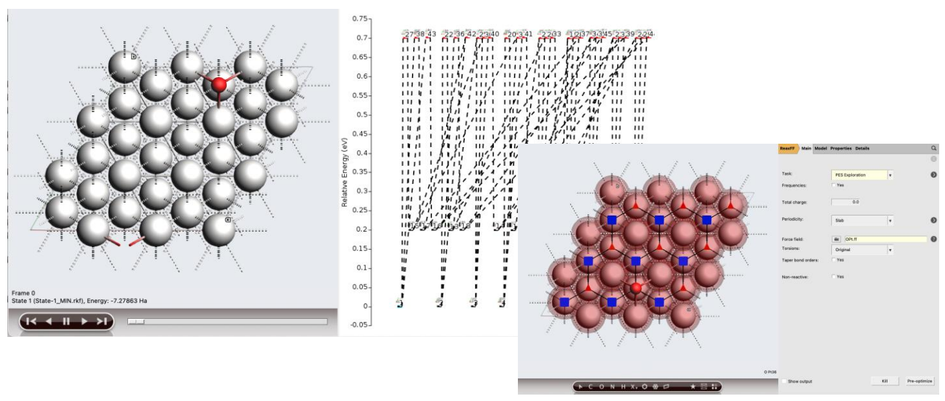
MLPotentials for chemistry

From organic chemistry to catalysis

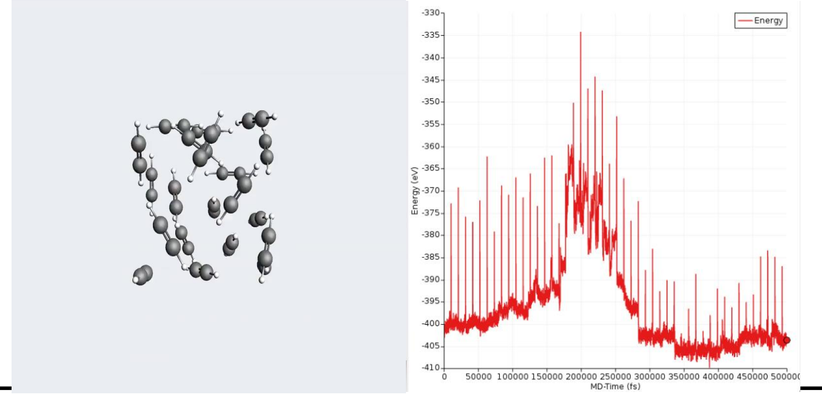
- Explore conformers with ANI-1x/2x
  - CREST & RDKit conformer generation
  - Re-score with DFT
  - Boltzman averaged properties



- Screen adsorption sites with PESexploration
  - Refined (or fully computed) with MLP
  - OCP (Open Catalyst Project) via ASE engine https://opencatalystproject.org/



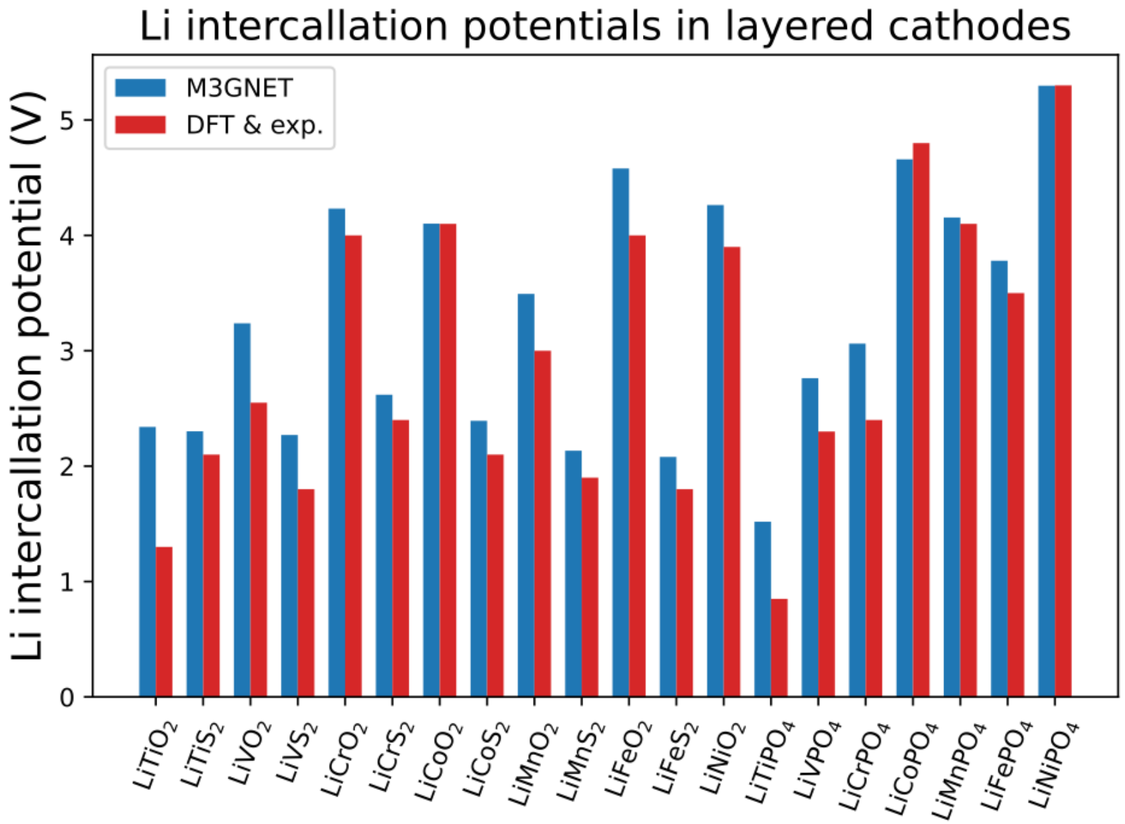
- Discover chemical reactions with ANI-1xnr (2024)
  - Use the nano reactor to accelerate reactions
  - Analyze all chemical reactions with ChemTrayZer



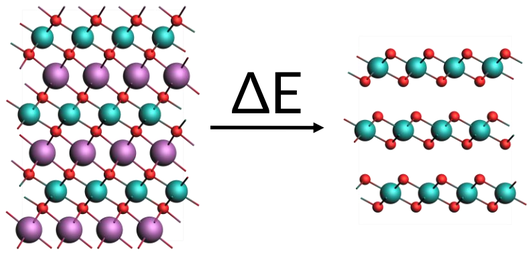
MLPotentials for (battery) materials

M3GNet, trained to Materials Project 1000x faster than DFT

C. Chen, S.P. Ong., Nature Comp. Sci. 2, 718–728 (2022)



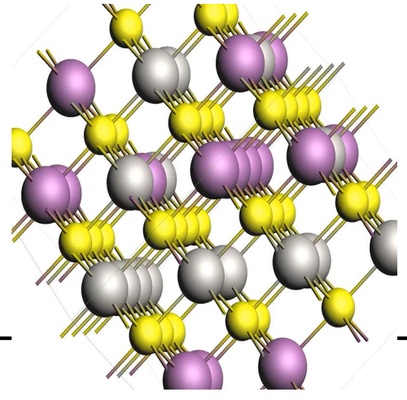
- Li intercalation potentials accurately predicted with DFT (~100 atoms)
- M3GNet reproduces DFT really well

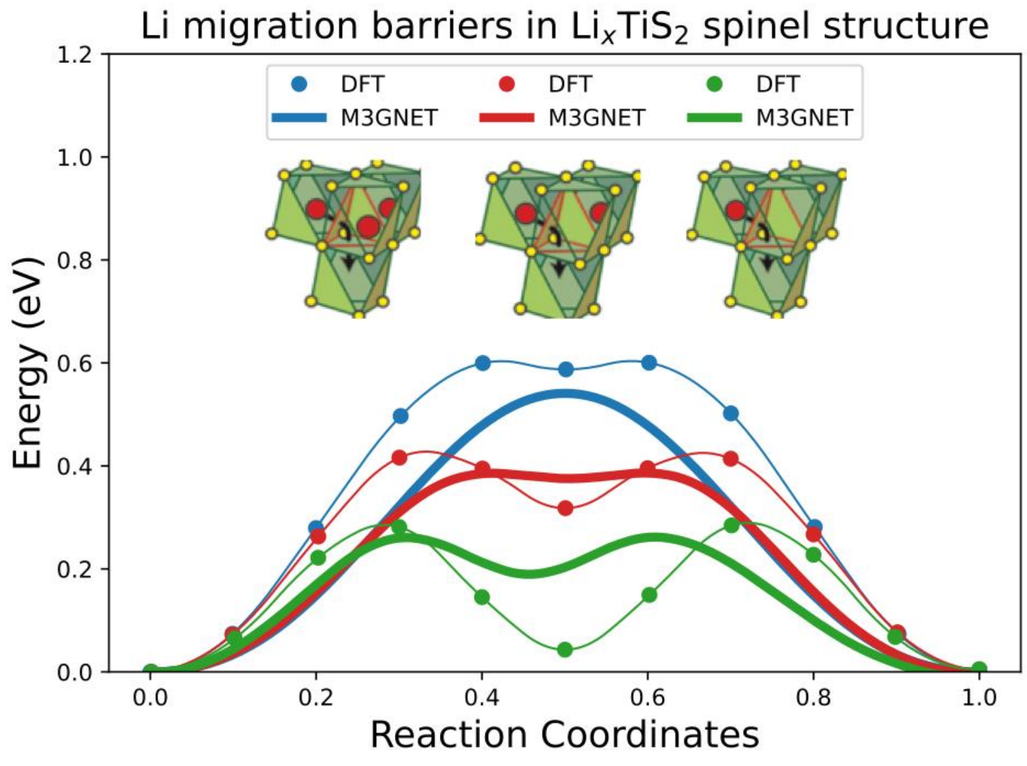


- Li potential
- Mechanical properties of electrode (volume change upon lithiation)

- Li diffusion through NEB or PES scan
- M3GNet can accelerate by 1000x
- (Dis)charge curves: GCMC

- Activation energy
- Diffusion (kinetics)

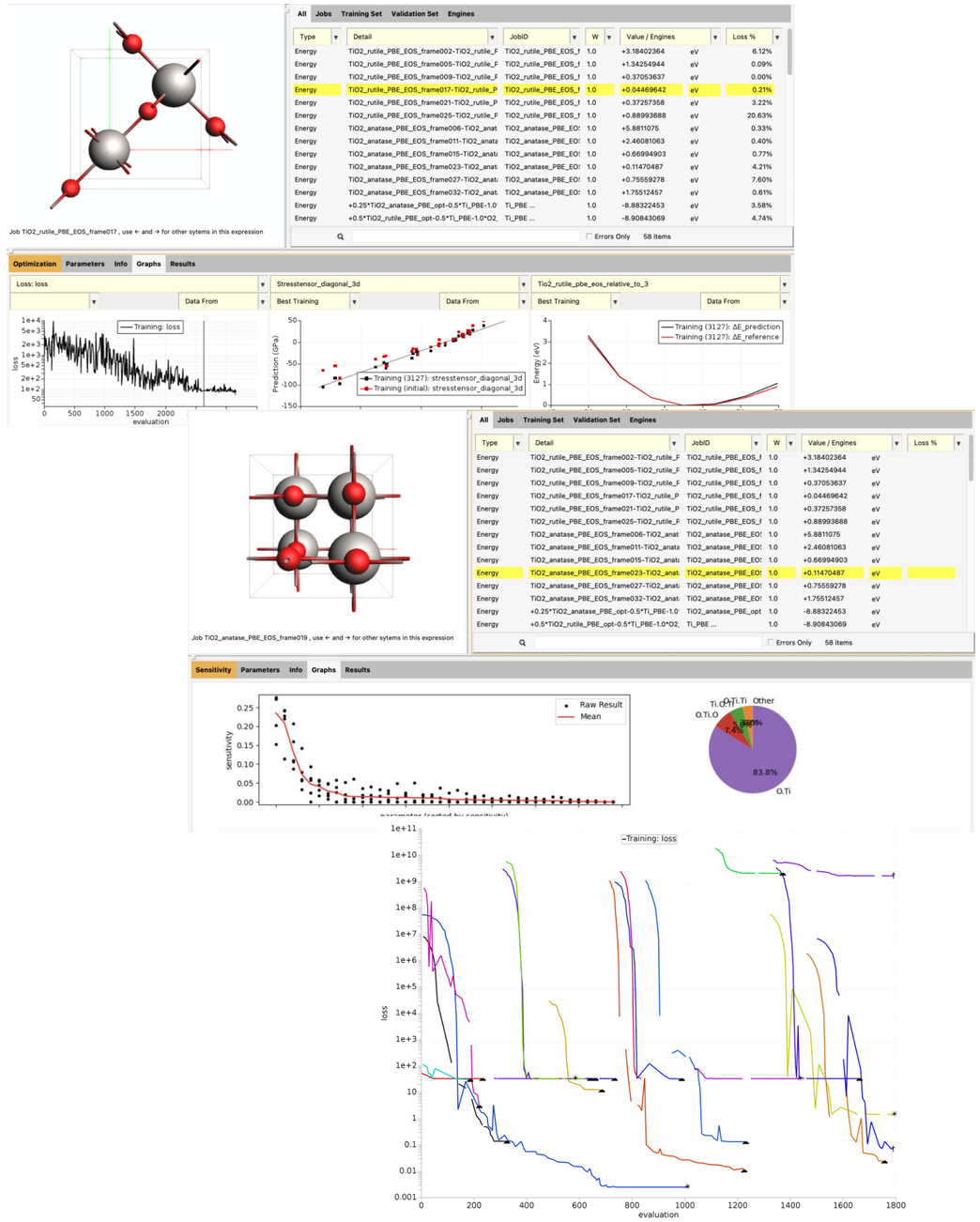




Training FF with ParAMS

ParAMS making parameters optimization easier

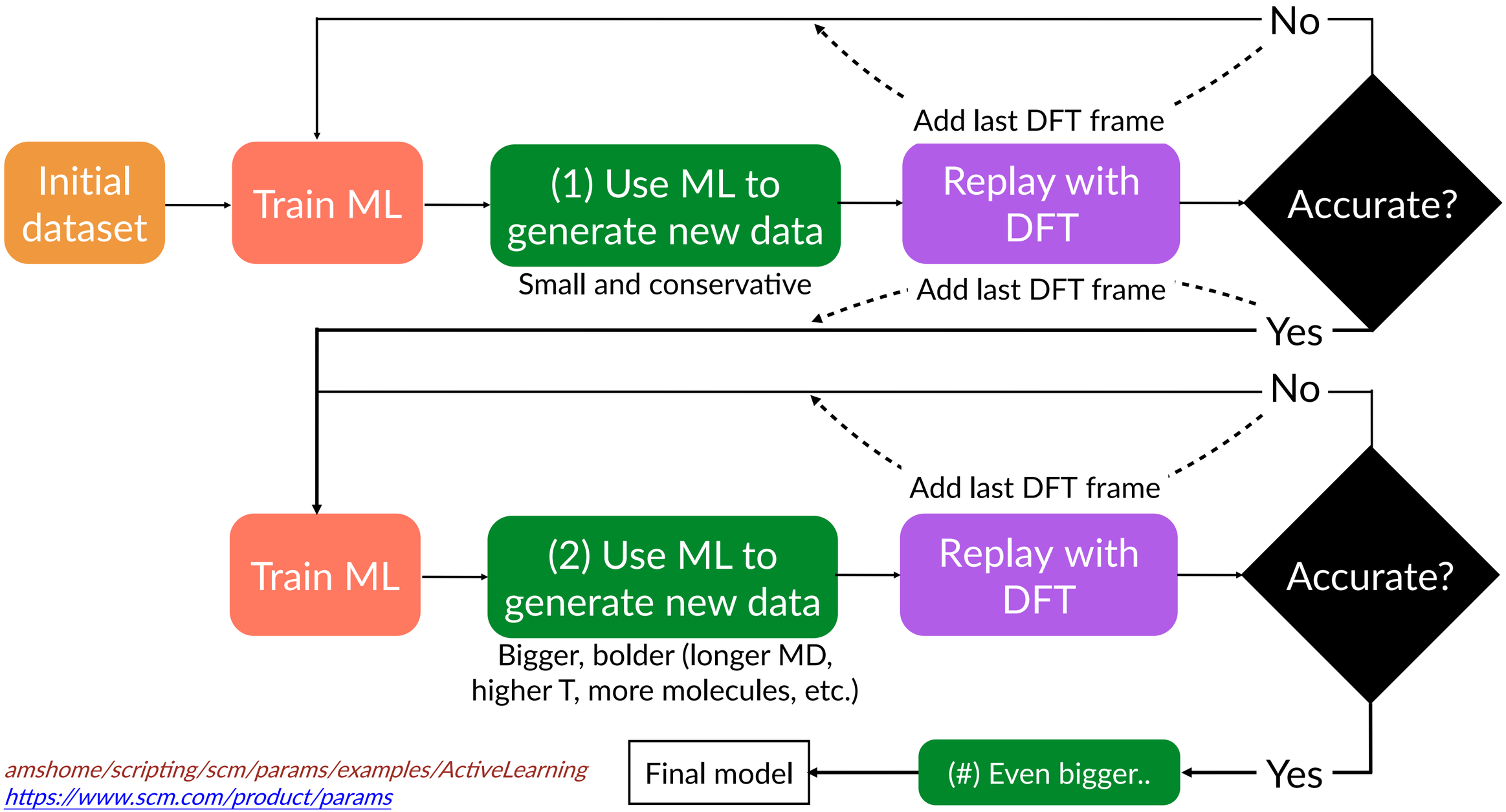
- Import, build and visualize training data
- Use data from AMS, VASP, QE, and experiments
- Tune parameters to describe energies, forces, geometries, stress tensor, charges, bandgaps, etc.
- Use validation sets to prevent overfitting
- Submit multiple optimization at the same time to explore parameters space
- Explore the sensitivity of the parameters to select the most important subset to optimize
- Train DFTB, ReaxFF, Force field, and … MLP



J. Chem. Inf. Model. 2021, 61, 8, 3737–3743
J. Chem. Theory Comput. 2023, 19, 9, 2557–2573
https://www.scm.com/product/params

Training MLP with ParAMS

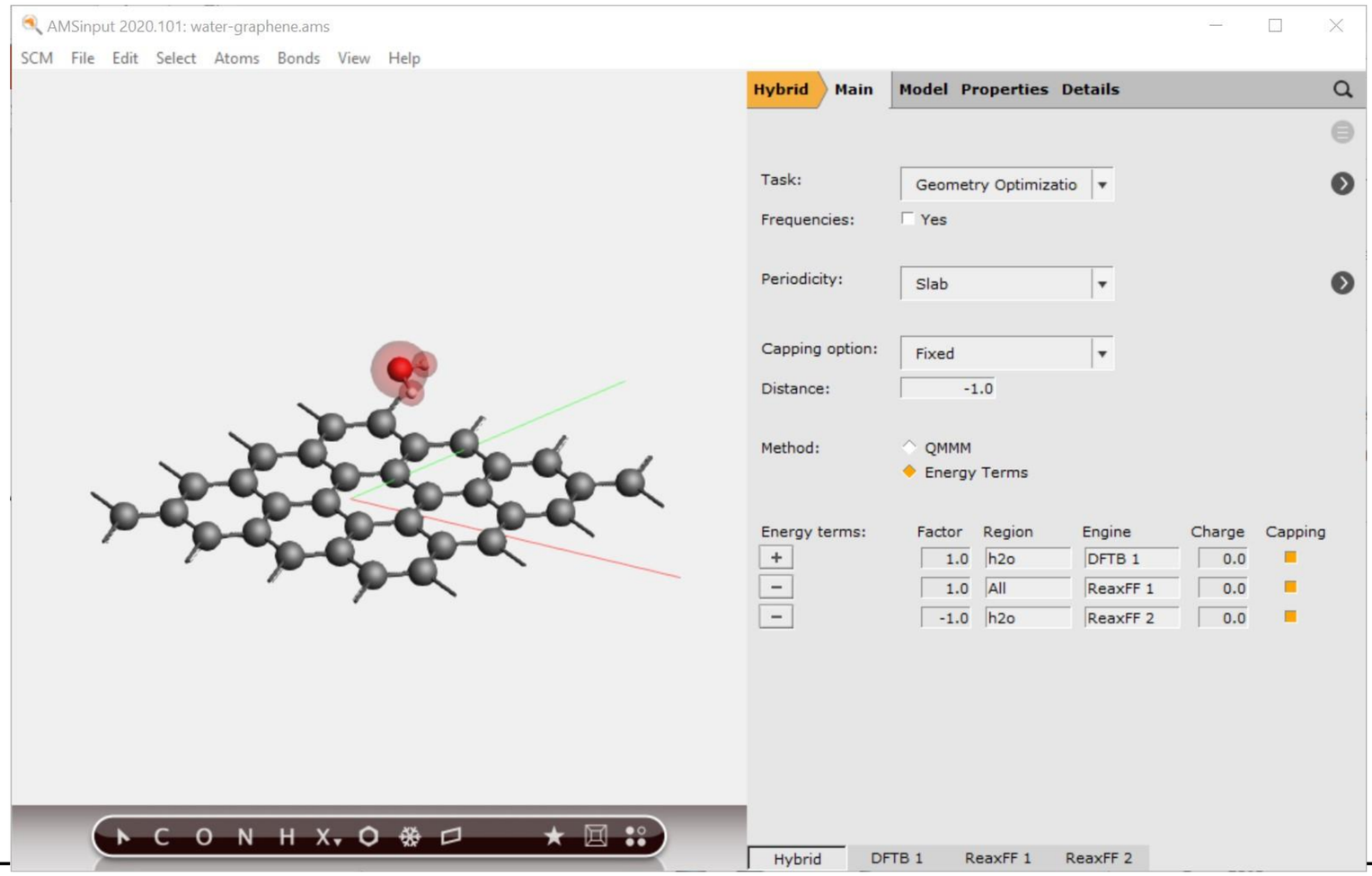
Active learning workflow

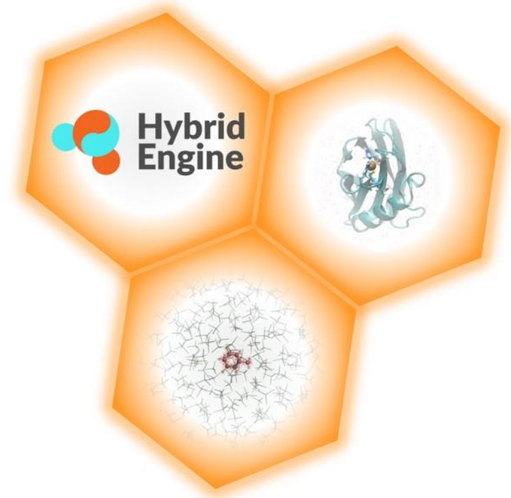


amshome/scripting/scm/params/examples/ActiveLearning
https://www.scm.com/product/params

Hybrid Engine

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



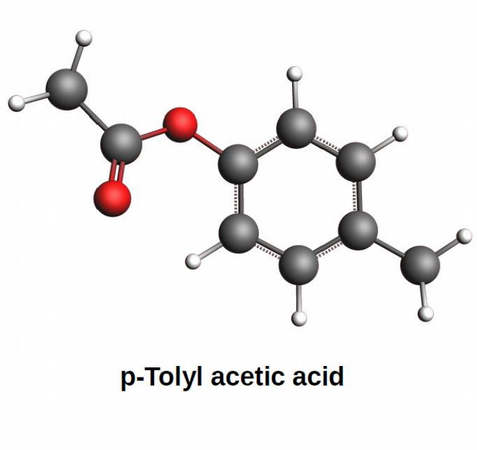


[Demo video]

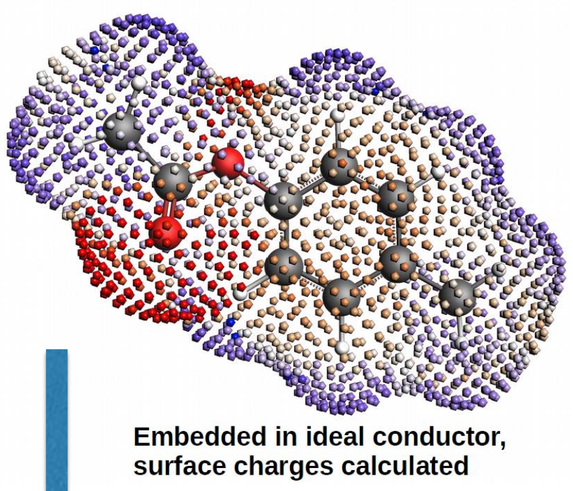
COSMO-RS/SAC: thermodynamic properties of fluids

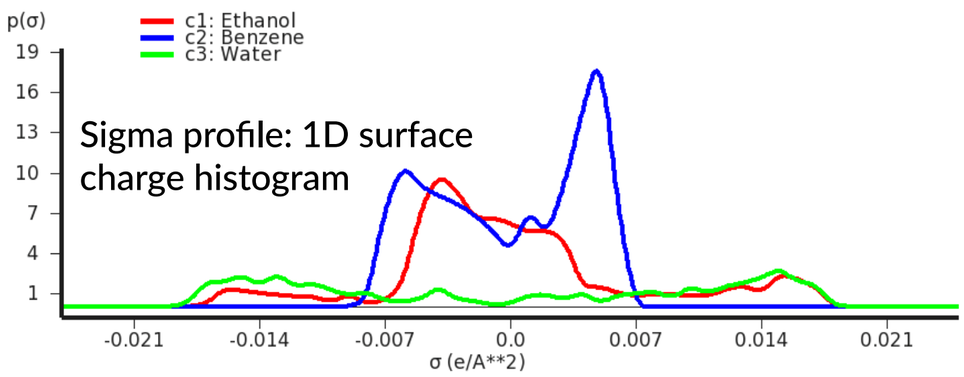
Quantum Chemistry & QSPR for quick property predictions

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

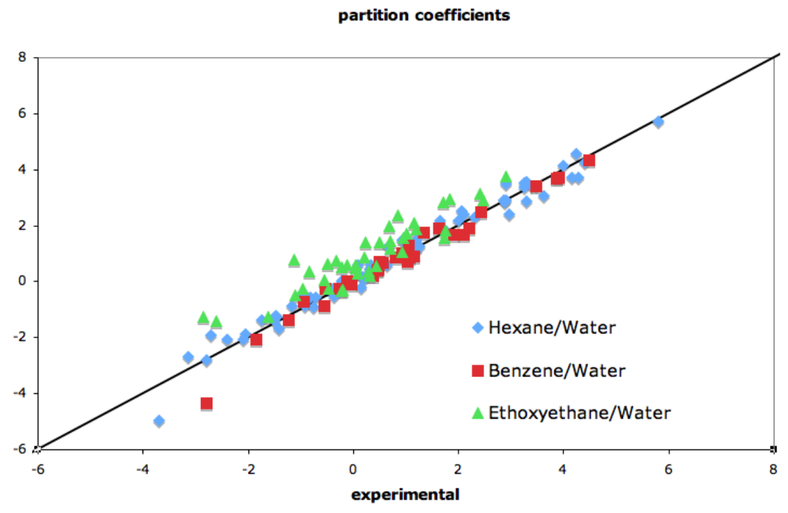


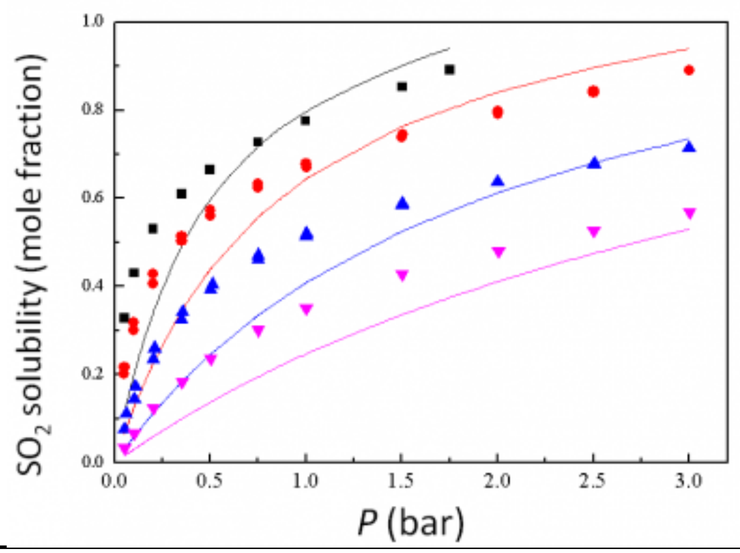
p-Tolyl acetic acid





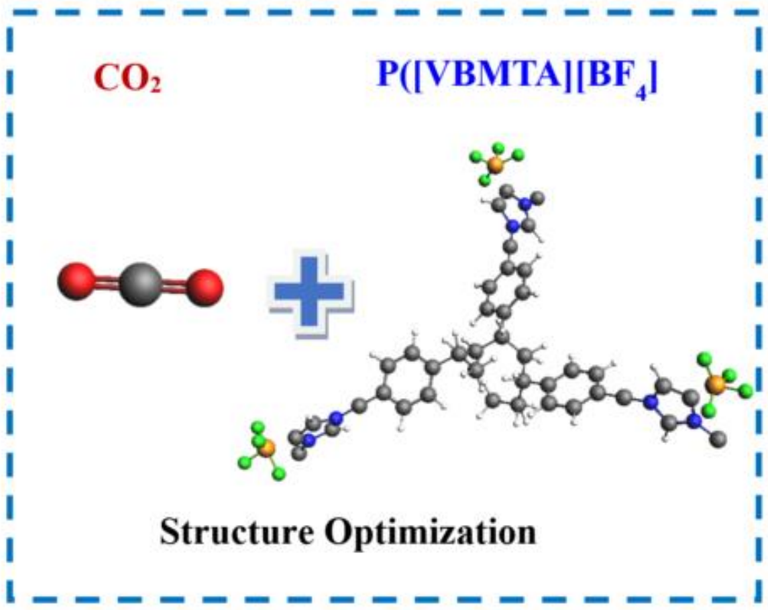
Sigma profile: 1D surface charge histogram

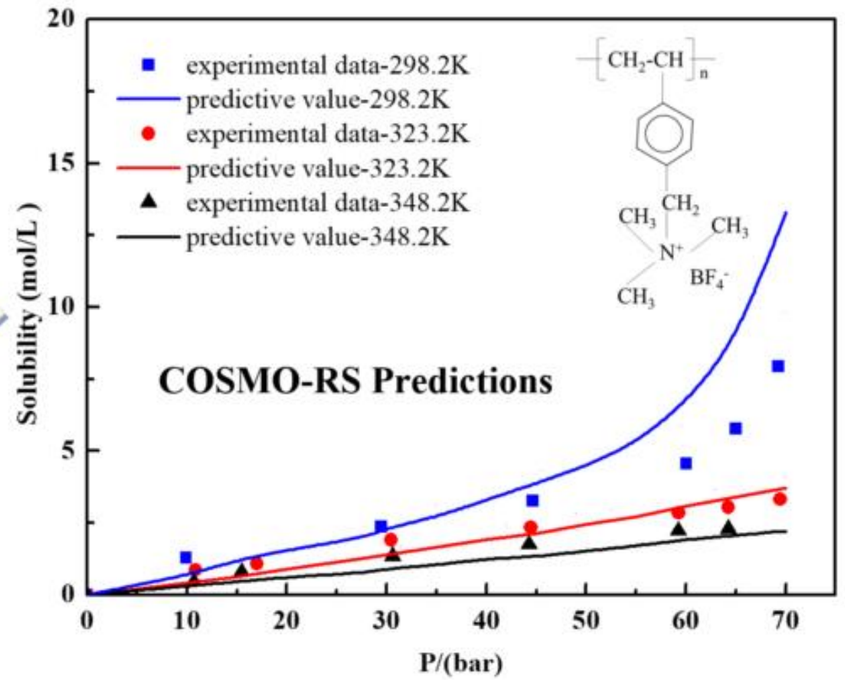


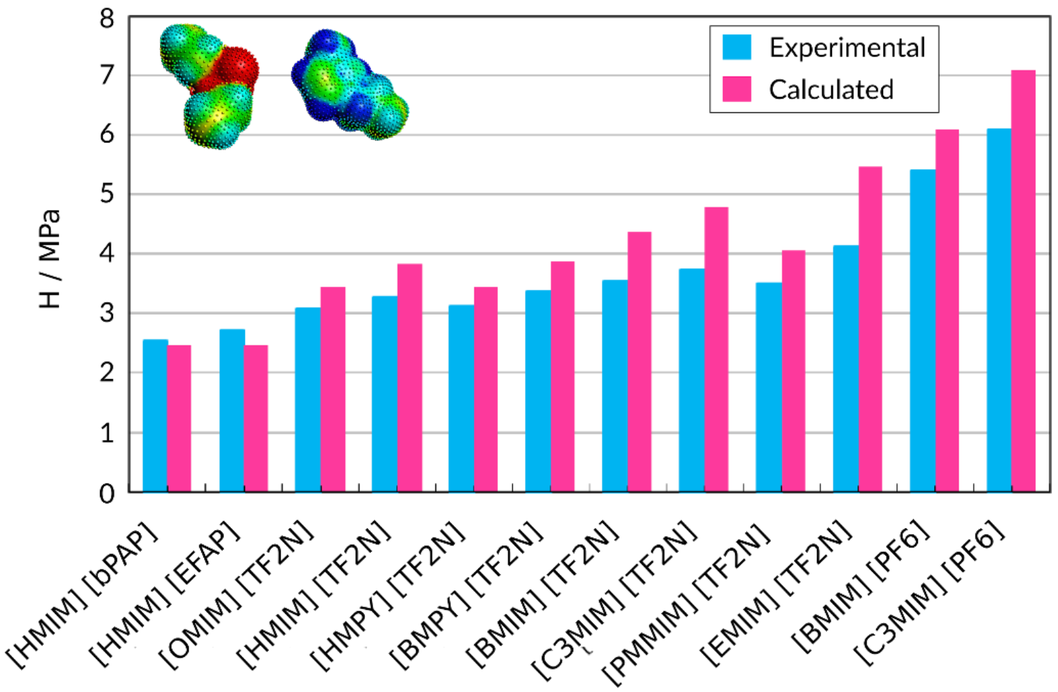


- Solvation & excess energies, pKa
- Solubilities, LLE, VLE, boiling points
- Optimize mixtures: solubility, LLE
- Polymers: Flory-Huggins X
- QSPR & fast sigma: using SMILES

COSMO-RS: solubility in (polymeric) ionic liquids







- COSMO-RS predicts beyond parametrization (opposed to UNIFAC)
- Works well for SO_2 solubilities
- Improvements for CO_2 (2018)
- Combined with polymer terms (2021)

Lei group: Chem. Rev., 114, 1289−1326 (2014), Green Energy & Environment (2018), Green Energy & Environment (2021)

Enjoy the workshop!

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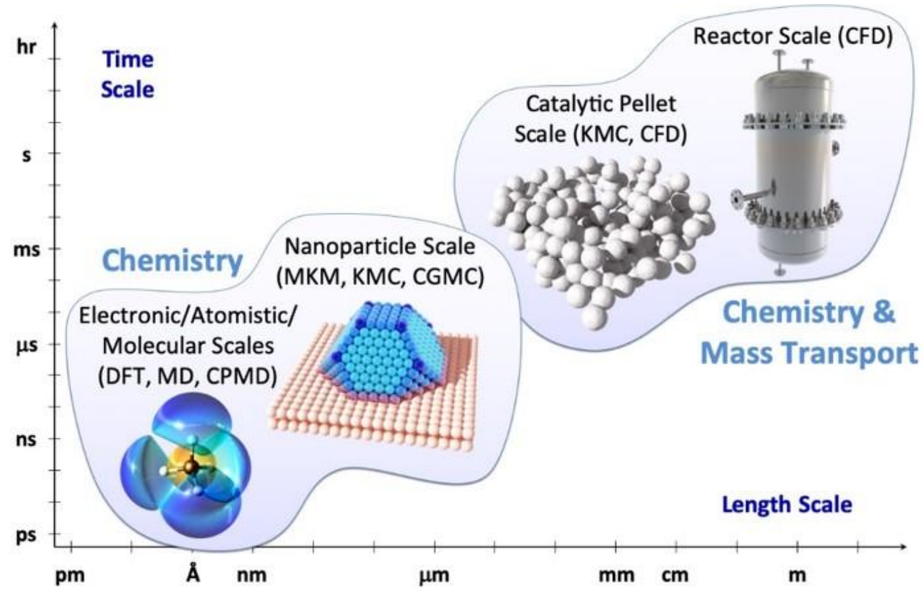
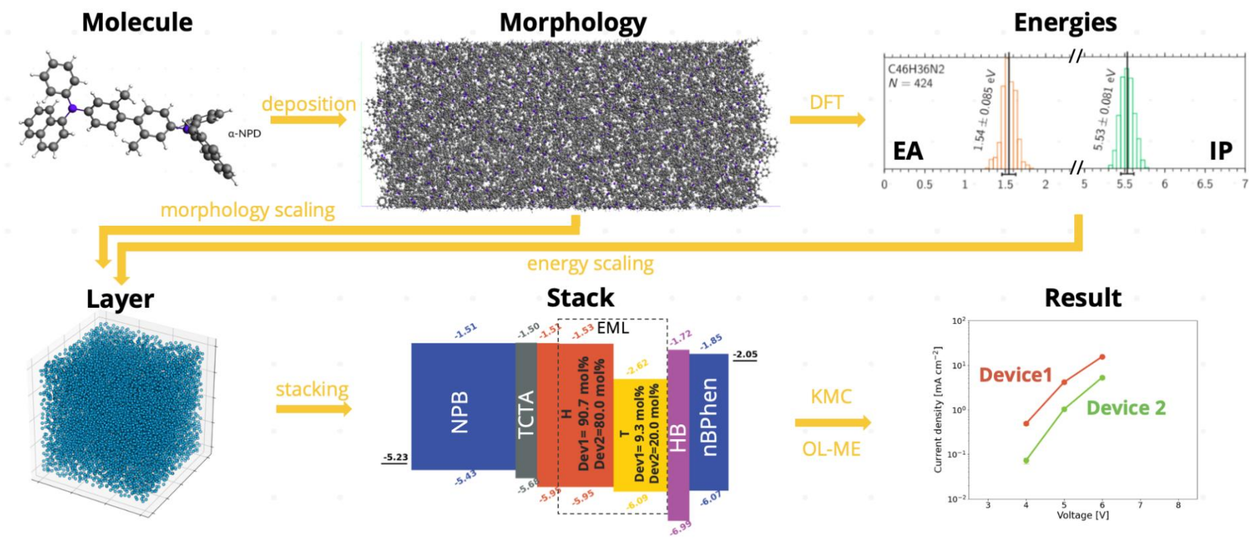
Summary: Amsterdam Modeling Suite

- Couple atomistic, meso & macro

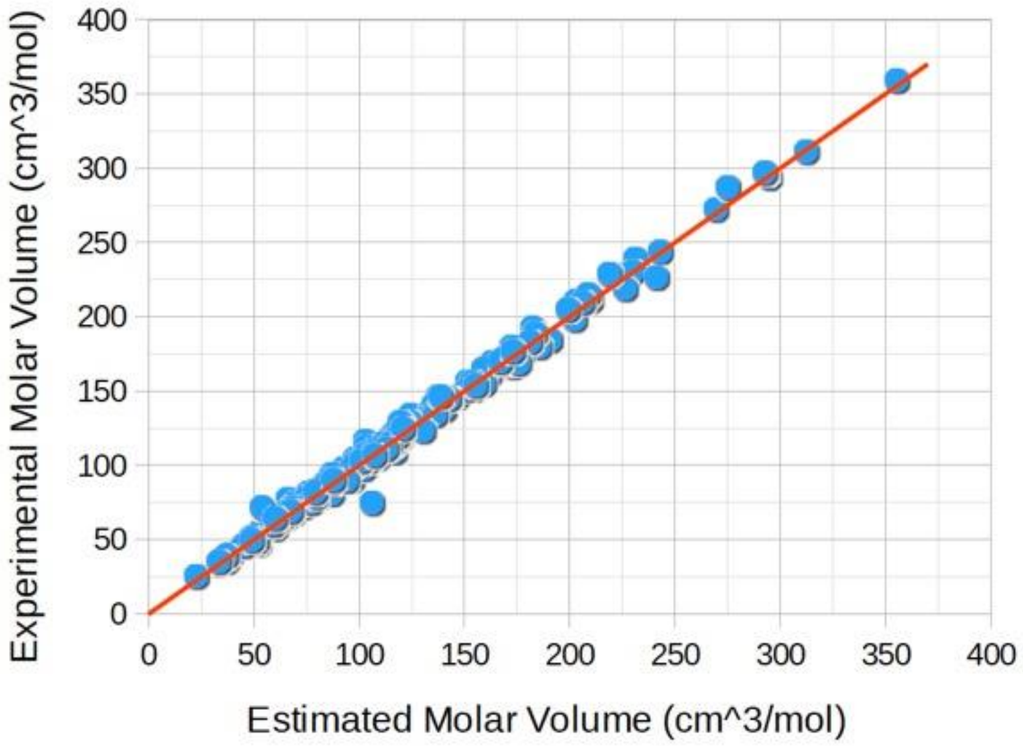
- Integrated User Interfaces
  - Graphical
  - Python layer
  - Connections to third party

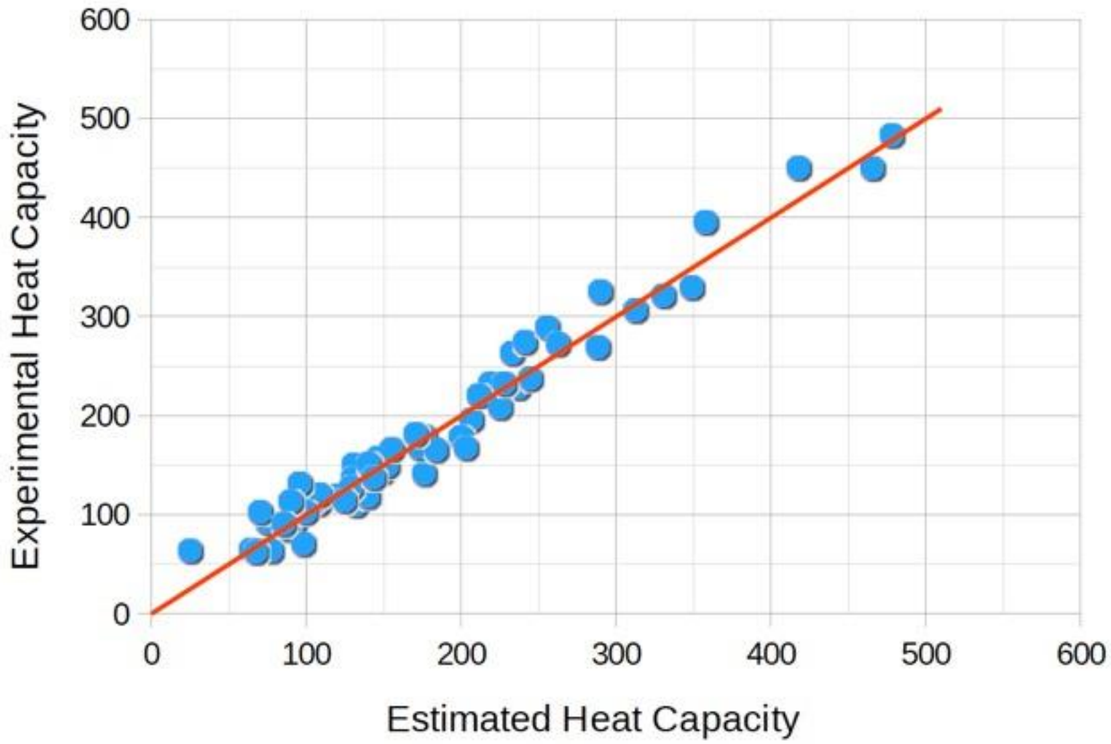


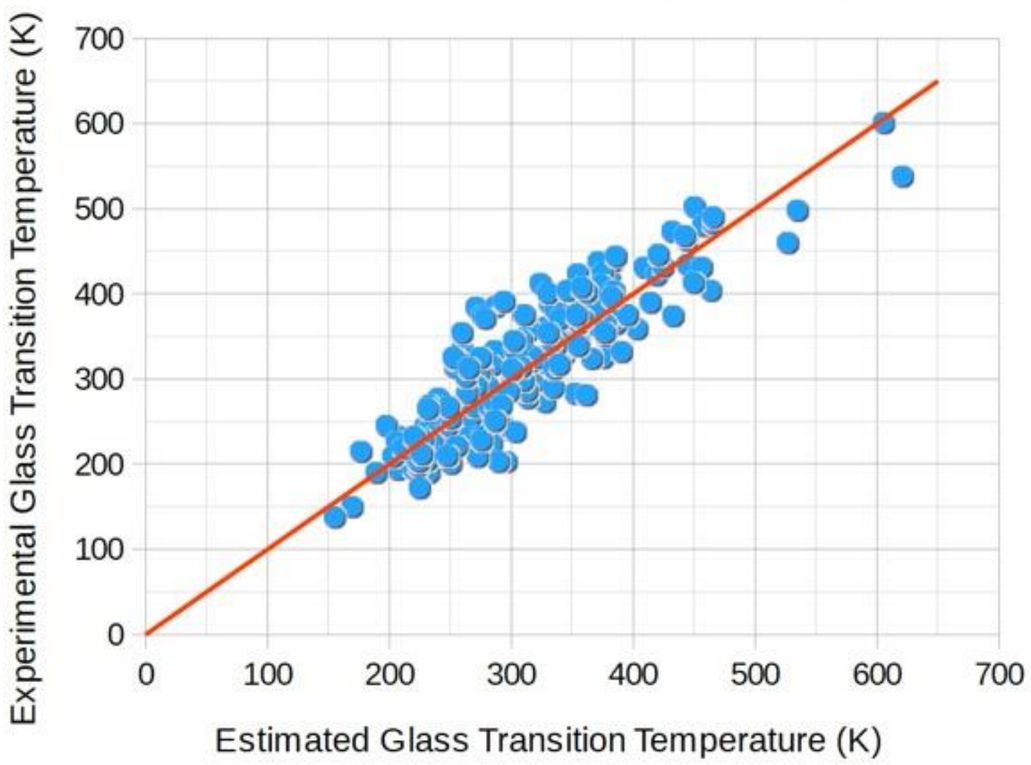
- Applications
  - Catalysis
  - Organic electronics
  - Batteries
  - Polymers



Polymers: QSPR with sigma-moments







Basic statistics

r^2:

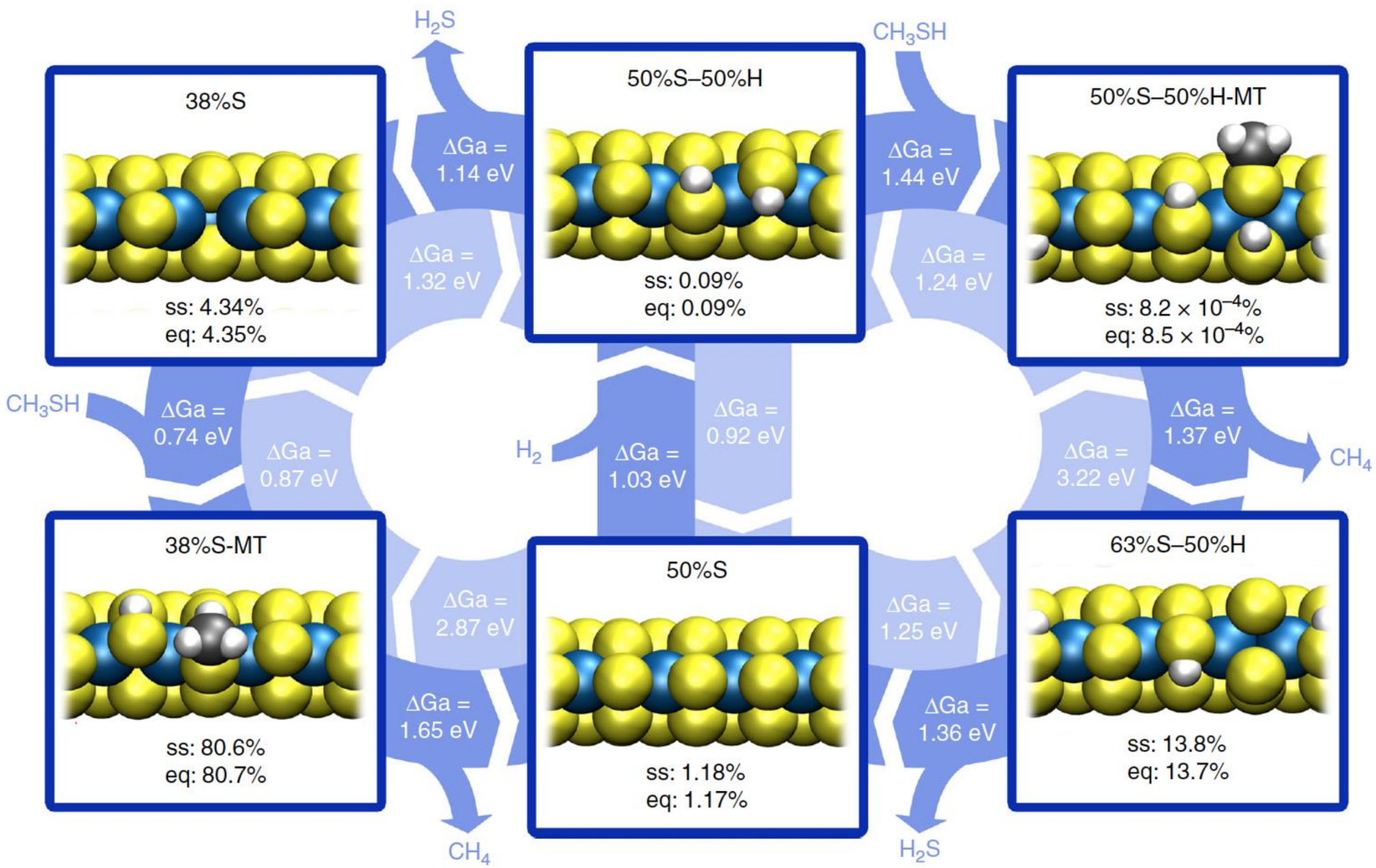
- Molar volume: > 0.99
- Heat Capacity: 0.96
- Glass Transition Temp. : 0.81

Average absolute error:

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

Hydrodesulfurization MoS_2: exp + calc

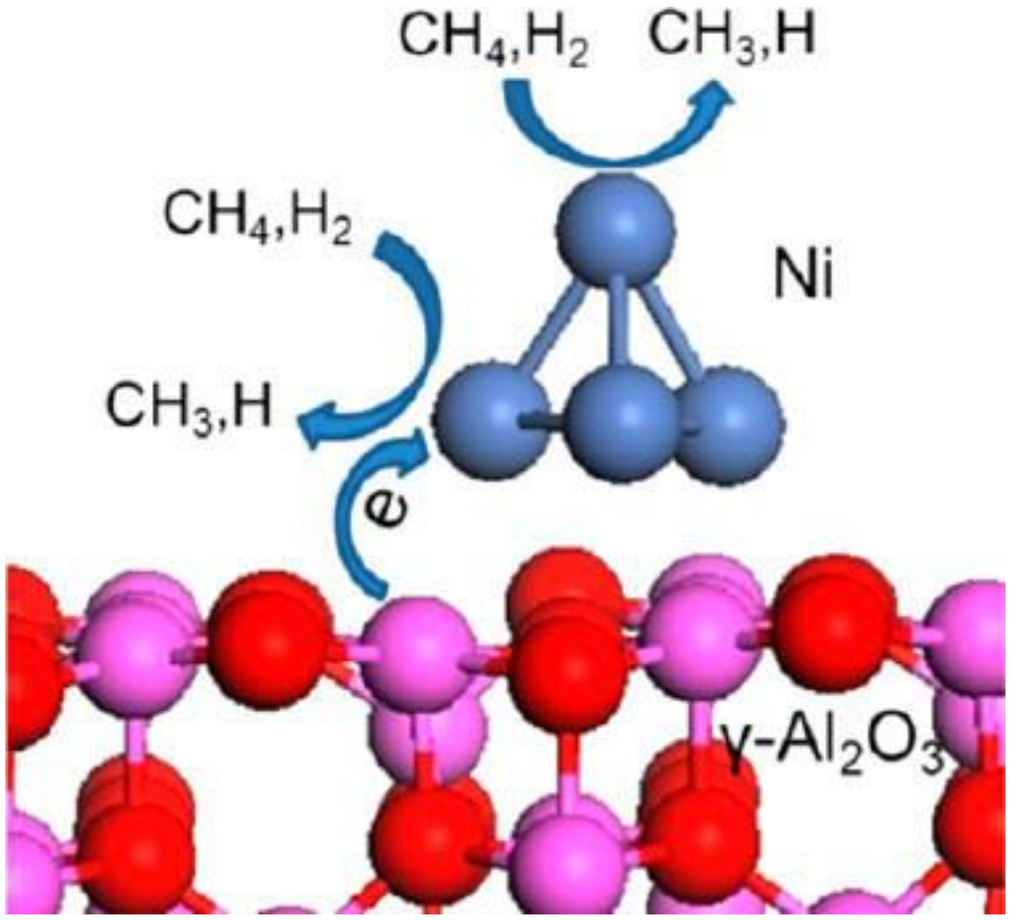
- STM with high p & T (different H_2/H_2S pressure)
- BAND calc. 2D=> phase diagram of edge -> compare with exp
- Reaction pathways (Albemarle: partial Hessian in BAND very useful!)

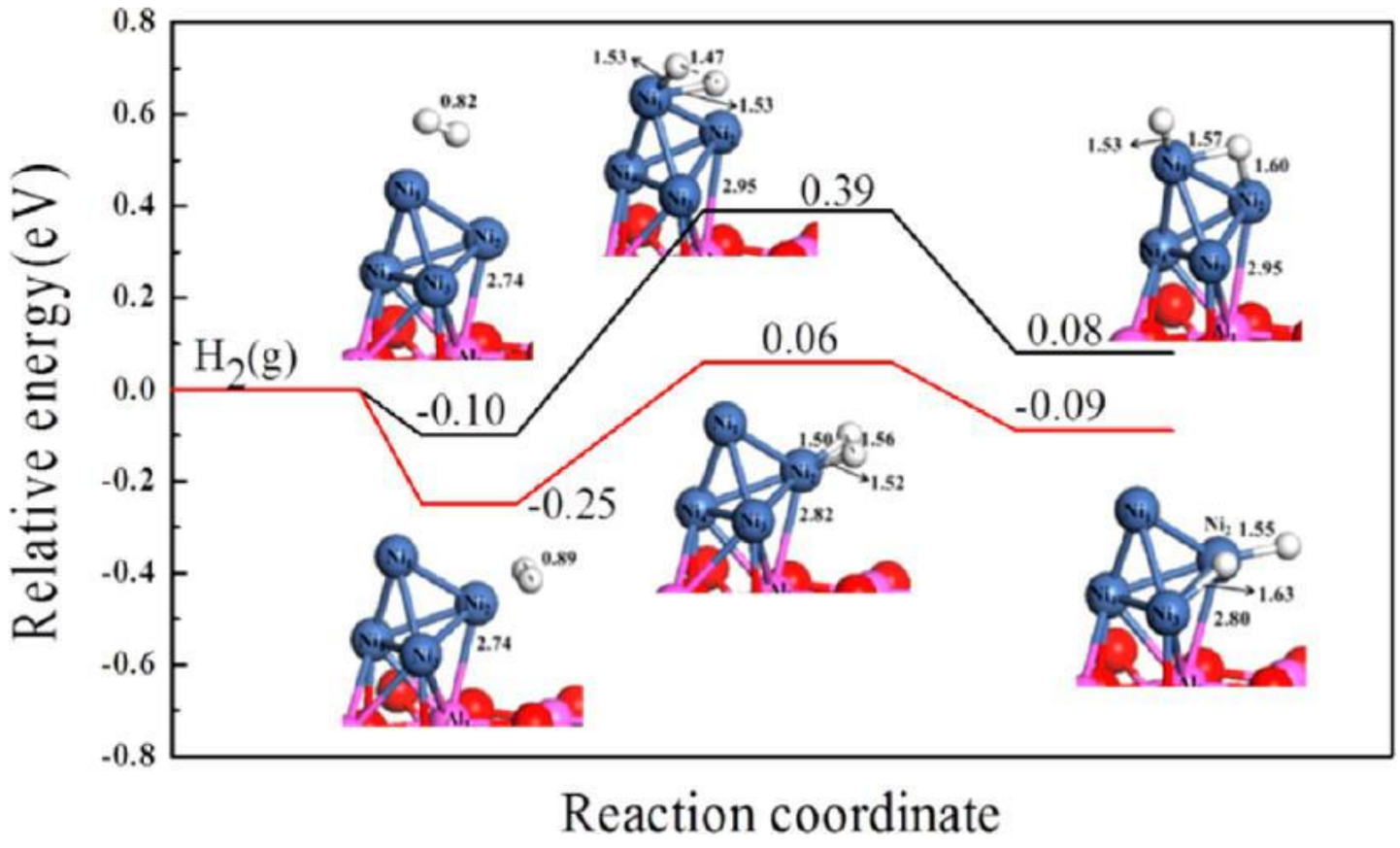


Nature Communications **10**, 2546 (2019)

CH_4 and H_2 dissociation on Ni/γ-Al_2O_3

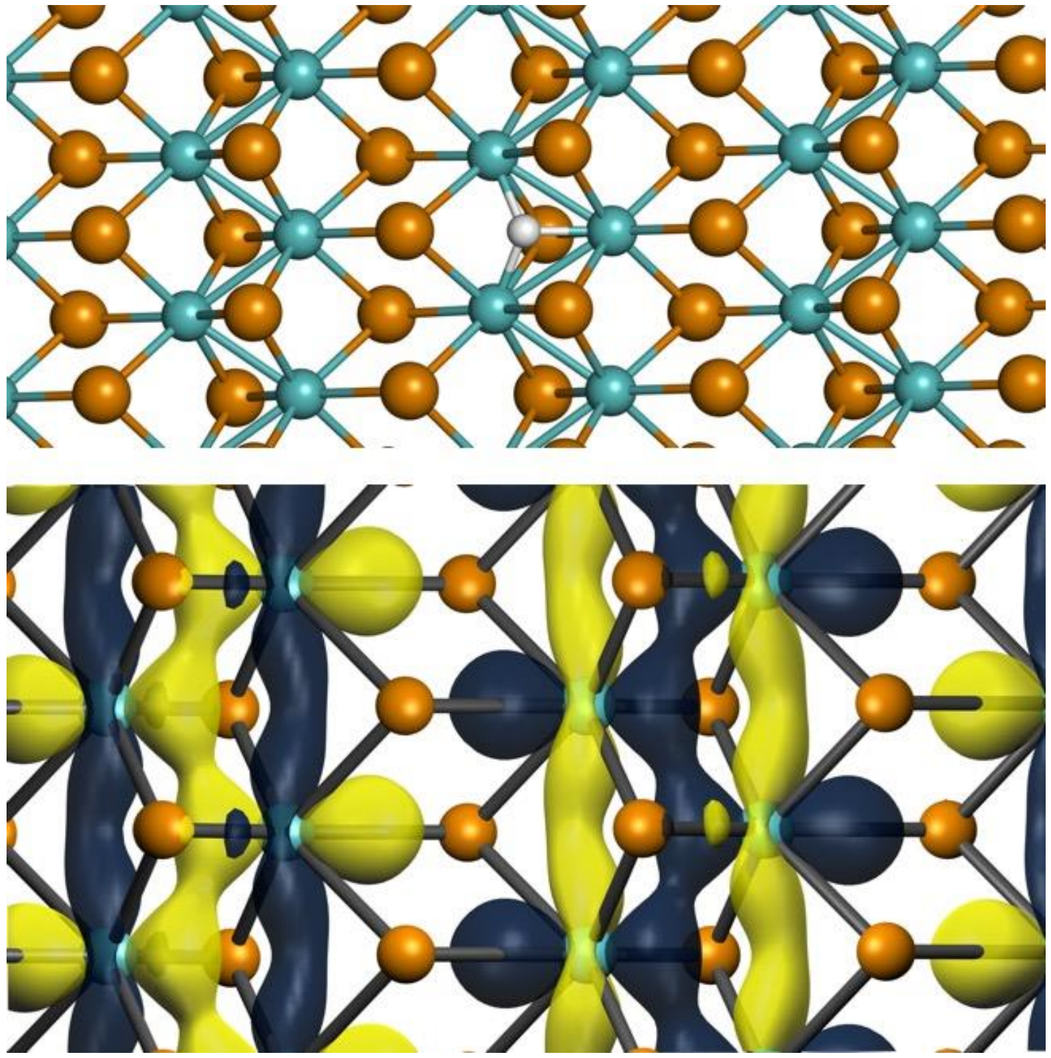
- Dissociation at TMO – NP interface preferred
- Aluminum acts as electron donor
- Need 2D to polarize surface





Li, Croiset, Ricardez-Sandoval, *J. Phys. Chem. C* **2013**, *117*, 16907

Electrochemical activation $MoTe_2$ for H_2 evolution



- 1T′-$MoTe_2$ catalyst improved at bias
- 2D surfaces + COSMO solvation
- H adsorbs on Te at cathodic bias

The rapid electrochemical activation of $MoTe_2$ for the hydrogen evolution reaction Nature Comm. 2019

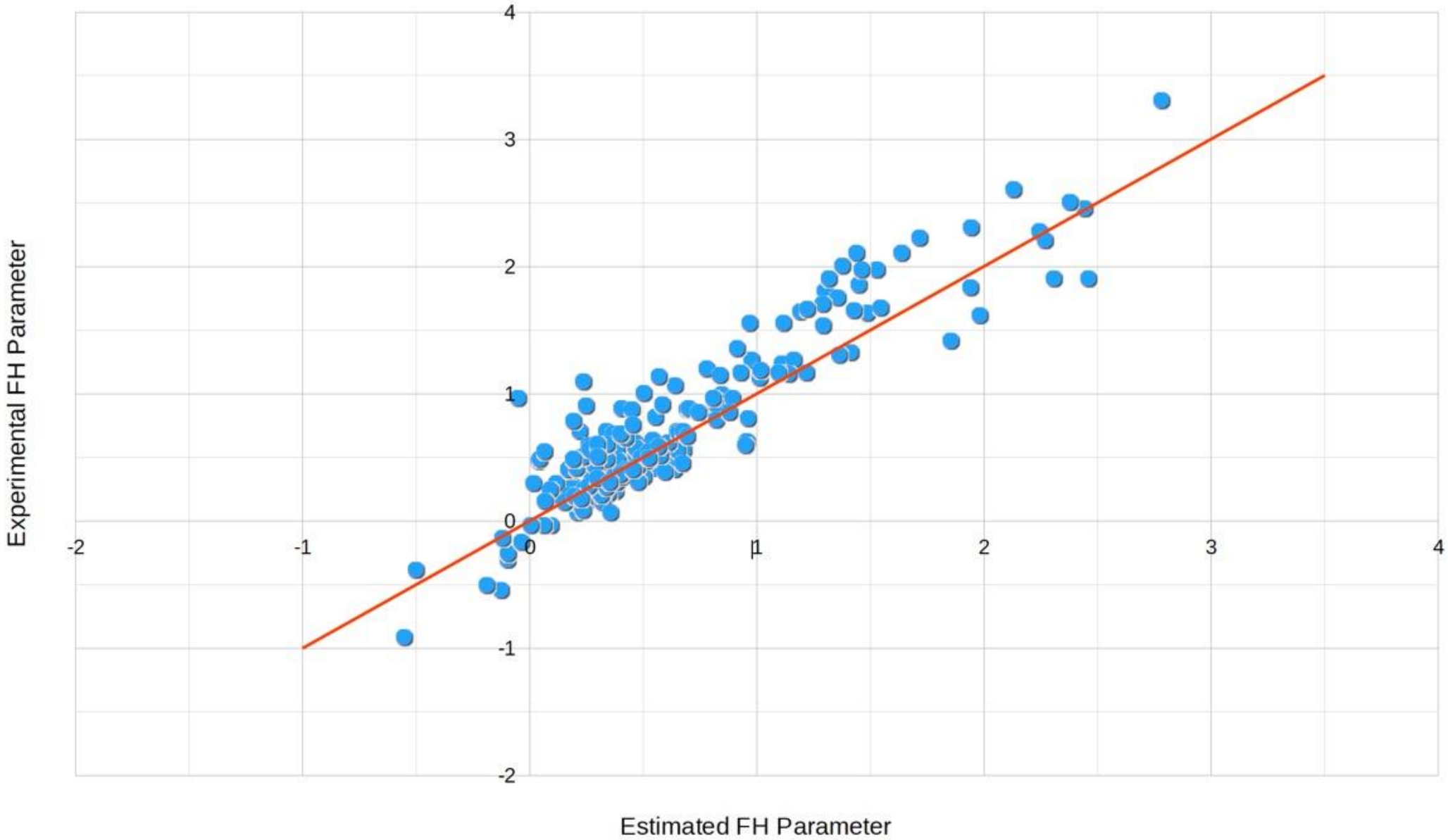
Flory-Huggins from COSMO-RS

$N_{p/s}$ – *Number of moles of polymer / solvent*

$\phi_{p/s}$ – *Volume fraction of polymer / solvent*

χ_{ps} – *Flory – Huggins Parameter*

$$\frac{\Delta G_{mix}}{RT} = N_p \ln(\phi_p) + N_s \ln(\phi_s) + N_s \phi_p \chi_{ps}$$

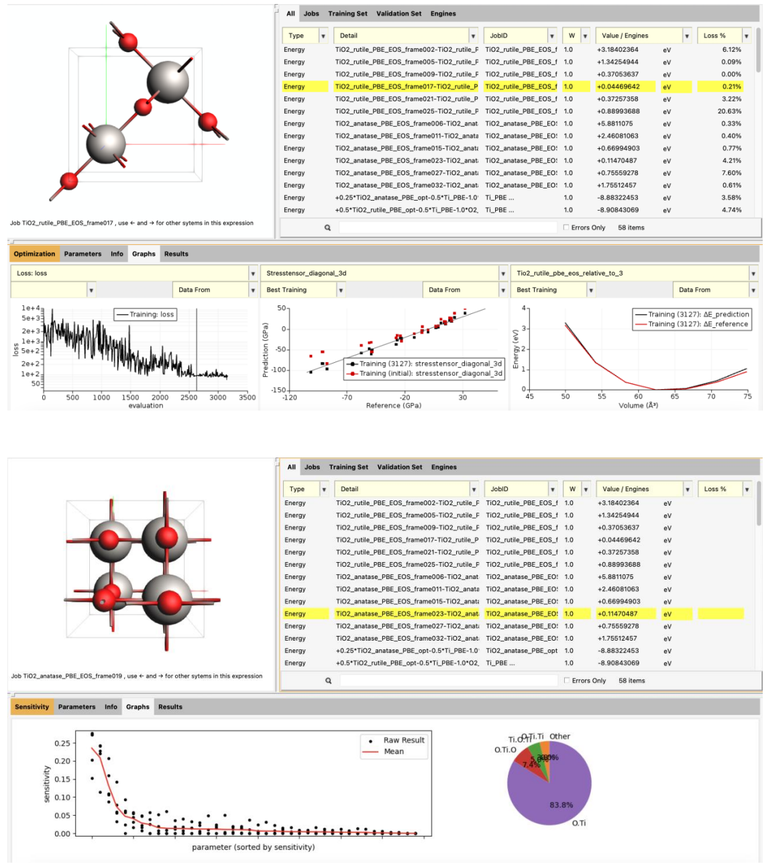


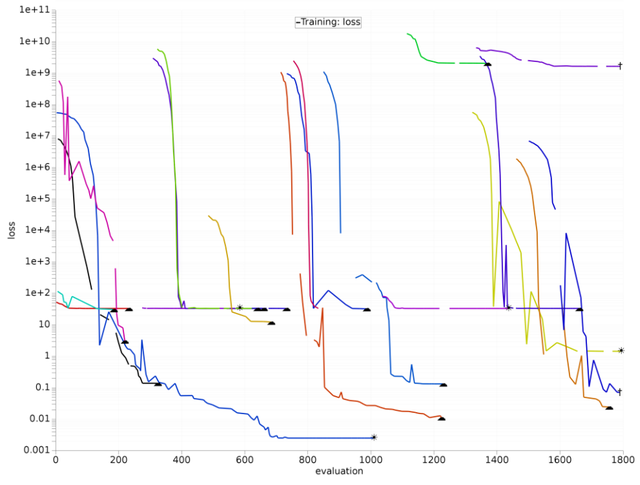
Training FF with ParAMS

Making parameters optimization simple

What can you do with ParAMS?

- Import, build and visualize training data
- Use data from AMS, VASP, QE, and experiments
- Tune parameters to describe energies, forces geometries, stress tensor, charges, bandgaps, etc.
- Use validation sets to prevent overfitting
- Submit multiple optimization at the same time to explore parameters space
- Explore the sensitivity of the parameters to select the most important subset to optimize
- Train DFTB, ReaxFF, Force field, and … MLP

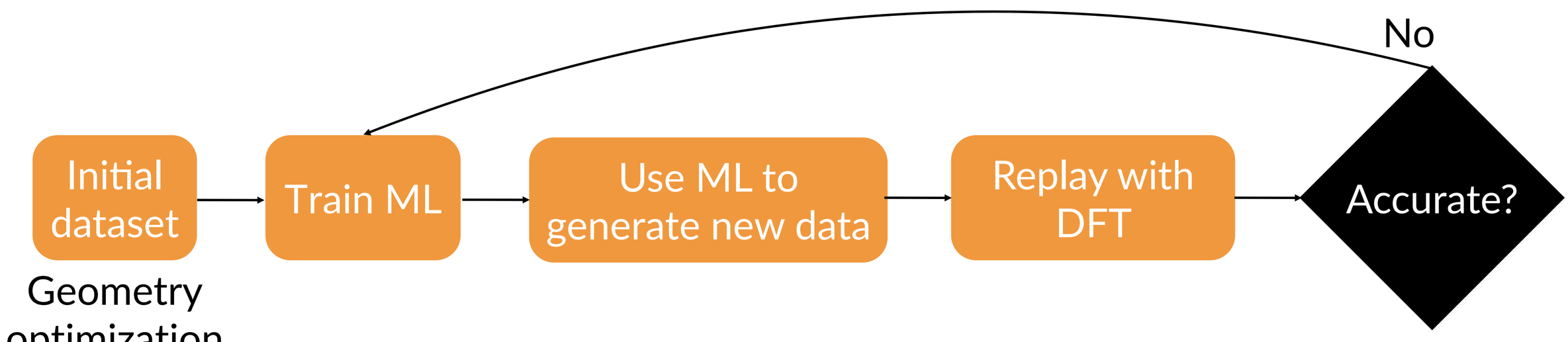


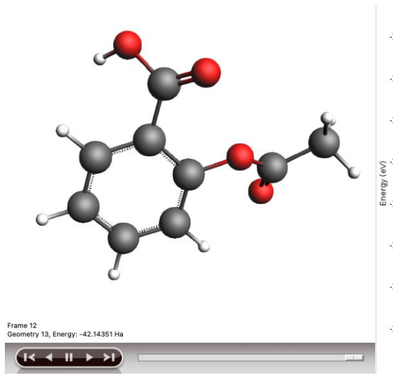


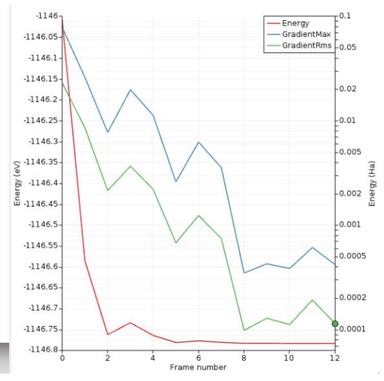
J. Chem. Inf. Model. 2021, 61, 8, 3737–3743
J. Chem. Theory Comput. 2023, 19, 9, 2557–2573
https://www.scm.com/product/params

Training MLP with ParAMS

Active learning workflow

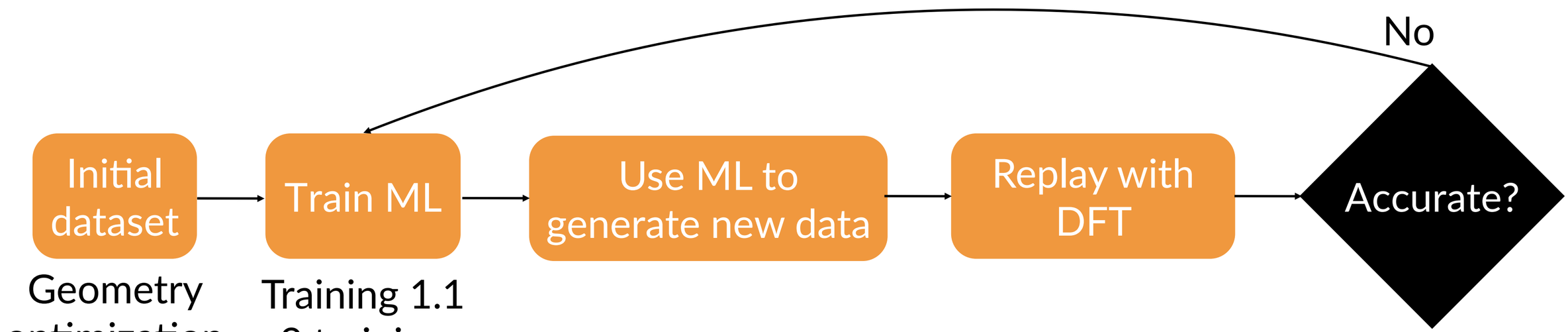


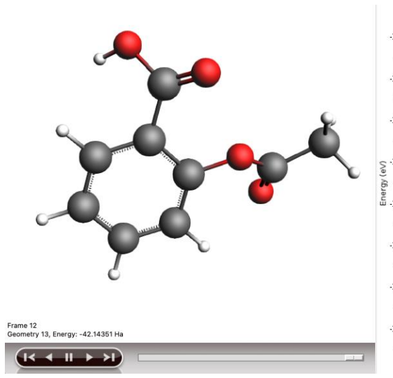


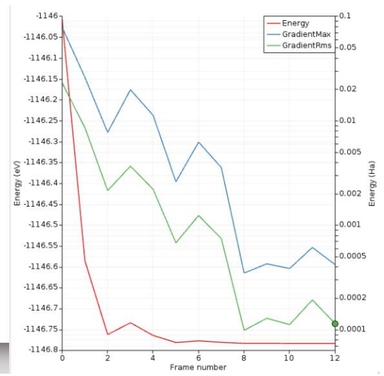


Training MLP with ParAMS

Active learning workflow

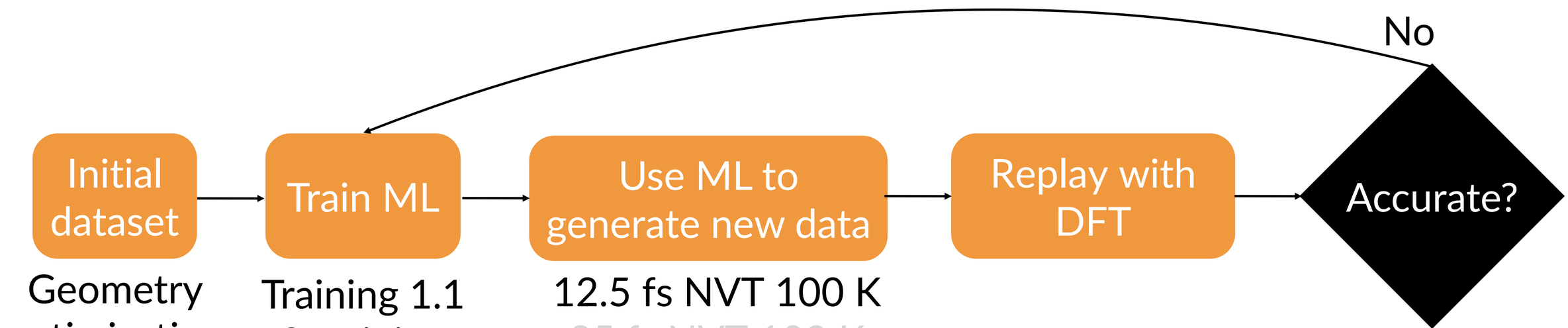


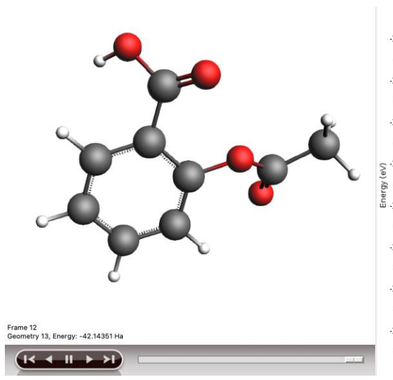


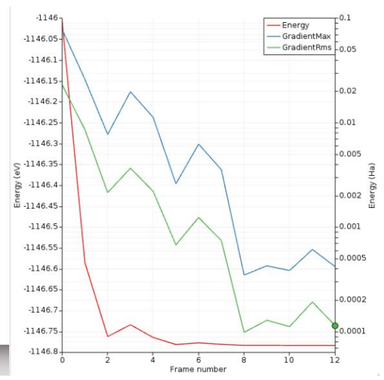


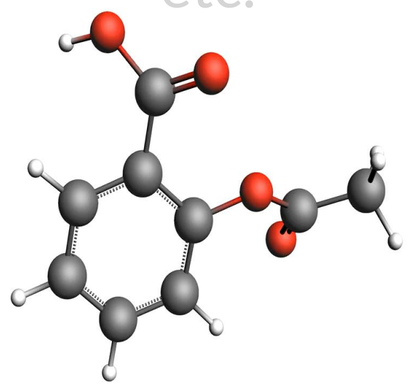
Training MLP with ParAMS

Active learning workflow



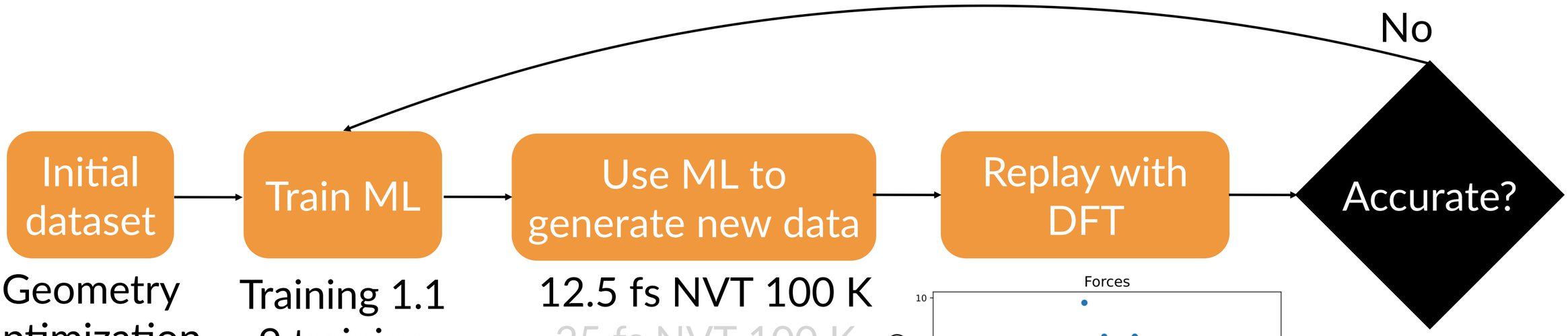


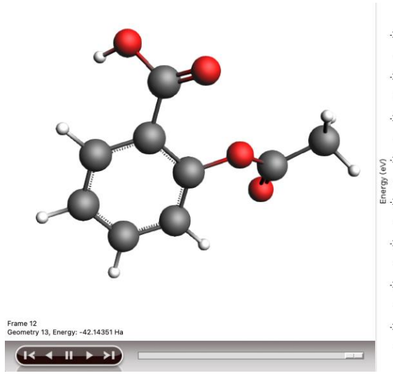


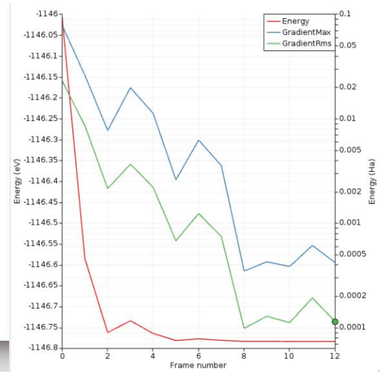


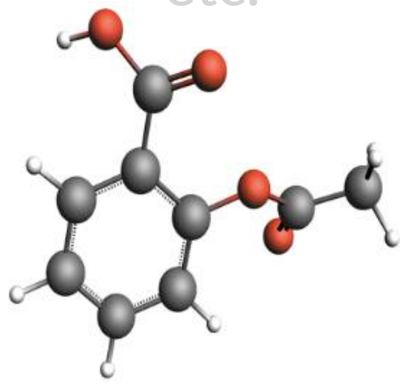
Training MLP with ParAMS

Active learning workflow





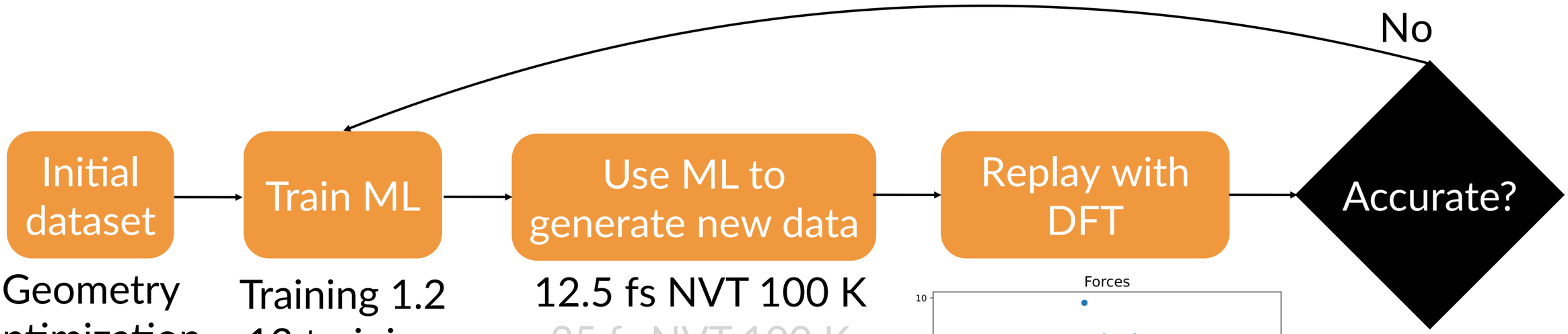




MAE = 1.83 eV/Å

Training MLP with ParAMS

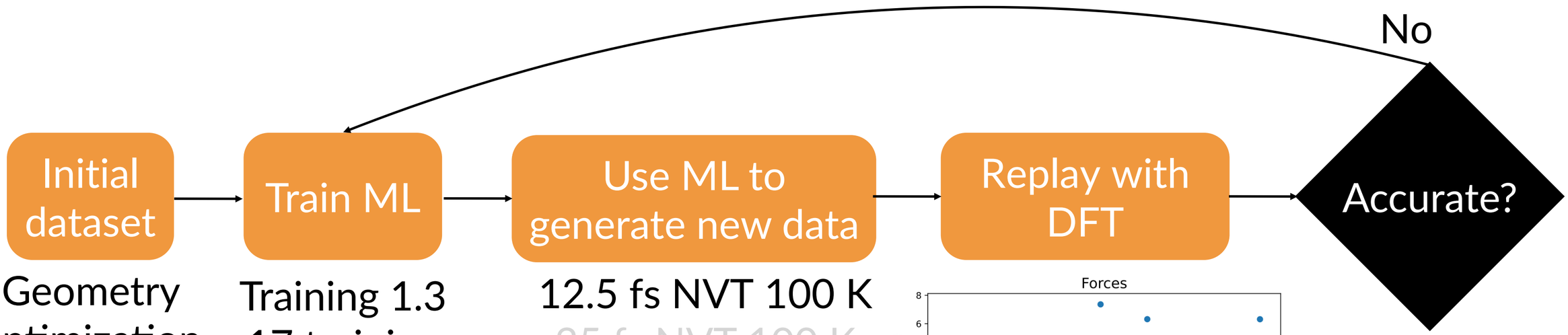
Active learning workflow

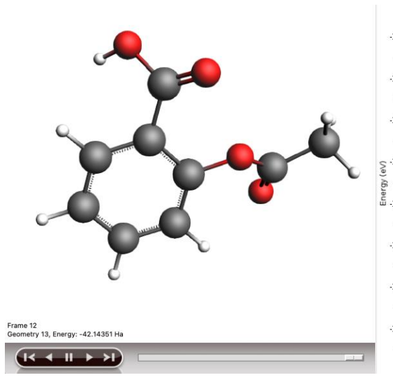


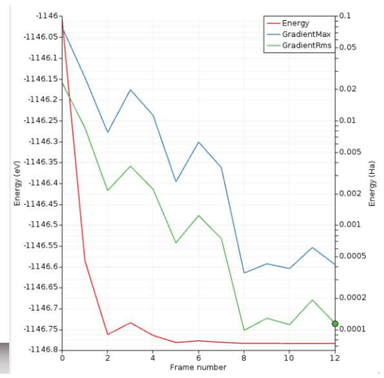
MAE = 1.83 eV/Å

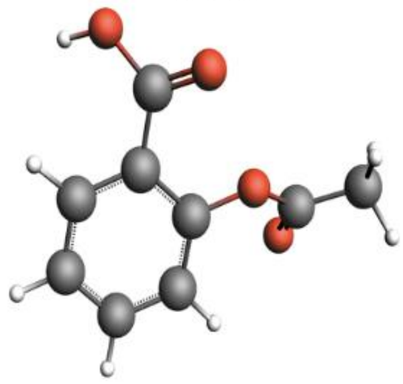
Training MLP with ParAMS

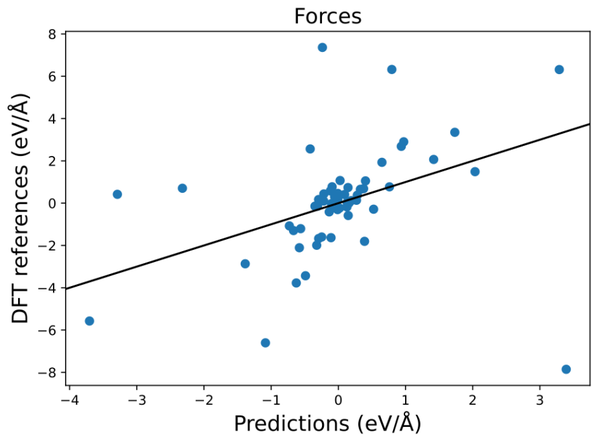
Active learning workflow







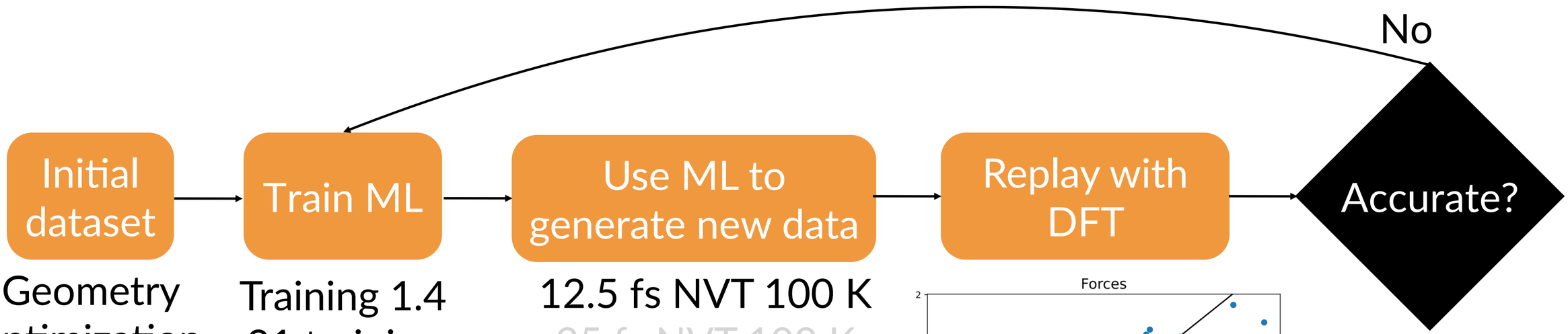


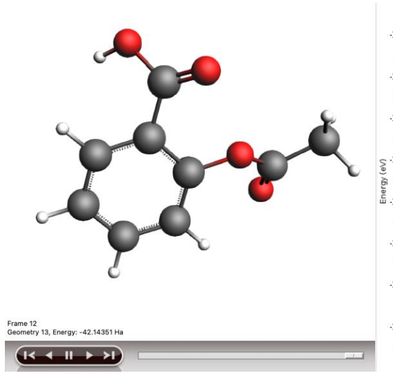


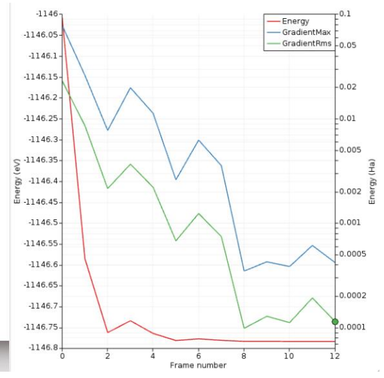
MAE = 1.32 eV/Å

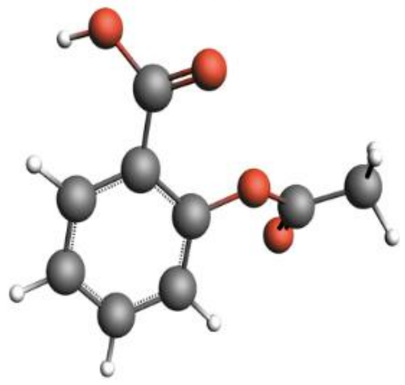
Training MLP with ParAMS

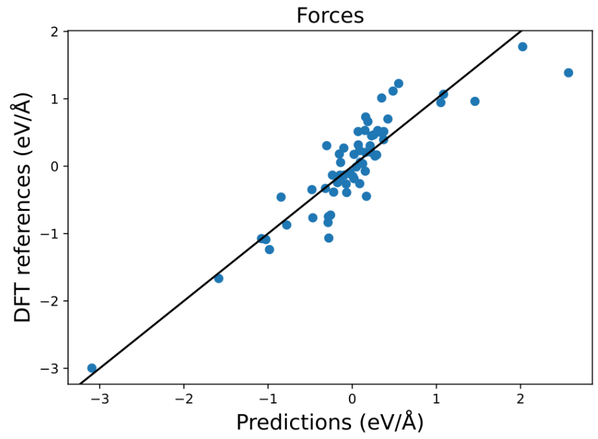
Active learning workflow







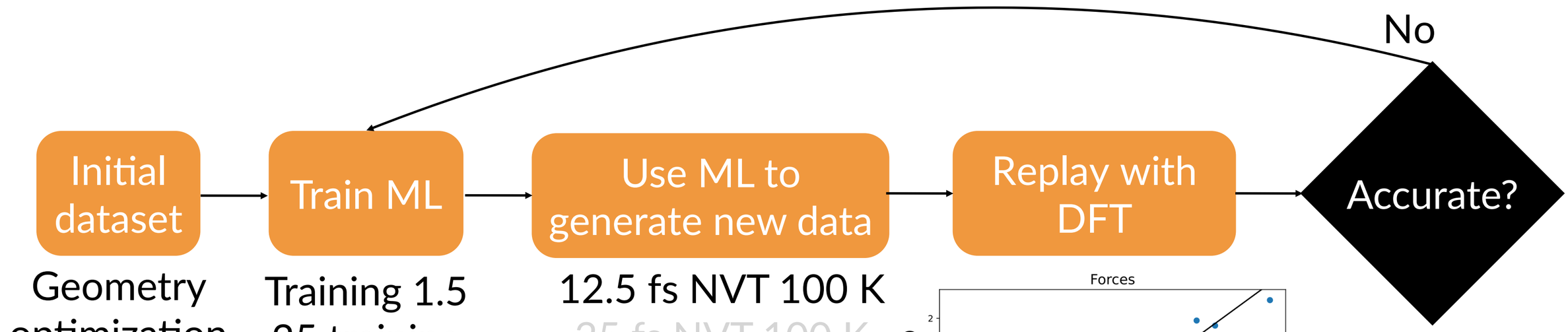


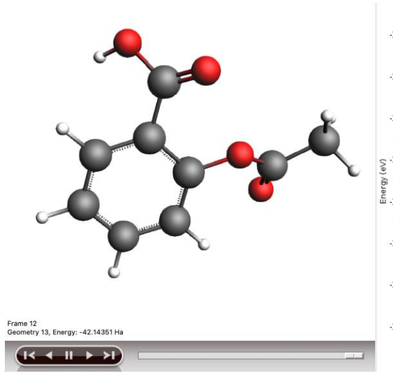


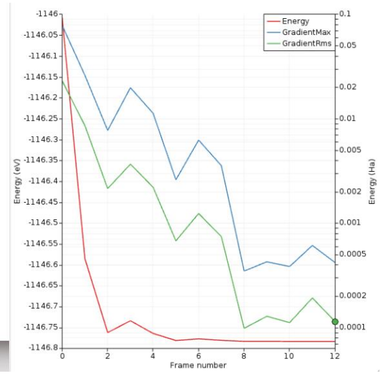
MAE = 0.26 eV/Å

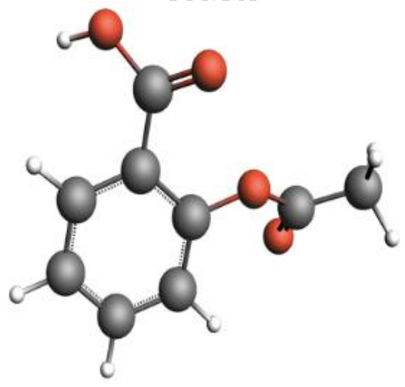
Training MLP with ParAMS

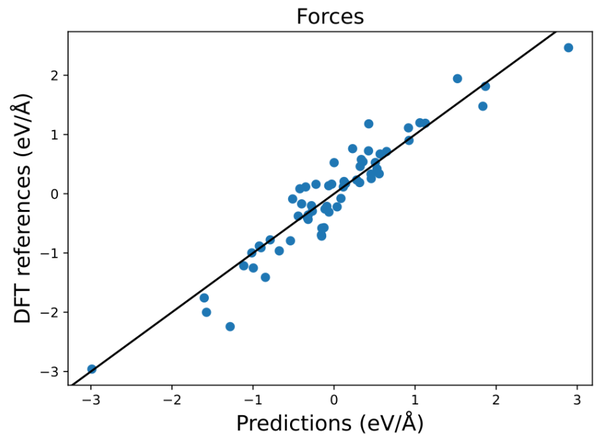
Active learning workflow







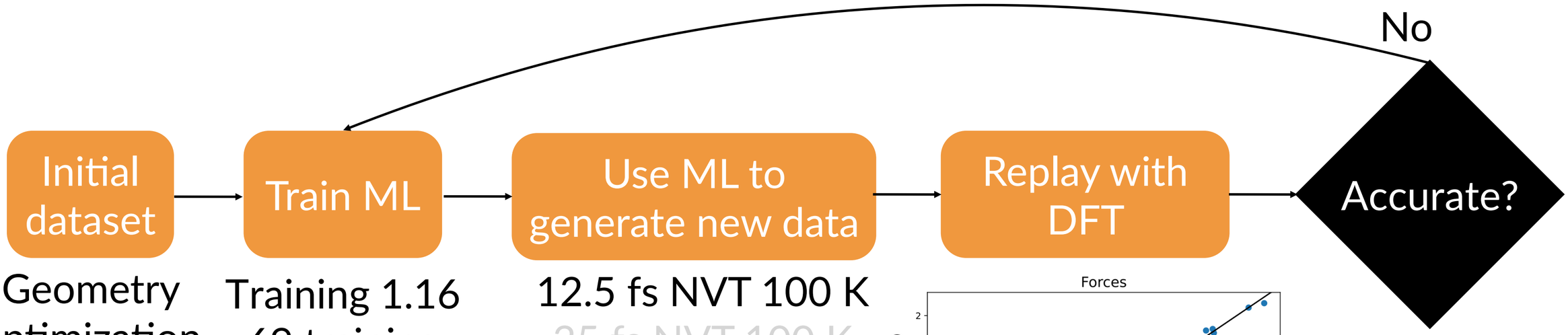


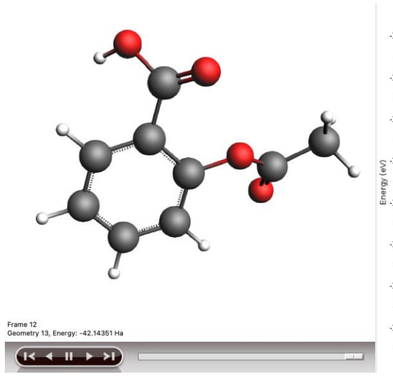


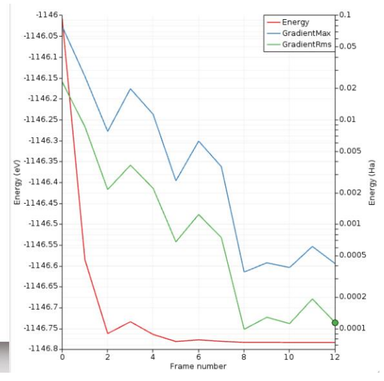
MAE = 0.25 eV/Å

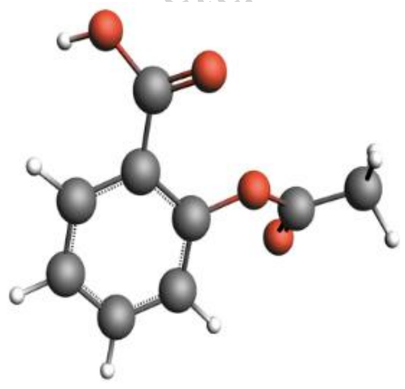
Training MLP with ParAMS

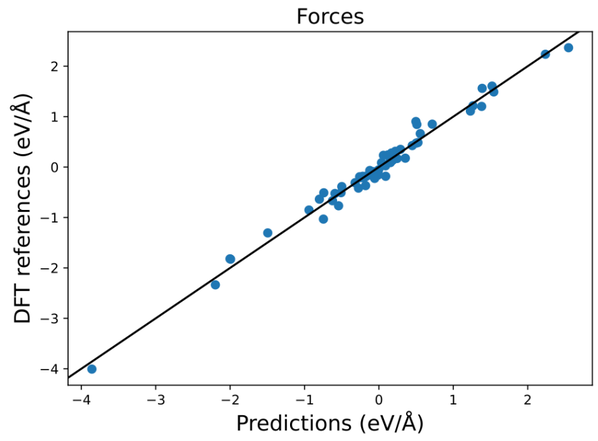
Active learning workflow







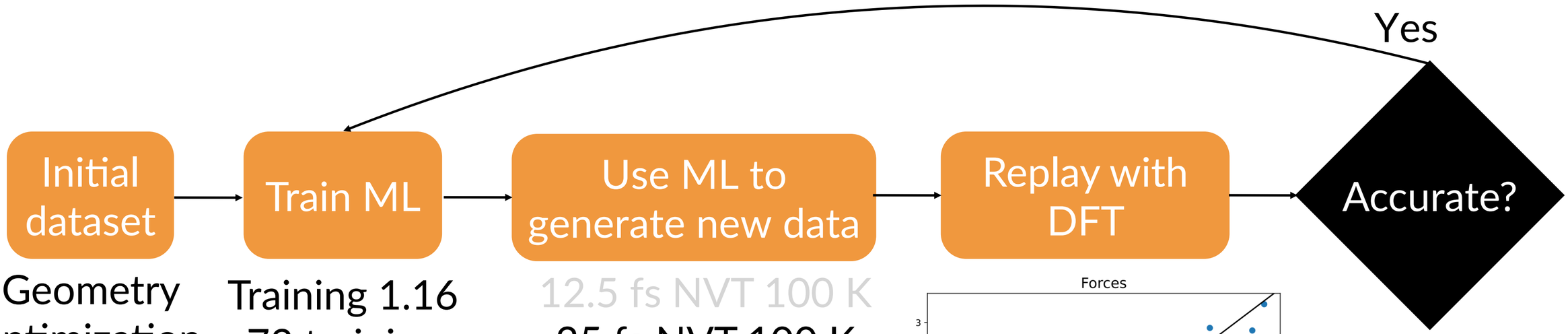




MAE = 0.10 eV/Å

Training MLP with ParAMS

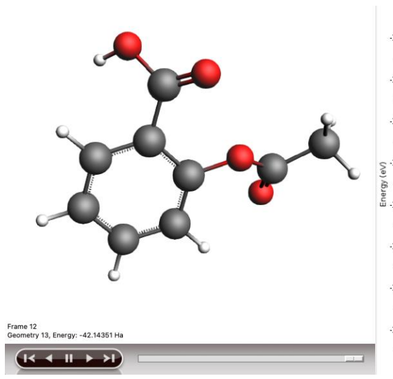
Active learning workflow

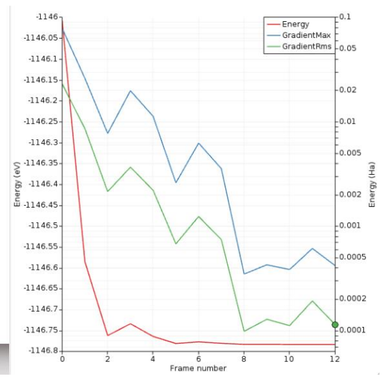


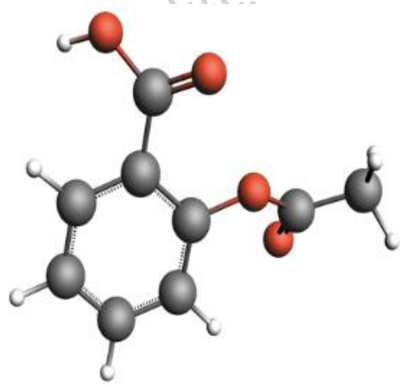
Geometry optimization ~10 steps

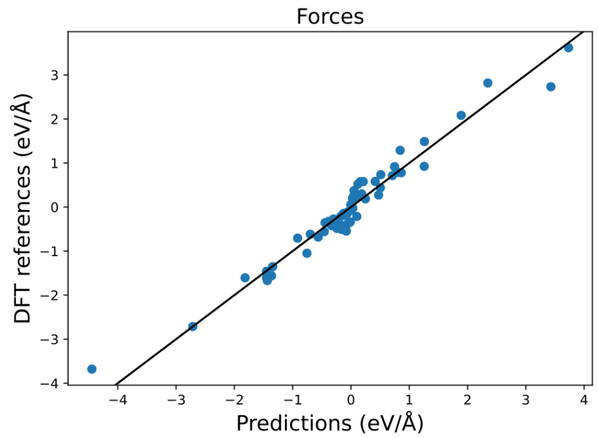
Training 1.16
73 training
21 validation

12.5 fs NVT 100 K
25 fs NVT 100 K
50 fs NVT 100 K
etc.





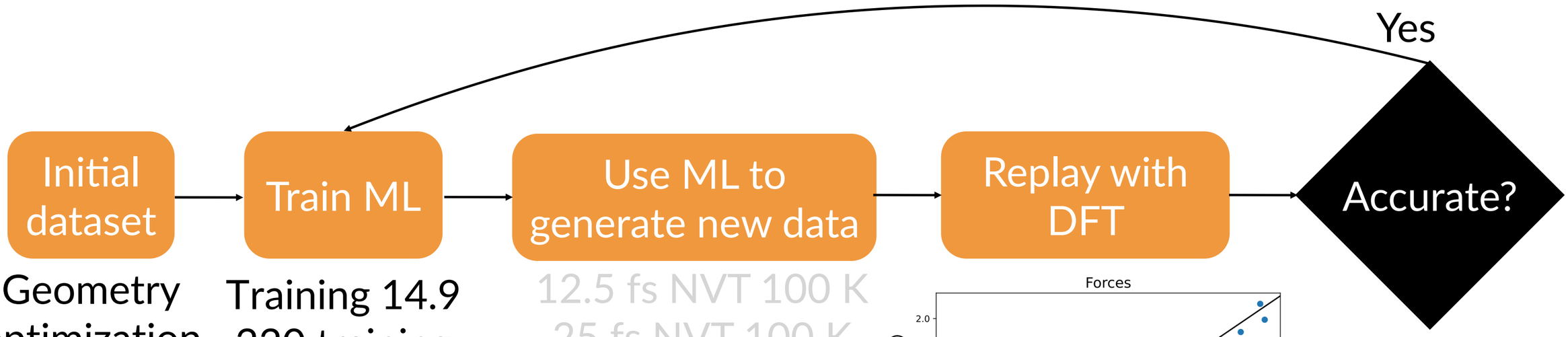


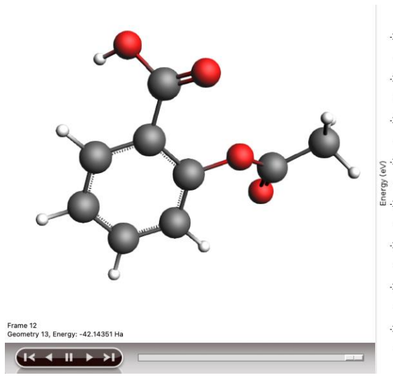


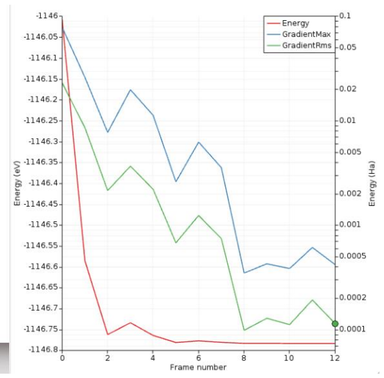
MAE = 0.18 eV/Å

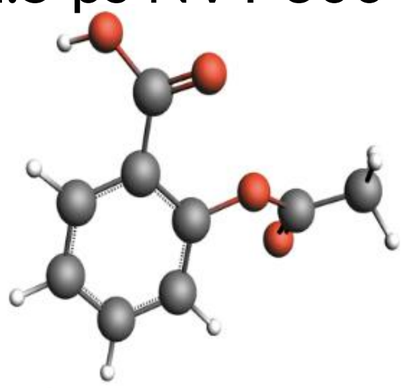
Training MLP with ParAMS

Active learning workflow

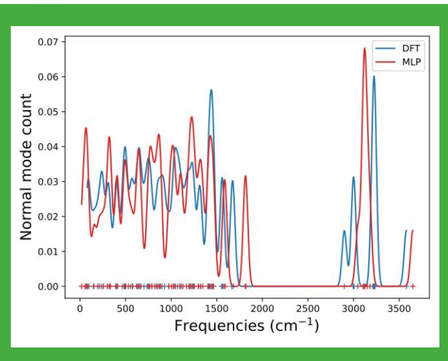




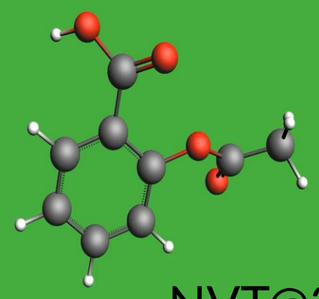




MAE = 0.05 eV/Å



Final model



NVT@300K

amshome/scripting/scm/params/examples/ActiveLearning

Training MLP with ParAMS

Summary

- Compute the initial training set of reference calculations
- Select the ML model to optimize (nequip, flare)
- Define the series of test tasks (default/custom)
- Run an interactively follow the training procedure
  - MAE plot
  - Parity plot
  - Dynamics