Sandia National Laboratories
Annual ML/DL Workshop

September 9-12, 2024

Physics-based and Graph-based Machine Learning Models for Surface Property Predictions

Dr. Peter Schindler

Assistant Professor at Northeastern University
(Dept. of Mechanical & Industrial Engineering)

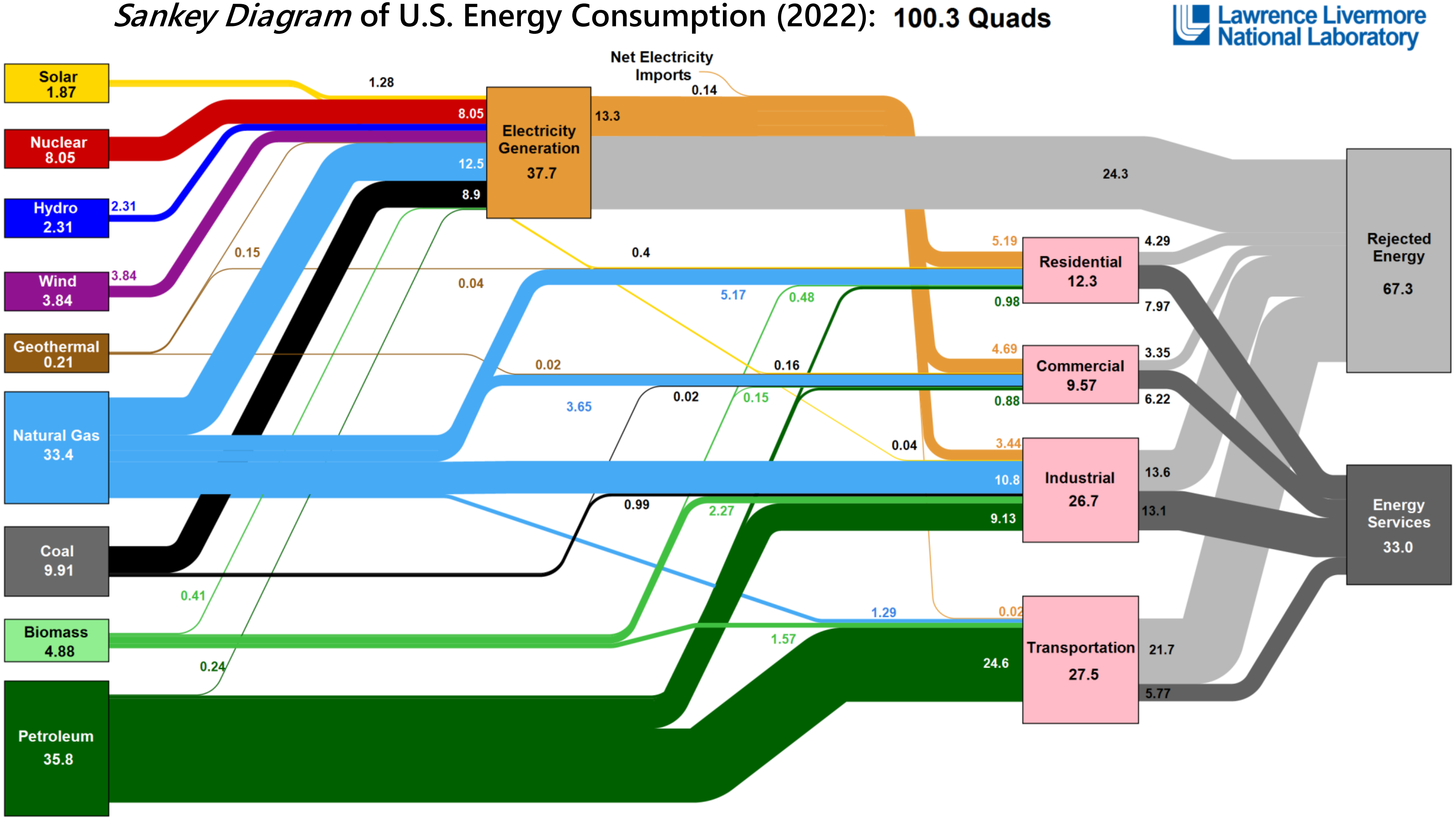
Northeastern University

p.schindler@northeastern.edu
https://www.d2r2group.com

Data Driven Renewables Research

Majority of Energy Goes to Waste(-Heat)

Sankey Diagram of U.S. Energy Consumption (2022): **100.3 Quads**

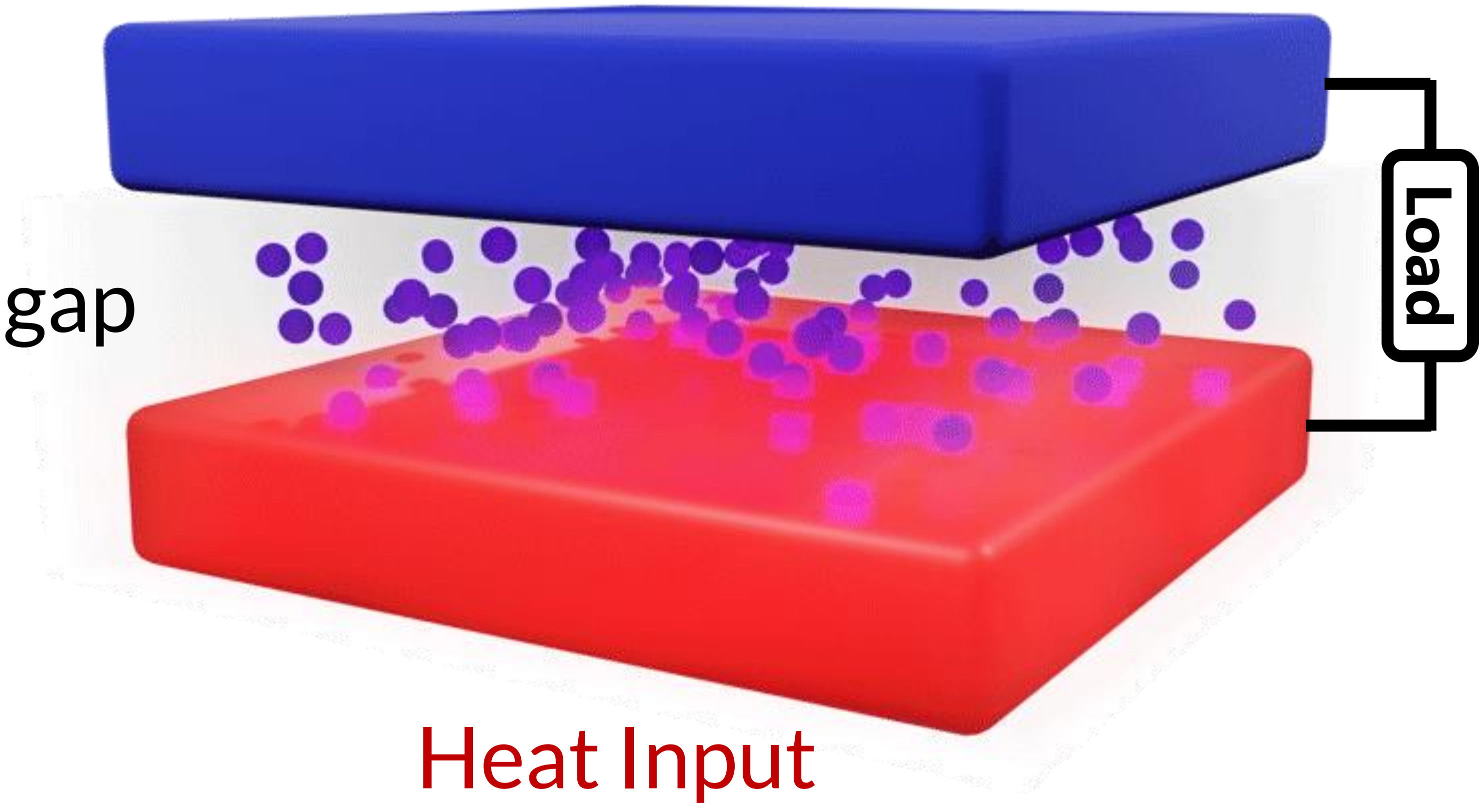


Thermionic Energy Converter (TEC)

Anode

Vacuum gap

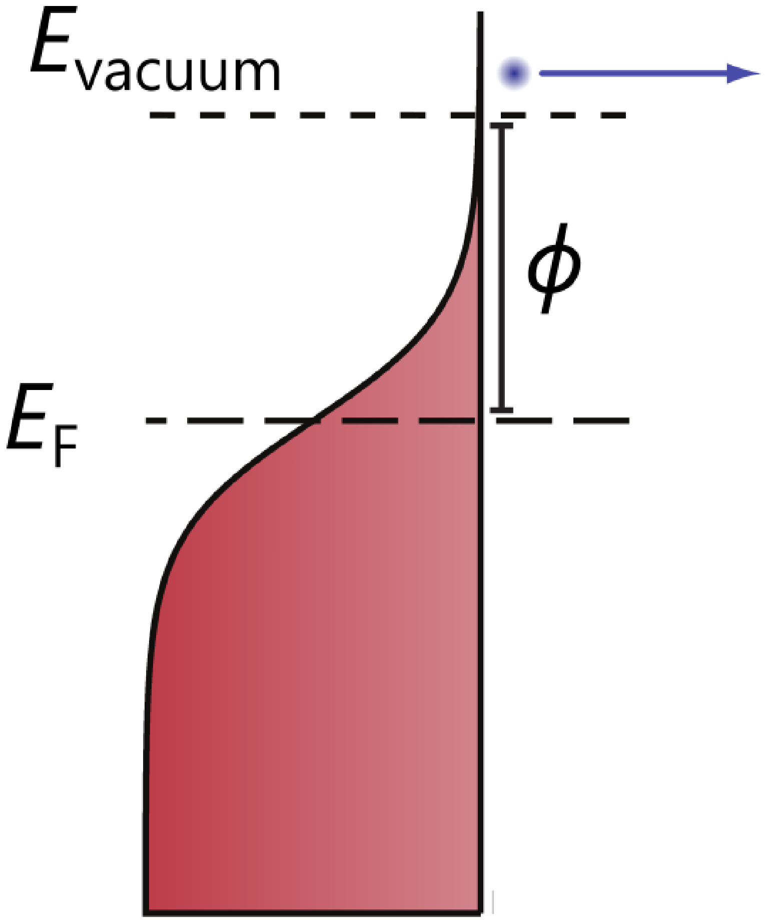
Cathode

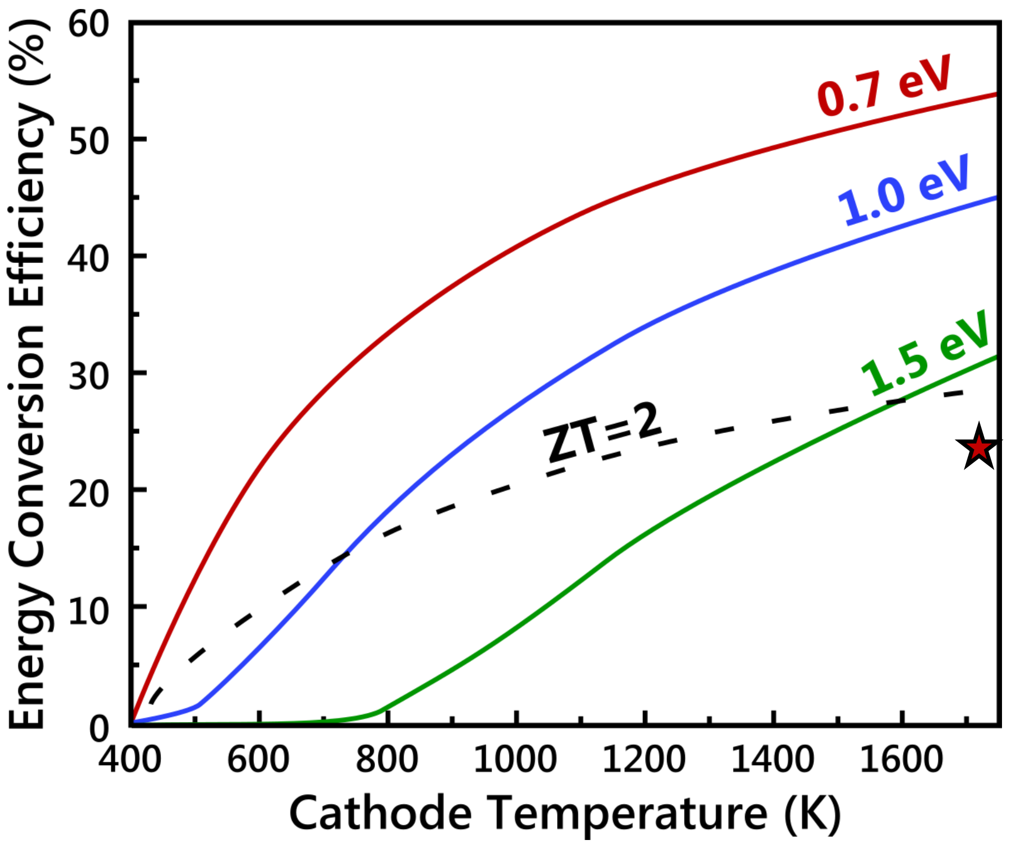


Heat Input

- No moving parts
- Power output scales with area

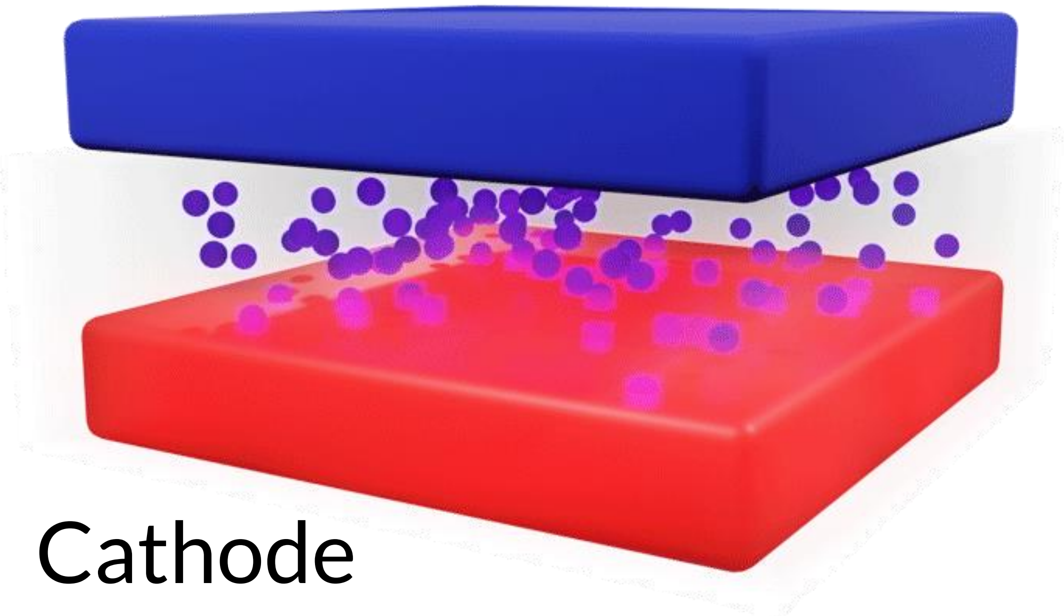
TEC Efficiency





Question: Which Electrode in a TEC Requires Ultra-low Work Function?

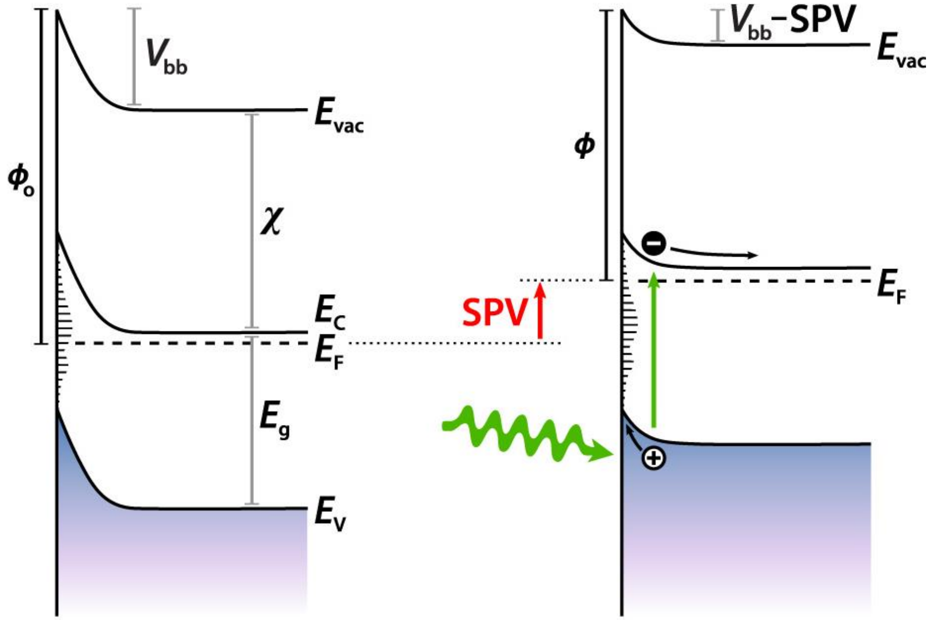
Anode

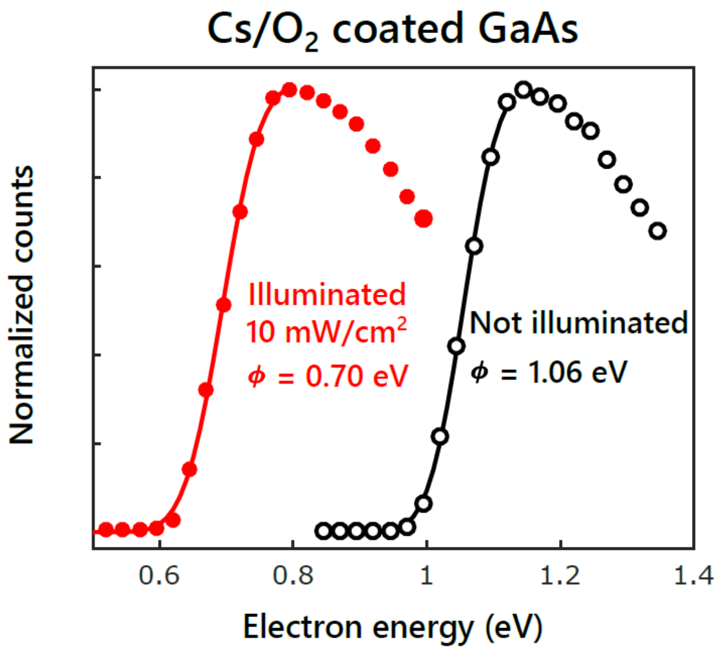


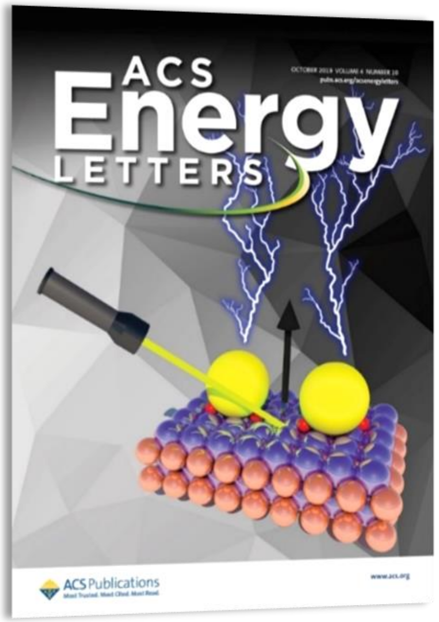
Cathode

State-of-the-Art Low Work Function Surfaces

WF < 2 eV typically require **alkaline coatings** or **non-steady state conditions**



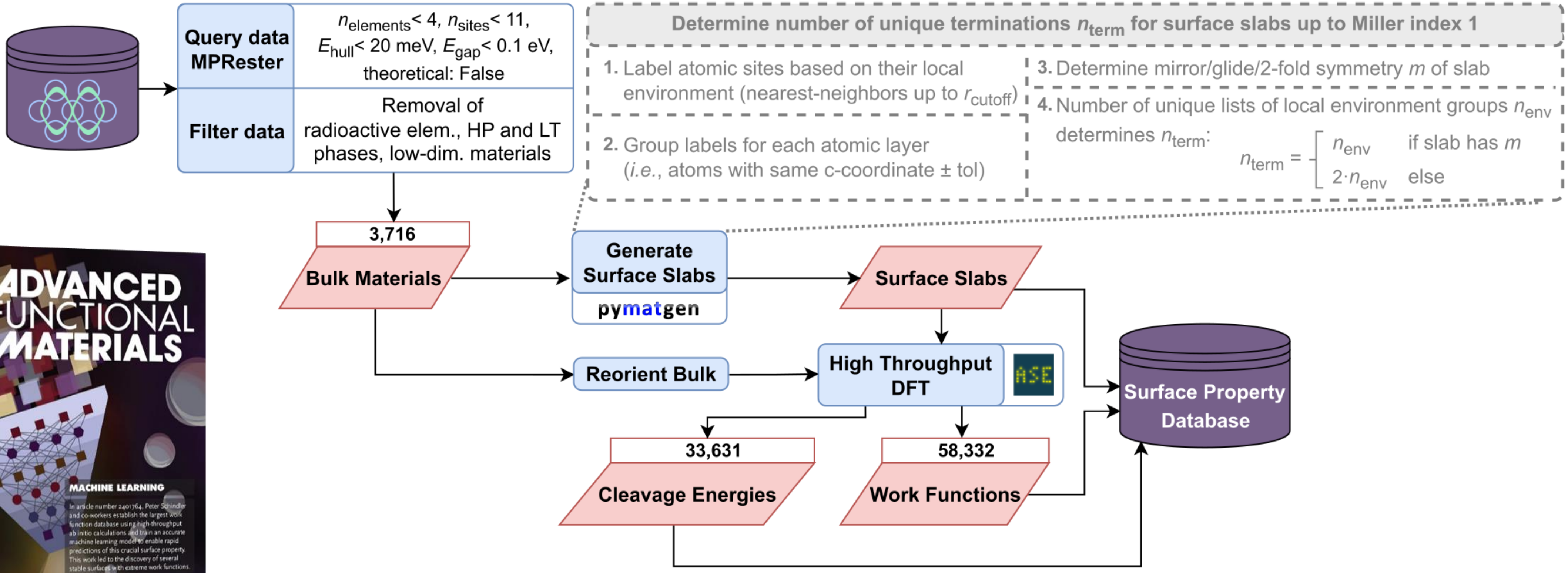


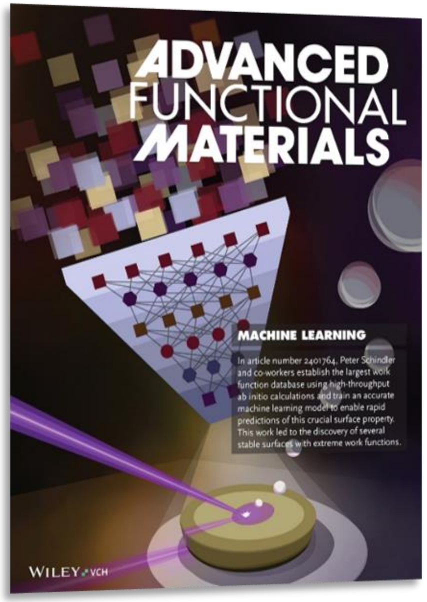


P. Schindler, *et al. ACS Energy Letters* 4, 10 (2019)

Data-driven Approach, Part I: Work Function Database

No large databases of work functions in literature (except for elemental crystals and 2D materials)

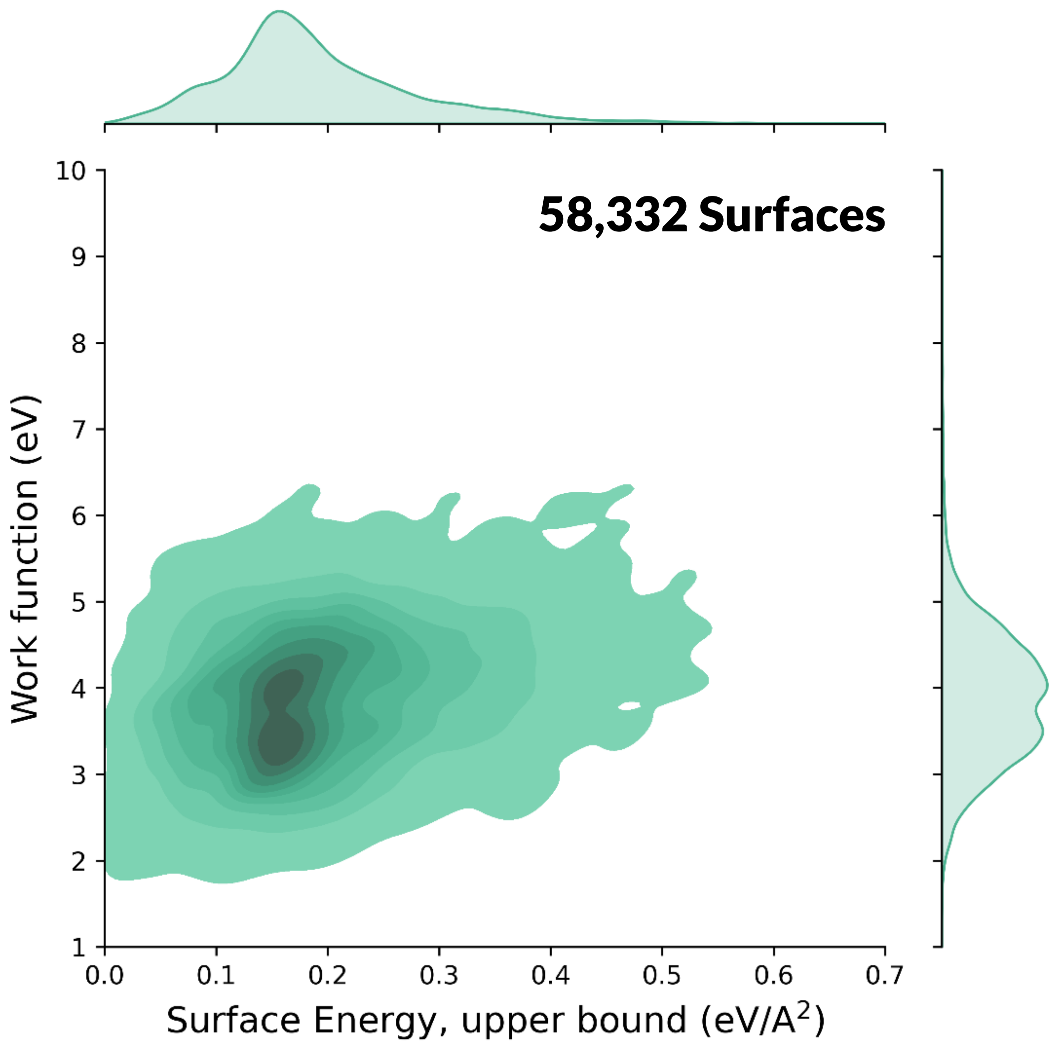


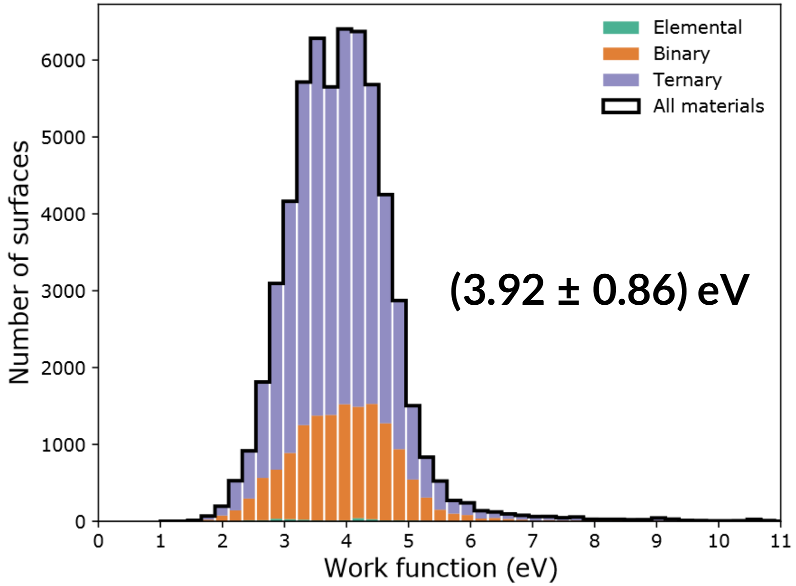


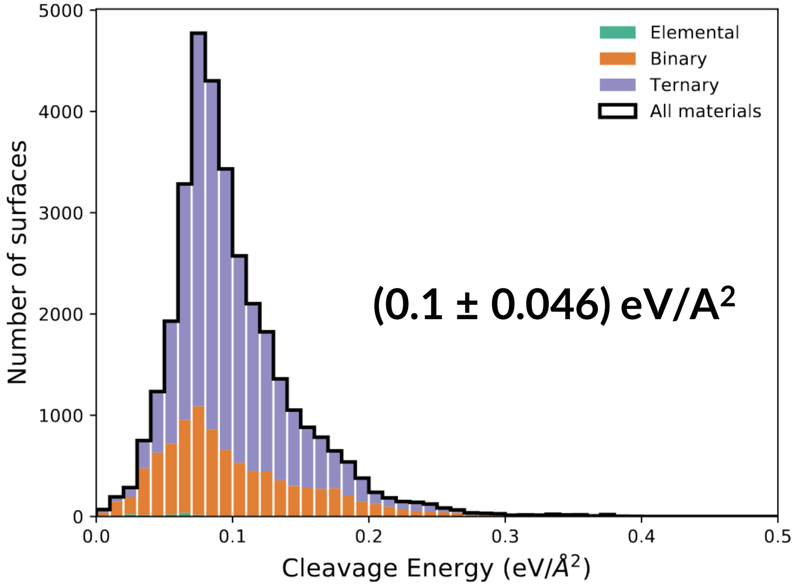
$$E_{cleavage} = \frac{E_{slab} - n_{bulk} \cdot E_{bulk}}{2 \cdot A_{slab}} = \frac{\gamma_{top} + \gamma_{bottom}}{2}$$

P. Schindler, et al. *Adv. Func. Mat.* 34, 19 (2024)

Work Function Database

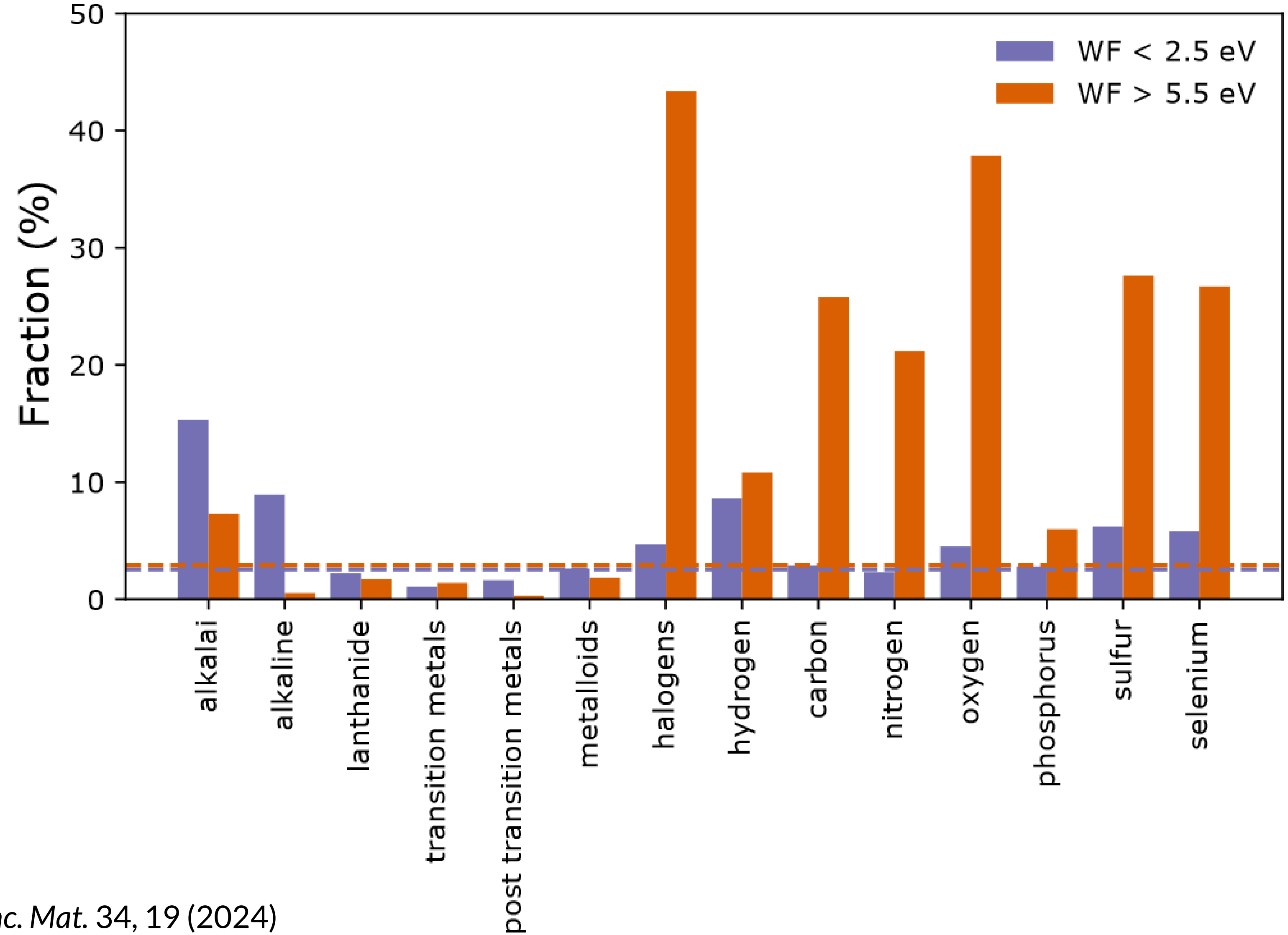






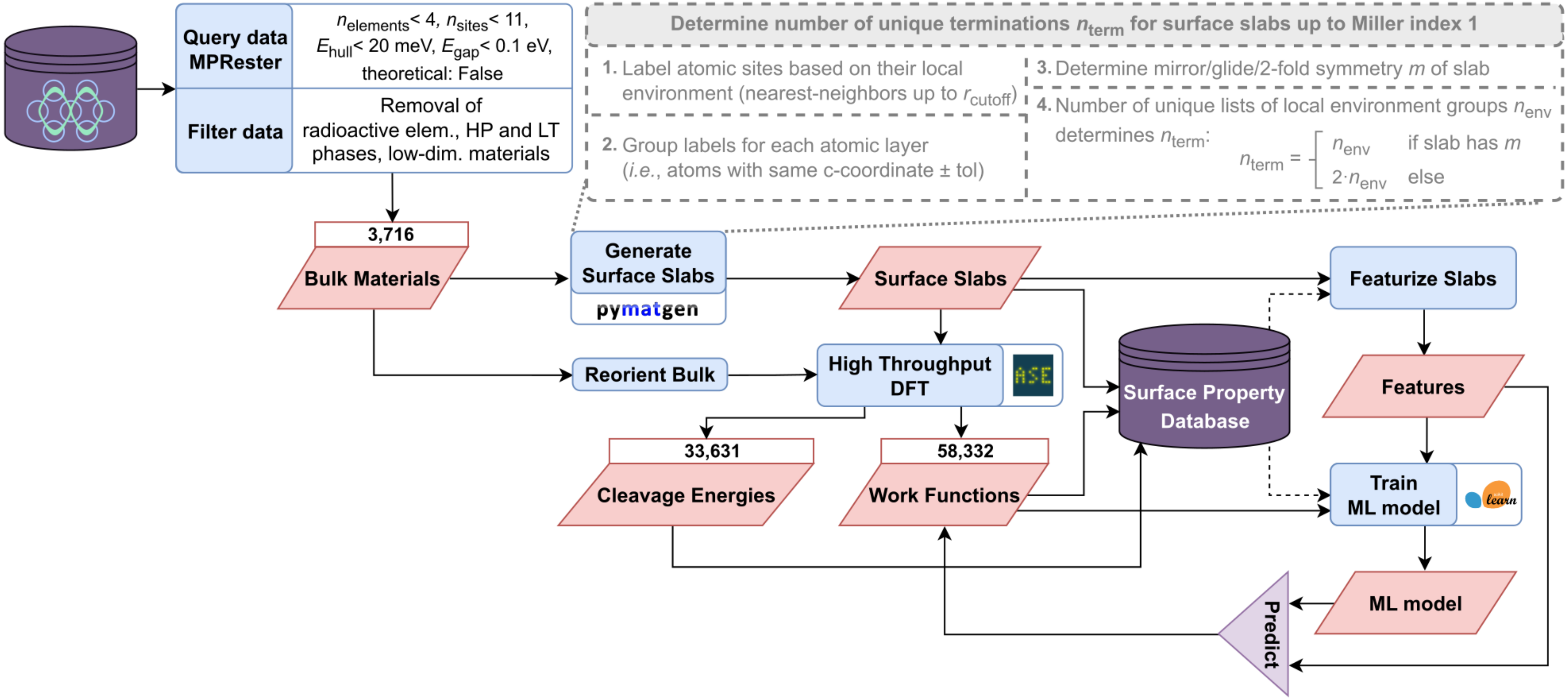
P. Schindler, et al. *Adv. Func. Mat.* 34, 19 (2024)

Work Function: Chemical Trends



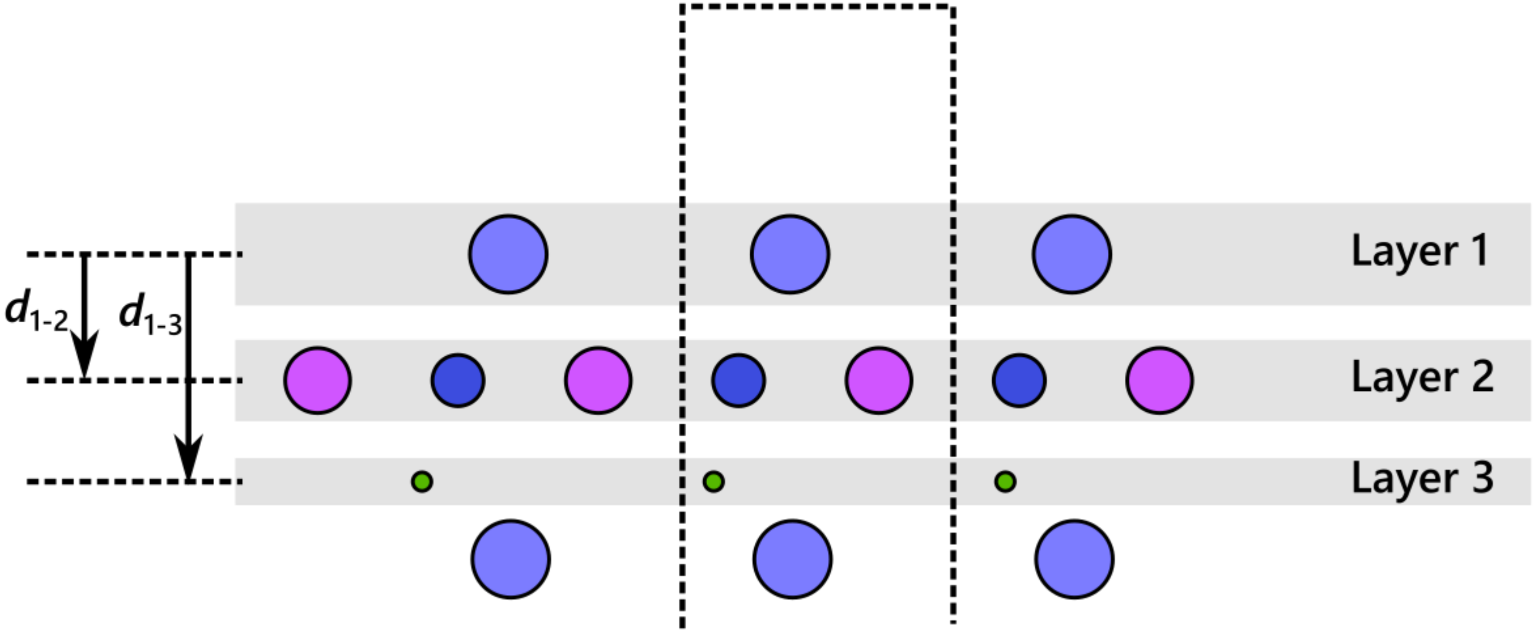
P. Schindler, et al. *Adv. Func. Mat.* 34, 19 (2024)

Data-driven Approach, Part II: Featurization & ML



P. Schindler, et al. *Adv. Func. Mat.* 34, 19 (2024)

Physically Motivated Surface Descriptors



For topmost 3 layers:

4 elemental features x (min, max, average) x 3 layers

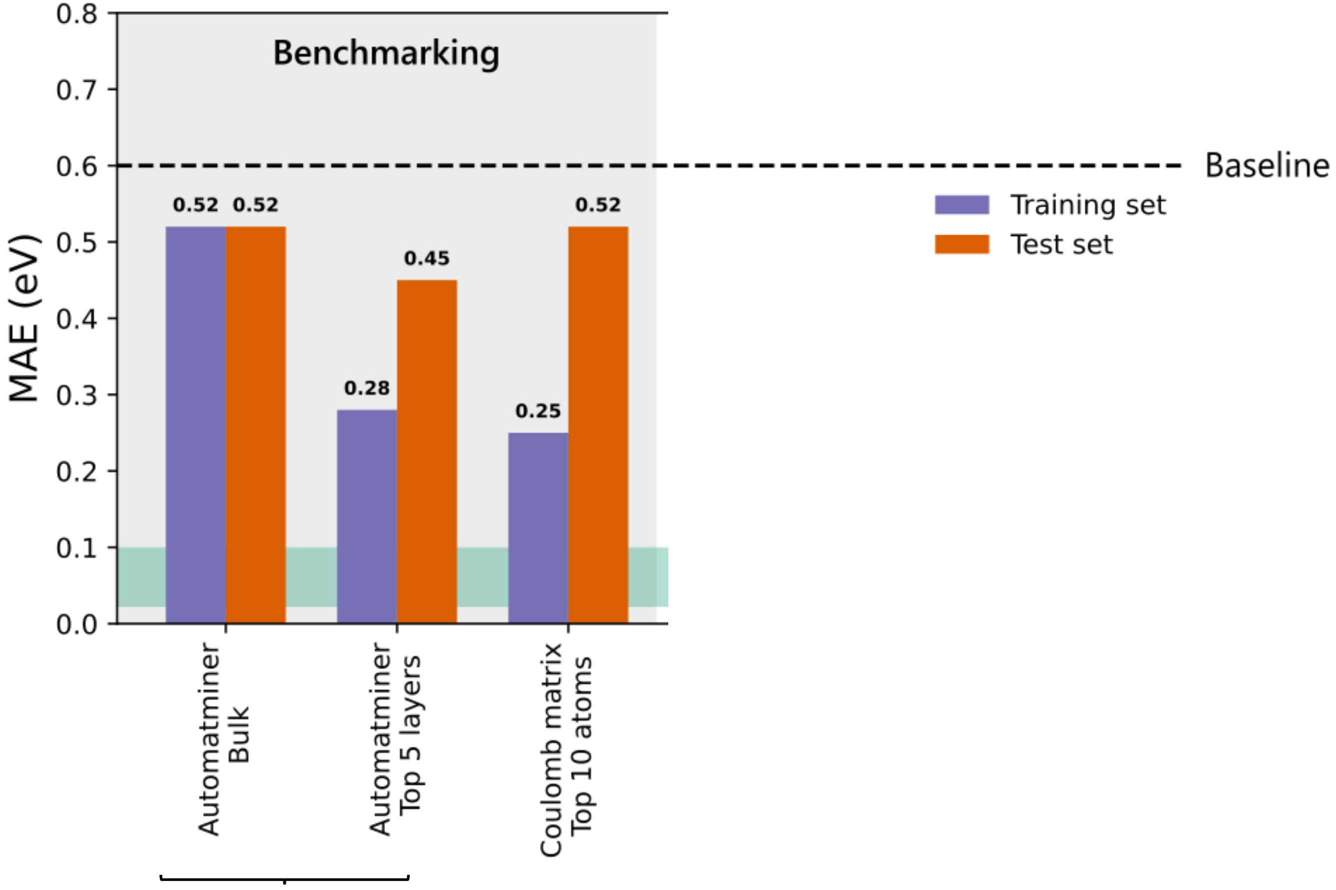
= 36

+ *structural features*: packing area (n_{atoms} / area) x 3 layers,
+ $d_{1\text{-}2}$ and $d_{1\text{-}3}$ + θ_{min} and θ_{min}

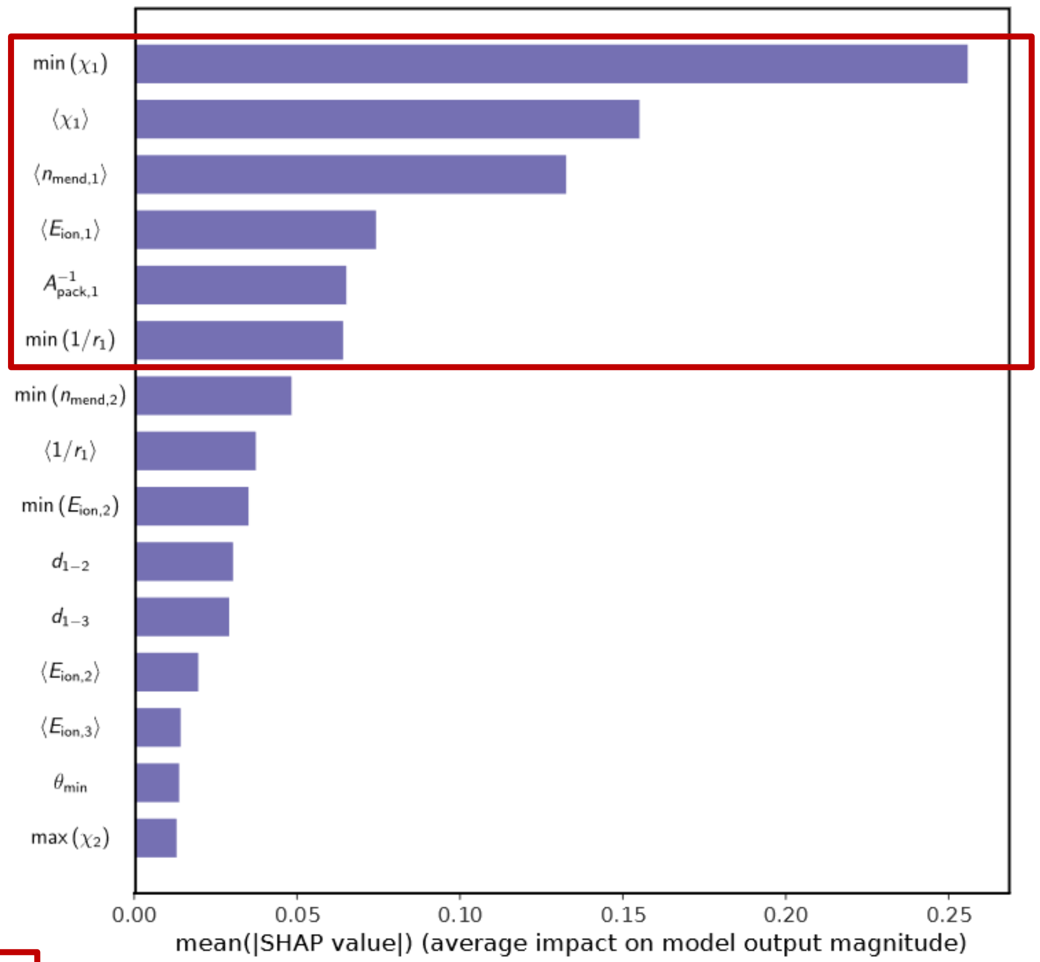
= **43** total features → pick **top 15 features** with RFE

χ
$1 / r$
$E_{ionization}$
$n_{mendeleev}$

Model Performance with Surface-based Descriptors



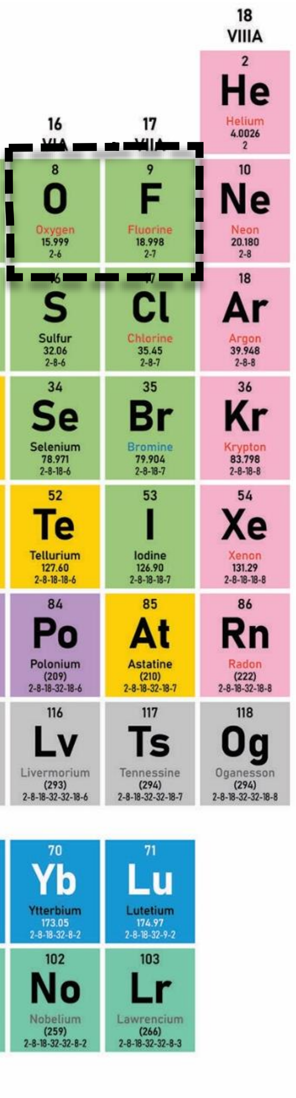
200 features



~10^5 faster than DFT

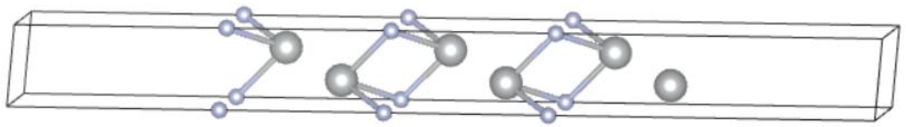
P. Schindler, et al. *Adv. Func. Mat.* 34, 19 (2024)

Promising Candidate Surfaces with Extreme WFs

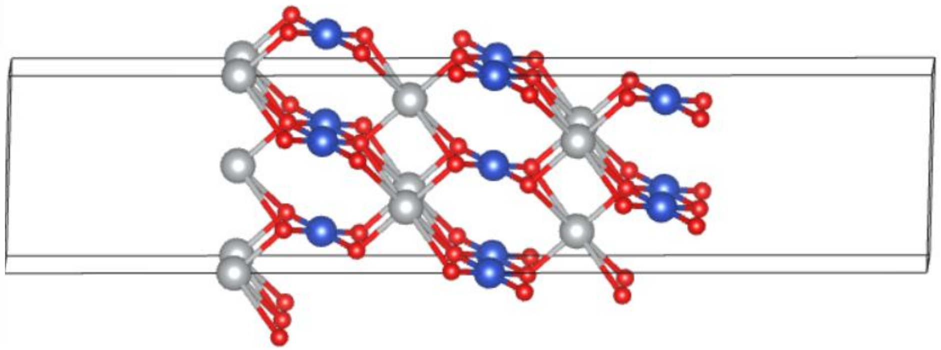


WF > 8 eV
56 stable surfaces

Ag_2F, (001)-F

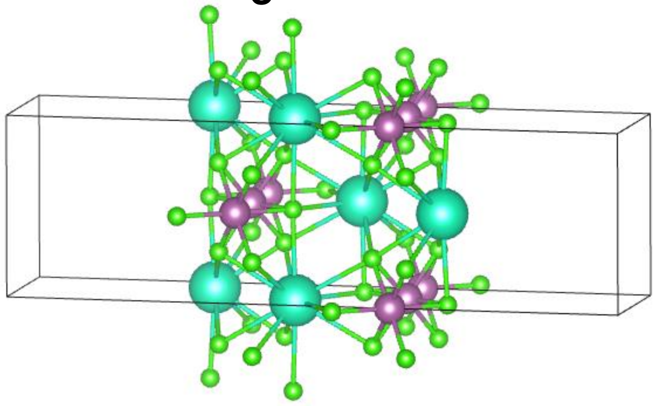


$CuAgO_2$, (001)-O

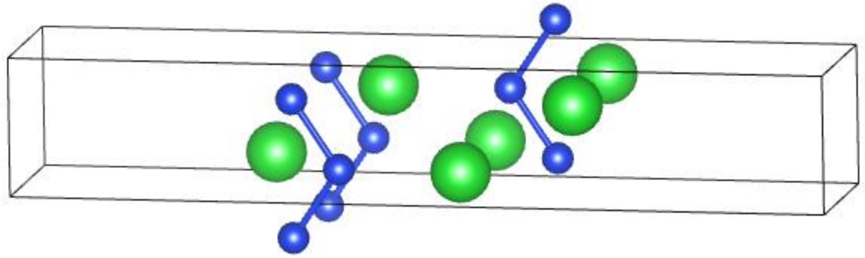


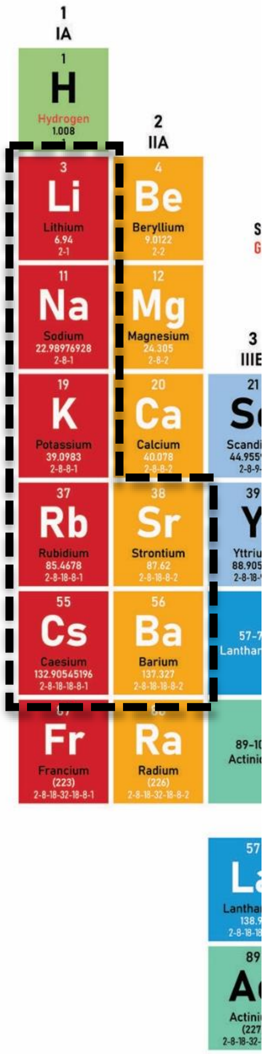
WF < 1.7 eV
34 stable surfaces

$CsScCl_3$, (100)-Sc



BaX [X=Si, Sn], (110)-Ba-X



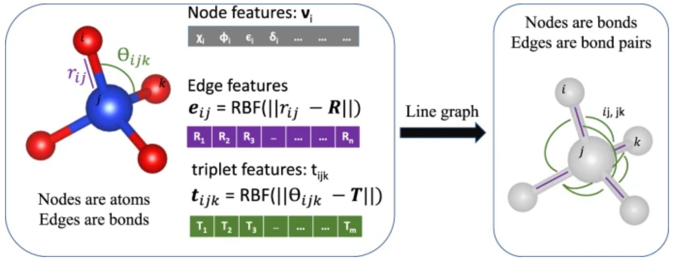


P. Schindler, et al. *Adv. Func. Mat.* 34, 19 (2024)

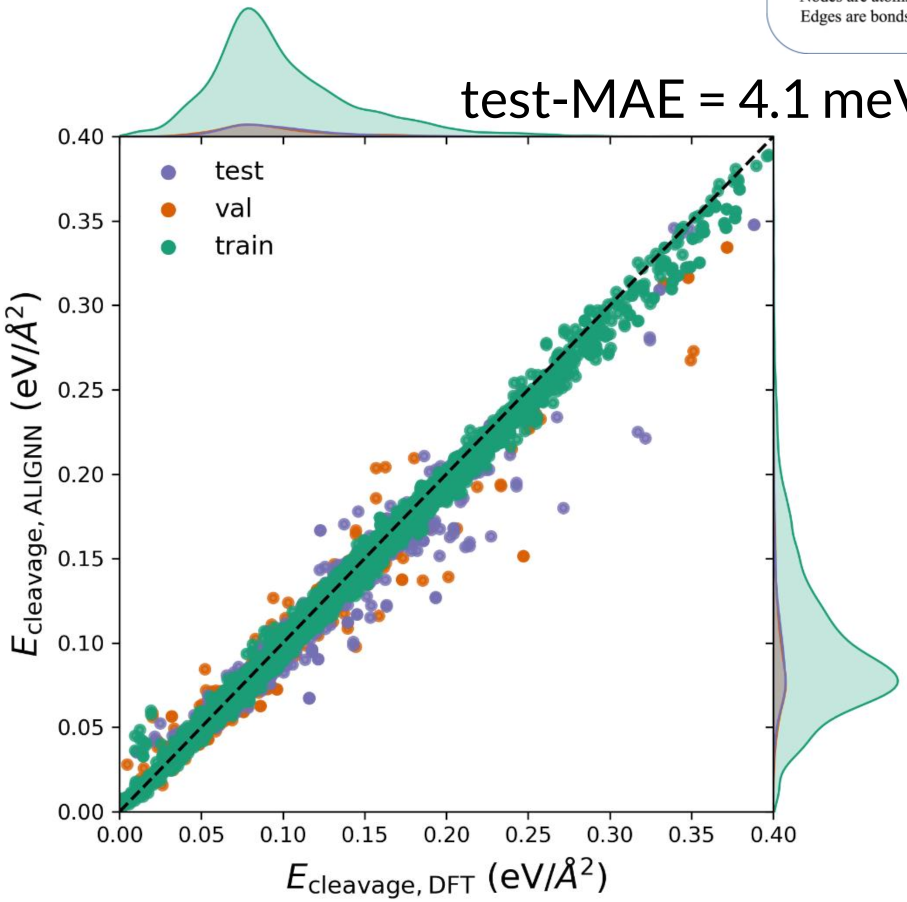
Graph-Convolutional Neural Networks to Predict Surface Properties

ALIGNN Framework:

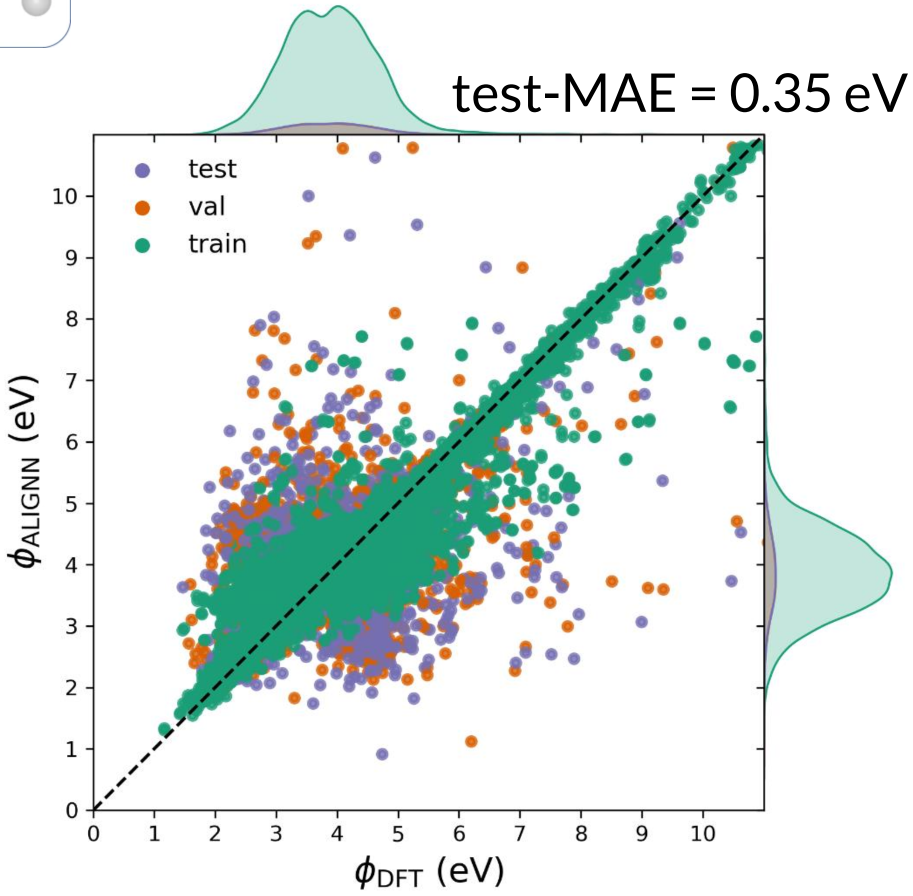
K. Choudhary & B. DeCost, npj Comp. Mat. 7 (2021)



test-MAE = 4.1 meV/A^2



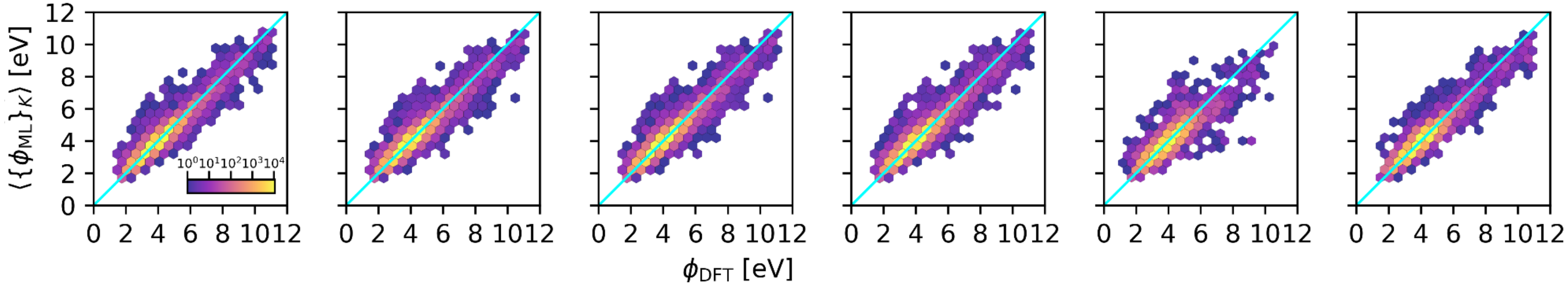
test-MAE = 0.35 eV

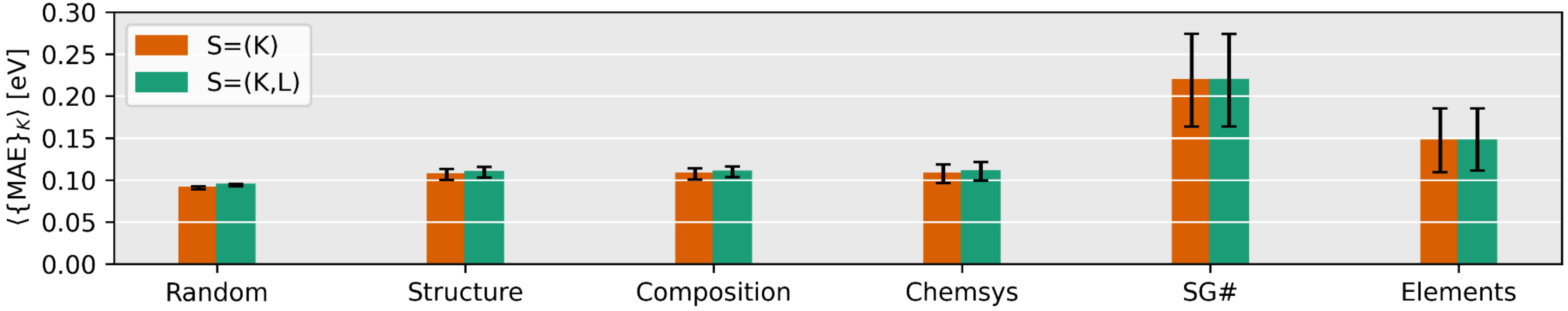


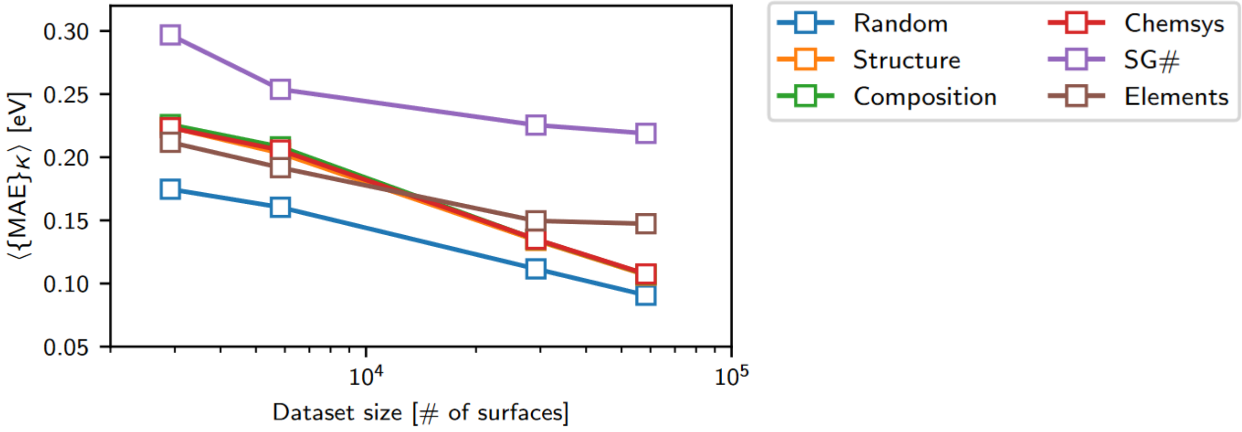
Currently testing: E(3)-equivariant Models

(in preparation)

MatFold: Cross-Validation Protocols to Probe OOD Generalization in MatSci

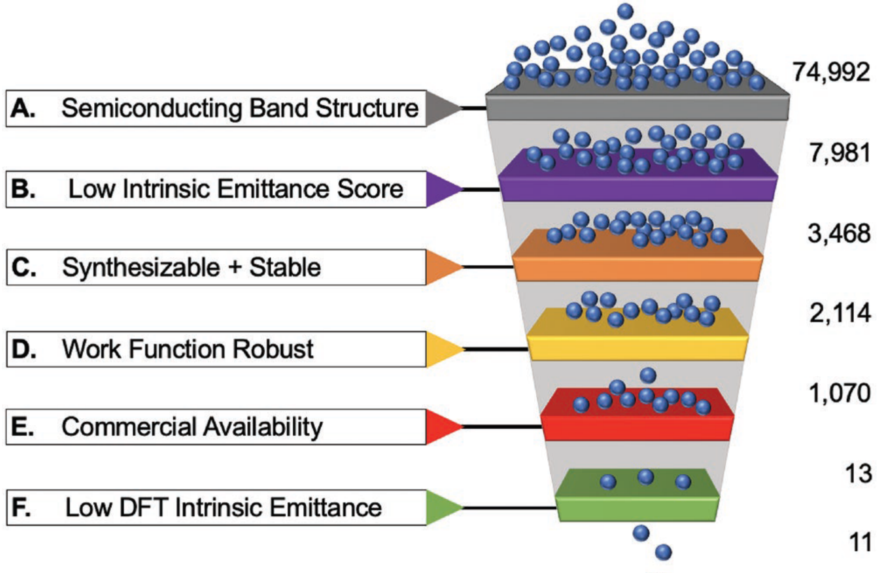




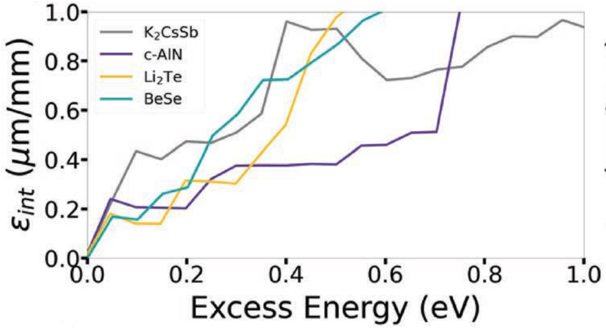


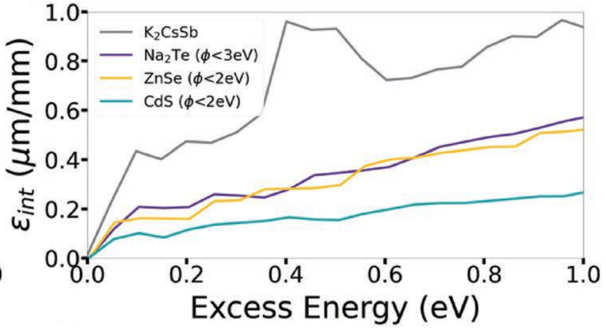
M. D. Witman and **P. Schindler**, *ChemRxiv (2024)*
[Preprint will be available in early August.
MatFold Python package will be available
open-source after journal publication]

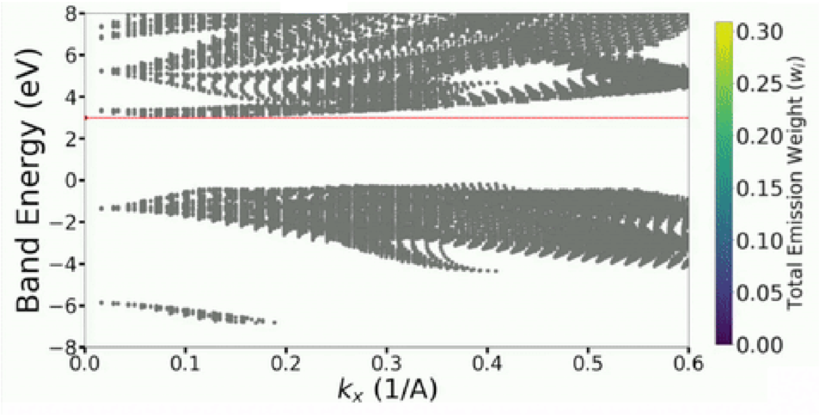
ML Driven Screening: Novel Ultra-bright and Air-Stable Photocathodes

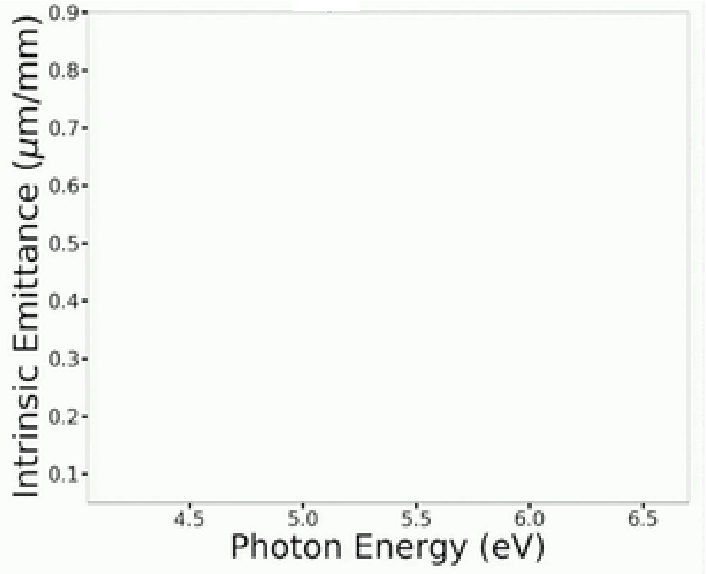


11 materials with intrinsic emittance < 0.3 µm/mm







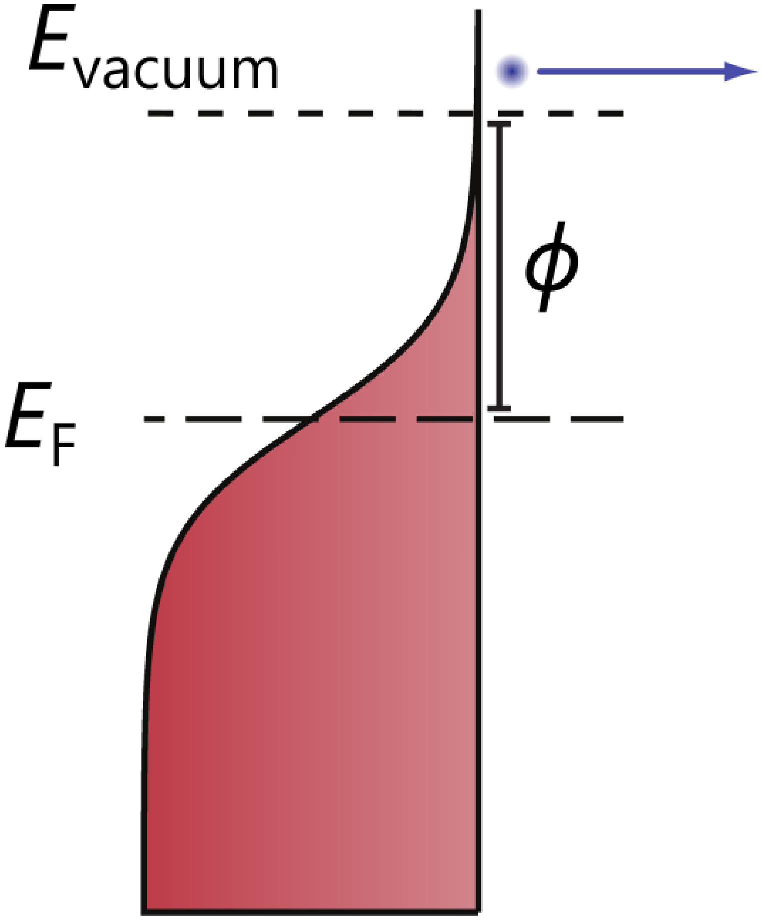


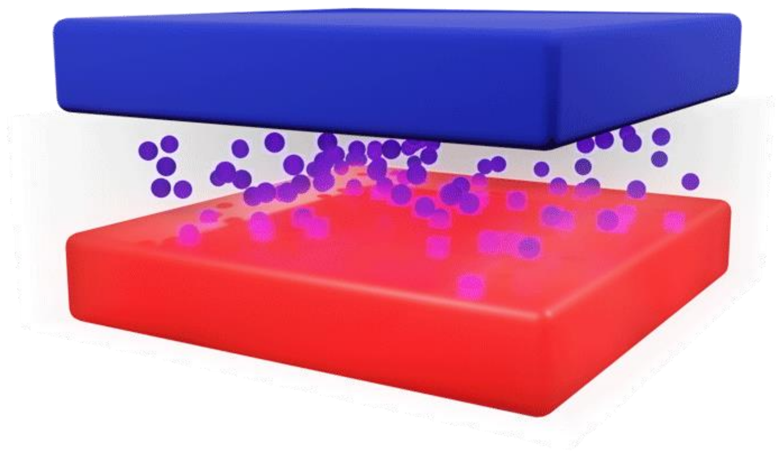
+ 3 air stable low intrinsic emittance materials M_2O (M = Na, K, Rb)

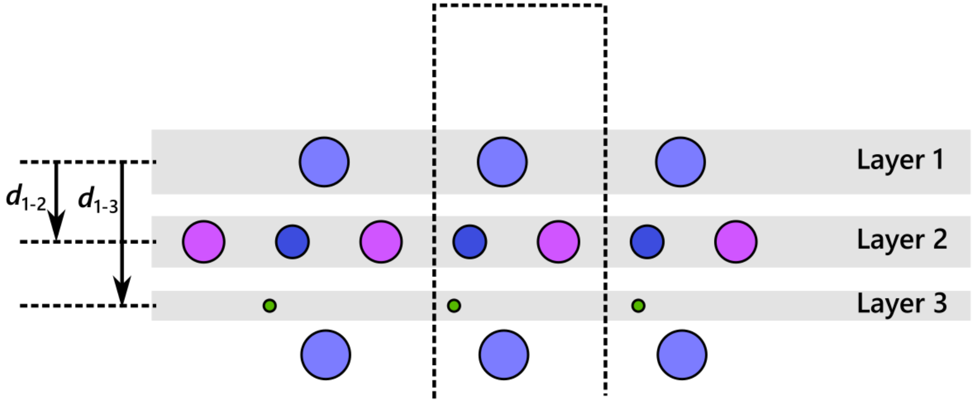
E. R. Antoniuk, Y. Yue, Y. Zhou, **P. Schindler**, W.A. Schroeder, B. Dunham, P. Pianetta, T. Vecchione, and E.J. Reed. *Physical Review B*, 101 (2020).

E. R. Antoniuk, **P. Schindler**, W. A. Schroeder, B. Dunham, P. Pianetta, T. Vecchione, and E. J. Reed. *Advanced Materials*, 33, 44 (2021).

Ultra-low Work Function







Thank you for listening!

Questions?

For further information and references:

www.d2r2group.com

Open Access: Full database and ML model (click on "*Resources*")